Non-linear Model Predictive Control of the Hashimoto Simulated Moving Bed Process

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Summary. 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 the solid phases 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 catalyst and adsorbent have been proposed and demonstrated successfully. However, a full integration often is not efficient because in the columns in the separating zones, the catalyst is not used or even counterproductive. By placing reactors between the separation columns at specific positions around the feed port, a more efficient process, the Hashimoto SMB process, is established. In this contribution, a non-linear predictive control concept for the Hashimoto SMB process is presented. The controller computes optimal control variables (flow rates and the switching time) to optimize an economic objective over a moving horizon. The purity requirements of the product streams are implemented as constraints and not as controlled variables. The optimization-based controller is combined with a scheme to estimate selected model parameters in order to reduce the influence of the inevitable model errors. Simulative results are presented for the example of the racemization of Tröger's base.

Keywords: Simulated moving bed chromatography (SMB), Hashimoto process, online optimization, parameter estimation.

1 Introduction

Chromatographic separation processes are based on different affinities of the involved components to a solid adsorbent packed in a chromatographic column. Most industrial applications are performed discontinuously involving one single chromatographic column which is charged with pulses of feed solutions. The feed injections are carried through the column by pure eluent. The more retained component travels through the column slower, and hence leaves the column after the less adsorptive component. The separated peaks can be withdrawn with the desired purity at the end of the column. However, batch operation leads to low productivity and high solvent consumption.

R. Findeisen et al. (Eds.): Assessment and Future Directions, LNCIS 358, pp. 473–483, 2007.
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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 the consumption of the solvent. The SMB process consists of several chromatographic columns which are interconnected in series to constitute a closed loop. A counter-current movement of the liquid phase and the solid phase is simulated by periodical and simultaneous switching of the inlet and outlet ports by one column in the direction of the liquid flow.

The Hashimoto SMB [1] is an extension of the Simulated Moving Bed process which integrates reaction into chromatographic separation and is therefore suitable to overcome the limitations of equilibrium reactions. The reactors are fixed in those separation zones of the Hashimoto SMB process where the forward reaction is favourable thus increasing the conversion of the feed.

Since SMB processes are characterized by mixed discrete and continuous dynamics, spatially distributed state variables with steep slopes, and slow and strongly nonlinear responses of the concentrations profiles to changes of the operating parameters, they are difficult to control. An overview of recent achievements in the optimization and control of chromatographic separations can be found in [3]. In [7] and [8], 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 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 error feedback of the predicted and the measured purities. In addition, the model parameters are regularly updated. In [4], the control concept was extended to the more complex processes Varicol and Powerfeed that offer a larger number of degrees of freedom that can be used for the optimization of the process economics while satisfying the required product purities. A slightly different approach to the control of SMB processes was reported by [5] and [6]. 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 this work, the switching period is considered as fixed, while in the previously mentioned work it is a parameter in the optimization. In [7] and [8], the prediction is based on the assumption that the columns are uniform (i.e. they all show the same behavior) and that the modelling errors are small. However, the properties of each individual column differ since they have different effective lengths, different packings with adsorbent and catalyst (for the case of reactive chromatography) and the column temperatures can exhibit some variation. In [10], a combined parameter and state estimation scheme for a nonlinear SMB process with individual column properties based on measurements of the concentrations in both product streams and one internal measurement is proposed.

In this paper, a control concept for the Hashimoto SMB process is presented. A non-linear predictive controller for the Hashimoto SMB process is established that computes optimal control variables (flow rates and the switching time) to optimize an economic objective over a moving horizon while the purity requirements

of the product streams are considered as constraints. In the optimization, a rigorous model of the general rate 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 by a parameter estimation scheme.

The remainder of this paper is structured as follows: in the next section, the model of the Hashimoto SMB process is introduced. Section 3 is devoted to the predictive control concept based upon an online optimization and parameter estimation scheme. Simulation results are presented in section 4. Finally, a summary and an outlook for future research are given.

2 Process Model

In this paper, the racemization of Tröger's base (TB) is considered. 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 equilibrium. Since the product TB- has a higher affinity to the chosen adsorbent, it is withdrawn at the extract port of the Hashimoto process. The TB+ part is withdrawn with the raffinate stream in order to improve the purification of the solvent in zone IV before it is passed on to zone I. Theoretically, this raffinate flow can be converted to the equilibrium by an additional external reactor and added to the feed stream. Alternatively, no raffinate flow can be taken out, thus the whole feed is converted to TB- making the additional unit to convert TB+ redundant. However, in this case a drastic increase of eluent would be required and the process would be less efficient. For the application in this paper, the first case is examined with the Hashimoto configuration depicted in Figure 1. The reactors are placed in zone III where a high concentration of TB+ is present. When the ports of the process are shifted by one column after the period τ has passed, the physical separation columns are switched by one column in the opposite direction to the liquid flow moving through the separation zones. The reactors, however, remain at their positions relative to the ports. The practical realization of the column switching via a process control system is sophisticated and indicated by Figure 2 which shows the

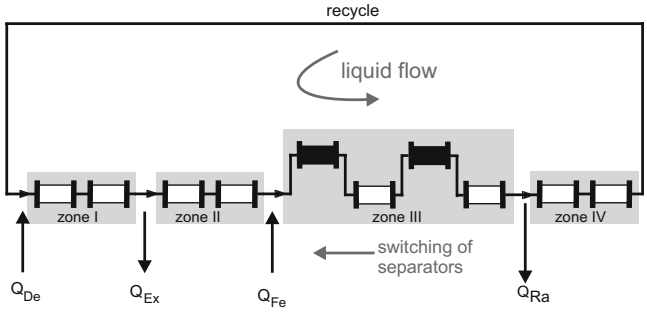

Fig. 1. Hashimoto configuration (reactors: black, separators: white)

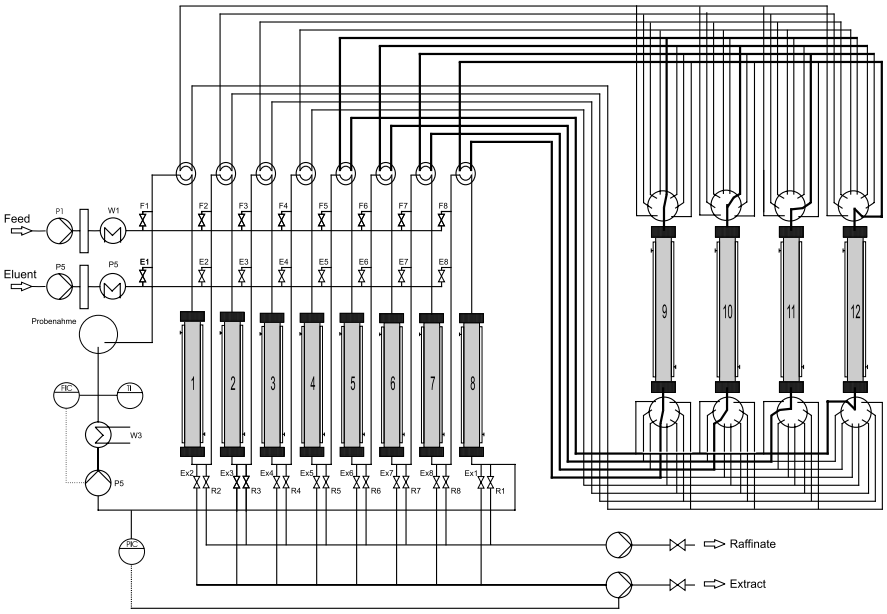

Fig. 2. Flowchart of the Hashimoto process

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.

Accurate dynamic models of multi-column continuous chromatographic processes consist of dynamic models of the single chromatographic columns and of the tubular reactors and the node balances which describe the connections of the columns and of the switching of the ports. 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. 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 given 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, 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 $c_{p,i}$. Furthermore, it is assumed that u and c_i are uniformly distributed over the radius. 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

$$\frac{\partial c_{b,i}}{\partial t} + \frac{(1-\epsilon_b)3k_{l,i}}{\epsilon_b R_p}\,(c_{b,i} - c_{p,i}|_{r=R_p}) = D_{ax}\,\frac{\partial^2 c_{b,i}}{\partial z^2} - u\frac{\partial c_{b,i}}{\partial z} \tag{1}$$

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

Reactor

$$\frac{\partial c_{b,i}}{\partial t} + r^{\text{liq}}_{\text{kin},i} = D_{ax}\,\frac{\partial^2 c_{b,i}}{\partial z^2} - u\frac{\partial c_{b,i}}{\partial z} \tag{3}$$

with appropriate initial and boundary conditions

$$c_{b,i}|_{t=0} = c_{b,i}(t=0,z), \quad c_{p,i}|_{t=0} = c_{p,i}(t=0,z,r), \tag{4}$$

$$\left.\frac{\partial c_{b,i}}{\partial z}\right|_{z=0} = \frac{u}{D_{ax}}(c_{b,i} - c_i^{in}), \quad \left.\frac{\partial c_{b,i}}{\partial z}\right|_{z=L} = 0, \tag{5}$$

$$\left.\frac{\partial c_{p,i}}{\partial r}\right|_{r=0} = 0, \quad \left.\frac{\partial c_{p,i}}{\partial r}\right|_{r=R_p} = \frac{k_{l,i}}{\epsilon_p D_{p,i}}(c_{b,i} - c_{p,i}|_{r=R_p})\,. \tag{6}$$

The adsorption equilibrium and the reaction kinetics have been determined experimentally in [11]. The adsorptive behaviour can be modelled best by an asymmetric multi-component Langmuir isotherm

$$q_i = \frac{H_i c_i}{1 + \sum\limits_j b_{i,j} c_j} \quad i = A, B, \tag{7}$$

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

$$r^{liq}_{kin,i} = \nu_i k_m (c_{b,i} - c_{b,j}) \quad i,j = A,B \qquad i \neq j. \tag{8}$$

From mass and concentration balances, the relations at the inlet and the outlet nodes result as

$$\text{Desorbent node} \quad Q_{IV} + Q_{De} = Q_I \tag{9}$$

$$c^{out}_{i,IV} Q_{IV} = c^{in}_{i,I} Q_I \qquad i = A,B \tag{10}$$

$$\text{Extract node} \quad Q_I - Q_{Ex} = Q_{II} \tag{11}$$

$$\text{Feed node} \quad Q_{II} + Q_{Fe} = Q_{III} \tag{12}$$

$$c^{out}_{i,II} Q_{II} + c_{i,Fe} Q_{Fe} = c^{in}_{i,III} Q_{III} \qquad i = A,B \tag{13}$$

$$\text{Raffinate node} \quad Q_{III} - Q_{Ra} = Q_{IV}, \tag{14}$$

where $Q_{I,II,III,IV}$ denote the internal flow rates through the corresponding zones I, II, III, IV, Q_{De}, Q_{Ex}, Q_{Fe}, and Q_{Ra} are the external flow rates of the inlet/outlet ports, respectively, and $c_{i,j}^{out}$ and $c_{i,j}^{in}$ denote the concentrations of the component i in the stream leaving or entering the respective zone j.

As eluent, an equimolar mixture of acetic acid and 2-Propanol is utilized. Acetic acid is the catalyst with a high activity at a temperature of 80 $°C$, but negligible activity at room temperature. Thus, the reactors are operated at 80 $°C$ while the separators are operated at room temperature. The separators are packed with the adsorbent Chiaralcel. An efficient numerical solution approach is used as proposed in [9] where a finite element discretization of the bulk phase is combined with orthogonal collocation of the solid phase.

3 Predictive Control

3.1 Online Optimization

The basic idea of the control algorithm is to perform an optimization of the operational degrees of freedom at future switching periods based upon a rigorous model of the plant with respect to an economic cost function (rather than e. g. a cost function involving a tracking error) in which the specifications of the SMB process (purity requirements, limitations of the pumps) as well as the process dynamics are handled as constraints. 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 with the control and prediction horizon shifted forward by one time interval, using new measurement data and eventually new estimated model parameters. In the application of optimizing control to SMB processes, the sampling time is chosen equal to the length of a cycle (length of a switching period times the number of chromatographic columns) and hence 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. We here consider a four-zone Hashimoto SMB process with raffinate flow that is described by the nonlinear discrete dynamics (16), (17). The objective of the optimizing controller is to minimize the eluent consumption Q_{De} for a constant feed flow and a given purity requirement of 99% in the presence of a plant/model mismatch. The inevitable mismatch between the model and the behavior of the real plant is taken into account by feedback of the difference of the predicted and the measured product purities. A regularization term is added to the objective function (15) to obtain smooth trajectories of the input variables. The controller has to respect the purity requirement for the extract flow (18) which is averaged over the prediction horizon, the dynamics of the Hashimoto SMB model (16), (17) and the maximal flow rate in zone I (20) due to limited pump capacities

(21). In order to guarantee that at least 70% of the mass of the components fed to the plant averaged over the prediction horizon leaves the plant in the extract product stream, an additional productivity requirement (19) is added. The deviation between the prediction of the model and the plant behavior is considered by the error feedback term (23). The resulting mathematical formulation of the optimization problem is:

$$\min_{\beta_I,\beta_{II},\beta_{III},\beta_{IV}} \quad \sum_{i=1}^{H_P} Q_{De,i} + \Delta\beta R \Delta\beta \tag{15}$$

$$s.t. \quad x^i_{smb} = x^i_{smb,0} + \int_{t=0}^{\tau} f_{smb}(x_{smb}(t), u(t), p)dt \tag{16}$$

$$x^{i+1}_{smb,0} = M x^i_{smb,\tau} \tag{17}$$

$$\frac{\sum_{i=1}^{H_P} Pur^*_{Ex,i}}{H_P} \geq \left(Pur^*_{Ex,min} - \Delta Pur_{Ex}\right) \tag{18}$$

$$\frac{\sum_{i=1}^{H_P} m_{Ex,i}}{H_P} \geq 0.7 m_{Fe} - \Delta m_{Ex} \tag{19}$$

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

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

where M is the shifting matrix, τ the period length. The extract purity, the purity error, the mass output, and the mass error are evaluated according to:

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

$$\Delta Pur_{Ex} = Pur^*_{Ex,plant,i-1} - Pur^*_{Ex,model,i-1} \tag{23}$$

$$m_i = \frac{\int_0^{\tau} (c_{i,A} + c_{i,B}) Q_i dt}{\tau} \tag{24}$$

$$\Delta m_{Ex} = m_{Ex,plant,i-1} - m_{Ex,model,i-1}. \tag{25}$$

Since the plant is operated close to 100% extract purity, the purities are scaled $(*)$ according to

$$purity^* = \frac{1}{1 - purity}, \tag{26}$$

that provides a large slope in the scaled purity for very high purities. The numerical tractability is improved by translating the degrees of freedom (period

length τ, desorbent flow Q_De, extract flow Q_{Ex}, and recycle flow Q_{Re}) into the so-called beta factors [2] that relate the liquid flow rates Q_i in each separation zone to the simulated solid flow rate Q_s.

$$Q_s = \frac{(1-\epsilon)V_{col}}{\tau} \qquad \frac{1}{\beta_{III}} = \frac{1}{H_A}\left(\frac{Q_{III}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{27}$$

$$\beta_I = \frac{1}{H_A}\left(\frac{Q_I}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \qquad \frac{1}{\beta_{IV}} = \frac{1}{H_B}\left(\frac{Q_{IV}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right). \tag{28}$$

$$\beta_{II} = \frac{1}{H_B}\left(\frac{Q_{II}}{Q_s} - \frac{1-\epsilon}{\epsilon}\right) \tag{29}$$

A feasible path SQP solver is applied to solve the optimization problem. The solver generates a feasible point before it minimizes the objective function. Since the SQP algorithm is a gradient based method, an additional constraint

$$\frac{\sum_{i=1}^{8} Pur_{Ex,i}}{8} \leq 99.9\% \tag{30}$$

is added that enforces the purity to be below 99.9% averaged over a cycle and prevents the purity from reaching 100 % at which the gradient information for the constraint (18) is lost.

3.2 Parameter Estimation

The parameter estimation scheme is based on a measurement device that is fixed behind the physical separation column positioned in front of the recycle line. The recycle measurements are collected over one cycle and simulated by the model to estimate the reaction rate constant k_m and the Henry coefficients H_A and H_B via a least-squares minimization according to:

$$\min_p \sum_{i=A}^{B} \left(\int_0^N (c_{i,meas}(t) - c_{i,Re})^2 dt \right), \tag{31}$$

where N is the number of measurement points.

4 Results

For the simulative run presented here, a column distribution as shown in Figure 1 is assumed (1400 states). The sampling time is set to one cycle (8 periods). The prediction horizon H_P and the control horizon H_C have a length of 6 intervals and 1 interval, respectively. The regularization terms R_i are set to 0.3 for each control variable. Both, controller and estimator, are started at the 72^{nd} period. In the control scenario, an exponential decrease of the catalyst activity is assumed that occurs in the case of a malfunction of the reactor heating. A

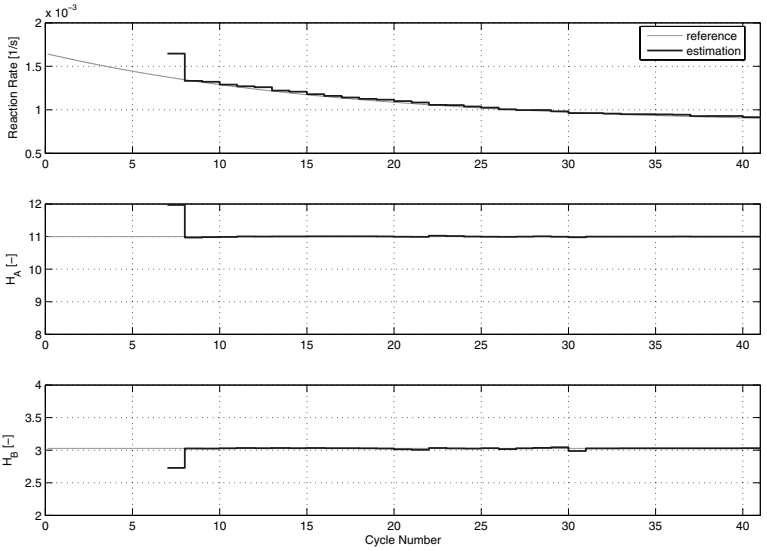

Fig. 3. Estimation of the reaction rate constant and Henry coefficients

separator:
separator length $L_s = 10cm$ particle diameter $d_p = 10\mu m$
separator diameter $D_s = 1cm$ axial diffusion coefficient D_{ax} acc. to [12]

adsorption coefficients	$H_A = 10.997$	**reactor:**	
	$b_{1,1} = 0.132\frac{l}{g}$	reactor length	$L_r = 100cm$
	$b_{1,2} = 0.543\frac{l}{g}$	reactor diameter	$D_r = 0.53cm$
	$H_B = 3.028$	reaction rate coef.	$k_m = 0.001645\frac{1}{s}$
	$b_{2,1} = 3.413\frac{l}{g}$	stoichiometry	$\nu = [-1; +1]$
	$b_{2,2} = 0.0\frac{l}{g}$	**eluent**:	
film transfer resistance	$k_{l,A} = 0.000302\frac{cm}{s}$	density	$\rho = 0.81867\frac{g}{ml}$
	$k_{l,B} = 0.000302\frac{cm}{s}$	viscosity	$\eta = 0.0271\frac{g}{cms}$
column void fraction	$\epsilon_b = 0.387$	**feed:**	$Q_{Fe} = 0.61\frac{ml}{min}$
particle void fraction	$\epsilon_p = 0.4$		$c_{A,Fe} = 2.5\frac{g}{l}$
overall void fraction	$\epsilon = 0.632$		$c_{B,Fe} = 2.5\frac{g}{l}$
particle diffusion coefficient	$D_p = 0.001\frac{cm^2}{s}$		

further plant/model mismatch is introduced by disturbing the initial Henry coefficients H_A and H_B of the model by +10% and -10%. Figure 3 shows that the parameter estimation scheme estimates the parameters of the plant well. The performance of the controller is illustrated by Figure 4. The controller manages to keep the purity and the productivity above their lower limits, while it improves the economic operation of the plant by reducing the solvent consumption. The

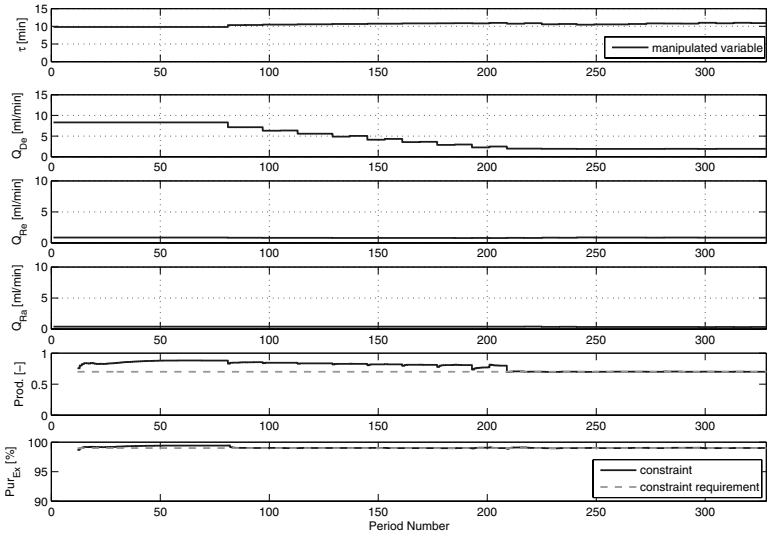

Fig. 4. Simulation of the predictive controller; manipulated variables and constraints

process converges to a stationary operating point. The optimizer converges to the optimum within one sampling time, and hence, can be applied in real-time.

5 Conclusion

An online model-based optimization and parameter estimation scheme for the Hashimoto Simulated Moving Bed process has been presented. The approach has the advantage that the process is automatically operated at its economic optimum while the purity and productivity requirements and plant limitations are fulfilled. In future research, the application to a pilot-plant is planned.

Acknowledgement

We would like to thank Larry Biegler for a fruitful discussion about how to avoid zero gradient information for a SQP solver. This investigation was supported by the Deutsche Forschungsgemeinschaft (DFG) under grant DFG En 152/34. This support is gratefully acknowledged.

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