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OPTIMAL CONTROL of the SIMULATED MOVING BED (SMB) CHROMATOGRAPHIC SEPARATION PROCESS

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Abstract: In this contribution, the open-loop optimal control (dynamic optimization) of a Simulated Moving Bed (SMB) chromatographic separation process is considered. The objective is to compute the optimal feed concentration and/or feed flow rate, over each switching period, with maximum flexibility. This problem is solved numerically using the combination of the control vector parameterization scheme with suitable state-of-the-art global nonlinear programming problem solvers. The advantages of the proposed approach are illustrated through the solution of two case studies, achieving significant improvements in the process productivity when compared to traditional feeding profiles.

Keywords: Simulated Moving Bed (SMB) chromatography, dynamic optimization, optimal control, feed concentration modulation, Control Vector Parameterization (CVP).

1. INTRODUCTION

The separation of the components of mixtures is an operation of key importance in many industrial areas (pharmaceutics, food, fine chemicals, etc.). In comparison to traditional batch processes, the continuous counter-current chromatographic techniques can achieve higher productivity, purity and product recovery, decreasing the desorbent consumption and keeping a constant product quality. However the movement of the solid phase is hardly realizable and, in practice, the Simulated Moving Bed (SMB) process is usually applied (Ruthven and Ching, 1998; Engell and Toumi, 2005).

During recent years a number of methods have been proposed to select the operation conditions in an attempt to maximize products purity or process productivity.

The "Triangle Theory" (Mazzotti et al., 1997) combines the material balances at the nodes of the SMB unit with the results of the Equilibrium Theory for Langmuir systems, to provide the constant flow rate ratios yielding complete separation. Unfortunately these operating conditions are not robust to small perturbations so that adequate control schemes should be used (as reviewed by Engell and Toumi, 2005).

It was soon realised that the modulation of certain operating conditions, instead of keeping them constant, could largely improve both productivity and products purity.

In this regard, the VARICOL (VARiable length COLumn, Ludemann-Hombourger and Nicoud, 2000) process makes use of a periodic but asynchronous shift of the ports to achieve a better allocation of the adsorbent and hence a reduced desorbent consumption. In this process, the number of columns per zone varies during a switching time, returning to the starting initial value at the end of the period. In the early 90s, Kearny and Hieb patented the modulation of several of the process flow rates, which resulted in a significant increase of products quality. A further development of this approach, the so-called PowerFeed process, was proposed by Zhang et al. (2003) who computed the optimal time-varying flow rates to maximize both product purities, revealing that the objective function is more sensitive to feed flow rate changes. As an alternative, Schramm et al. (2003a) proposed the so-called ModiCon that, based on the predictions of the nonlinear wave propagation theory, suggests the modulation of the feed concentration injected to the system during the switching time. It is particularly remarkable that concentration modulation outperforms feed flow rate modulation (Schramm et al., 2003b).

In this contribution, we present the statement and solution of two open-loop optimal control problems (OCP), using the modulation of the feed concentration and the feed flow rate as control variables in order to either maximize the process productivity, subject to specific raffinate purity requirements, and the system dynamics.

Due to the distributed nonlinear nature and the presence of coupled phenomena in the rigorous SMB model, the resolution of the problems becomes a challenging task. Thus, we apply state of the art solvers within a control vector parameterization (CVP) scheme, in order to both efficiently simulate the complex cyclic dynamics and surmount optimisation convergence problems. To illustrate the advantages of the optimal operating policies computed with these numerical techniques, the results are compared to those obtained using the classical ModiCon approach.

2. SIMULATED MOVING BED (SMB) PROCESS

2.1 SMB process description.

The Simulated Moving Bed (SMB) process consists of a number of chromatographic columns connected in series and arranged in four zones as represented in Figure 1.

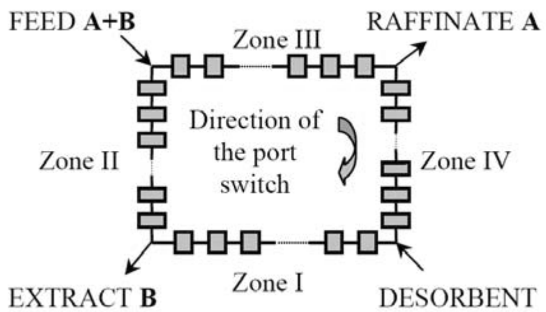

Fig 1. Simulated Moving Bed process.

The continuous separation of the components of the mixture takes place via the apparent counter-current flow between the solid and the liquid phases. The initial mixture (Feed) is introduced between zones II and III, the raffinate, essentially composed by the less adsorbed component of the mixture, is obtained between zones III and IV, and the extract, containing mainly the more adsorbed component, is extracted between zones I and II. The separation of the components occurs mainly in zones II and III, while the adsorbent regeneration and the desorbent recycling are the principal goals of sections I and IV, respectively.

2.2. SMB process model.

The system is considered as a set of static chromatographic columns with periodic switching times. The concentration of the component i in the column j ($Cs_{i,j}$) is given by the mass balance of the solid phase in this column:

$$\frac{\partial Cs_{i,j}}{\partial t} = k_{FS,i}(Cs^*_{i,j} - Cs_{i,j}) \;\; i = 1,2; \; j = 1,...,NC \qquad (1)$$

where t is the time, k_{FS} the mass transfer coefficient. $Cs^*_{i,j}$ is the equilibrium concentration defined, for instance, by a Langmuir competitive adsorption isotherm:

$$Cs^*_{i,j} = \frac{a_i \cdot C_{i,j}}{1 + \sum_{k=1}^{2} b_k \cdot C_{k,j}} \qquad (2)$$

In the liquid phase, the concentration of i in the column j ($C_{i,j}$) is described by the mass balance:

$$\frac{\partial C_{i,j}}{\partial t} = D_{L\,i,j}\frac{\partial^2 C_{i,j}}{\partial z^2} - u_j \frac{\partial C_{i,j}}{\partial z} - \frac{1-\varepsilon}{\varepsilon} \cdot \frac{\partial Cs_{i,j}}{\partial t} \qquad (3)$$

The right-hand-side terms in (3) represent the dispersive liquid transport, the convective transport and the mass transfer between the liquid and solid phases, $D_{L\,i,j}$ and u_j being the axial dispersion coefficient and the fluid speed, respectively.

Due to the cyclic repositioning, the input and output ports are moved one column in the liquid flow direction every switching interval *Δt*. Thus, the concentration profiles in the column j at the beginning of a switching period p are equal to those obtained in the column $j+1$ at the end of the previous period ($p-1$):

$$C_{i,j}(t_p = 0, z_j) = C_{i,j+1}(t_{p-1} = \Delta t, z_{j+1}) \qquad (4)$$

The boundary conditions for the mass balance in the liquid phase are of Dirichlet type at the beginning of each column.

$$C_{i,j}(t, z = 0) = C_{i0,j} \qquad (5)$$

A simple advection equation is used at the end of each column to express zero-dispersion conditions:

$$\frac{\partial C_{i,j}}{\partial t}(z = L) = -u_j \frac{\partial C_{i,j}}{\partial z}(z = L) \qquad (6)$$

To complete the specification of the boundary conditions, it is necessary to express the mass balance of each component at the transition between two consecutive zones.

For more details on the model see Haag et al. (2001).

2.3. Numerical methods for simulation.

In this contribution the numerical method of lines (NUMOL, Schiesser, 1991), as implemented in the MATMOL toolbox (Vande Wouwer et al., 2004), is used for solving the PDEs of the model. The spatial derivatives have been approximated by fourth-order finite difference formula, using biased-upwind finite difference schemes for the convective terms and centred finite differences for diffusive terms (Haag et al., 2001).

After applying NUMOL, the evolution of the field is described by a large scale, sparse and possibly stiff set of ordinary differential equations whose solution requires the use of a sparse implicit initial value problem solver (IVP) to enhance both the stability and the efficiency of the solution process.

3. DYNAMIC OPTIMIZATION OF SMB

3.1 Mathematical statement

The open loop optimal control problem consists in the computation of a set of time-dependent decision variables in order to minimize (or maximize) a pre-defined objective functional over a certain finite time horizon, while verifying the existing constraints.

This work presents the formulation and solution of 2 problems, OCP1 and OCP2, formulated as follows:

Find the feed concentration $C_{FEED}(t)$ (and the feed flow rate, Q_{FEED}, for OCP2) along $t \in [t_0, t_f]$ to maximize the process productivity PR defined as the amount of feed separated (equal to the amount of product) per amount of necessary adsorbent:

$$\max_{C_{FEED}} PR; \quad PR = \frac{\dot{m}_{PRODUCT}}{m_{ADSORBENT}} = \frac{Q_{FEED} \cdot \overline{C}_{FEED}}{NC \cdot (1-\varepsilon) \cdot S \cdot L} \qquad (8)$$

subject to:

- the system dynamics: Eqns.(1)-(6)

- lower and upper bounds on the control variables:

$$0 \le C_{FEED} \le 1.0\ \%vol \qquad (9)$$

$$0 \le Q_{FEED} \le 25\ ml/min\ \ (only\ for\ OCP2) \qquad (10)$$

The upper bound on the feed concentration has been chosen taking into account the upper limit of solubility of the mixture, while the maximum feed flow rate is limited by the maximum allowable pressure drop in the plant.

- and a constraint related to the minimum average raffinate purity at final time:

$$Pur_{Raf} = \overline{C}_{j,Raf} \Big/ \sum_{i=1}^{2} \overline{C}_{i,Raf} \ge Pur_{Raf,min} = 85\% \qquad (11)$$

j being the less adsorbed component.

3.2 Numerical methods for dynamic optimization.

The state-of-the-art direct methods transform the original infinite dimensional optimization problem into a non-linear programming problem (NLP). The complete parameterization (CP) or the multiple shooting approaches parameterise both controls and states, becoming usually rather expensive for distributed systems. In contrast, the control vector parameterization (CVP, Vassiliadis et al., 1994) only discretises the controls, resulting the most convenient for large-scale systems with complex dynamics, particularly those related to distributed parameter systems (Balsa-Canto et al., 2004) such as the SMB case. CVP proceeds dividing the controls in a number of elements (ρ) and approximating each element via a low order polynomial. The polynomial parameters become the decision variables in the NLP, whose solution can be approached with a standard solver.

In this regard, Successive Quadratic Programming (SQP) methods are the most popular. Nevertheless they can lead to suboptimal (local) solutions in presence of non convexities, i.e. multiple optima (Banga and Seider, 1996), as it will be illustrated later for the case studies considered, especially when started far away from the global optimum. To surmount these difficulties several global optimization techniques have been developed.

The proposed approaches may be classified in two main groups: deterministic and stochastic methods. The first methods, take advantage of the problem structure and ensure global convergence, but only for problems that comply with a set of requirements (such as smooth and twice continuous differentiable functions), which are not ensured in the present cases. Alternatively, stochastic and hybrid stochastic-local methods, although they can not guarantee global optimality, offer the option of achieving solutions close to the global ones (very often the best known solutions) in relatively short computation times. This makes them very attractive for dynamic optimization purposes (Banga and Seider, 1996).

4. RESULTS

We consider the particular case where the two components of a mixture (cyclopentanone and cyclohexanone) are separated, cyclopentanone being the more adsorbed component in the liquid phase. The fixed operation conditions are summarized in Table 1 and the model parameters estimated based on experimental data collected on a pilot plant available at the Max Planck Institute of Magdeburg (Germany) can be found in the work by Grosfils et al., 2004.

Table 1 Fixed operation conditions of the SMB process

Δt, t_f	Switching & final time	180s, 11520s
Q_{II}, Q_{IV}	Flow rates in zones II,IV	39.39,43.40ml/min
$Q_{FEED\ REF}$	Feed flow rate (ref.value)	17.74 ml/min
Q_{DESORB}	Desorbent flow rate	20.00 ml/min

Taking into account the complexity of the model of the SMB process and the presence of constraints in the optimal control problems formulated above, the selection of a robust and efficient optimization method is of key importance. In this concern, we used the recently developed Nonlinear Dynamic Optimization Toolbox (NDOT, García et al., 2005) implemented in Matlab, which is based on the use of the CVP scheme and offers a large variety of state of the art IVP and NLP solvers. So as to increase computational efficiency, the IVP solution is implemented in compiled Fortran 77 and is called from Matlab through suitable gateways (mex-files).

Regarding the SMB process simulation, the NUMOL method as described above was considered. A grid of 20 nodes over each column was finally selected as it offers a good compromise accuracy/computational cost. Since there are two columns per zone, this results in a set of 640 ODEs solved here using LSODES (Hindmarsh, 1983) a BDF method which exploits the sparsity pattern of the ODE set Jacobian.

Concerning the solution of the NLPs, from the different possibilities available in our toolbox, the SQP method SNOPT (distributed as part of TOMLAB, Holmström et al., 2004) was selected as the first candidate to solve the proposed case studies, as it has been particularly successful for the solution of a set of challenging OCPs (García et al., 2005). SNOPT as a local NLP method requires a starting initial guess. In order to check for the possible multimodality of the problems under consideration a set of 50 initial guesses were used in a multistart procedure. As it will be illustrated later, all problems resulted to be non convex, which lead us to use a population based stochastic algorithm, Differential Evolution (DE, Storn and Price, 1997), to guarantee convergence to the best possible solution.

4.1 Case Study 1 (OCP1).

The solution of the OCP1 was first approached using a multistart strategy with SNOPT. The results show different productivities depending on the initial guess, revealing the presence of several sub-optimal solutions (Figure 2). This fact clearly indicates the need of using global optimization strategies to surmount convergence to a local minimum.

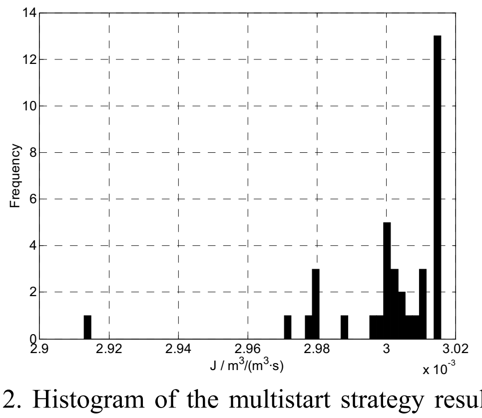

Fig 2. Histogram of the multistart strategy resulting from the dynamic optimization of OCP1 with ρ=2 and SNOPT (50 runs, J_{BEST}=3.016·10^{-3} $m^3/(m^3 \cdot s)$).

The stochastic global optimisation strategy DE was then applied using two variable-length time intervals since the practical implementation of more intervals would be difficult. The process productivity value obtained after 4 hours of computation was 3.313·10^{-3} $m^3/(m^3 \cdot s)$ with a raffinate purity of 85.00% (active constraint). The corresponding optimal control profile is shown in Fig. 3.

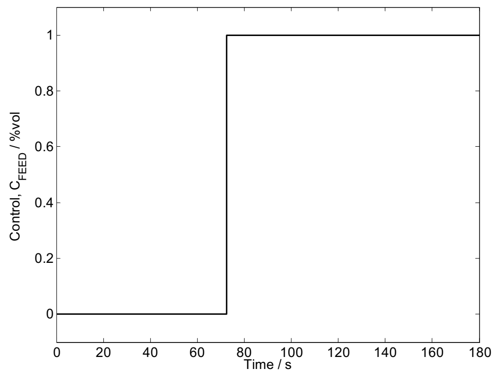

Fig 3. Optimal control profile for OCP1.

In the first period of the corresponding optimal control profile (t < 72.3s), the feed supplied to the unit is pure solvent, while in the second part (72.3s ≤ t < 180s) the feed concentration reached the maximum value (1%vol). The corresponding spatial evolution of the concentrations is shown in Fig. 4.

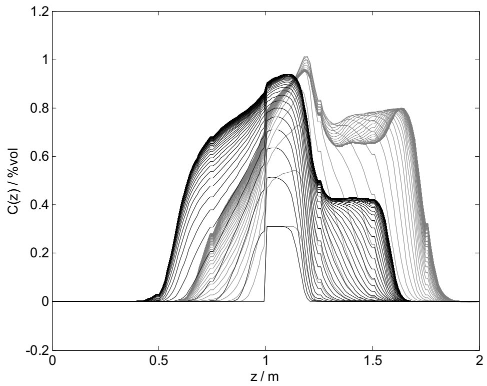

Fig 4. Spatial evolution of concentrations for OCP1.

These results are in agreement with the wave theory which states that the components of a mixture go across the SMB unit as nonlinear travelling waves and the movement of these waves affect the purities of the output streams (Schramm et al., 2003b). The wave propagation velocities influence the stream purities via the modification of the wave fronts.

It is clear that the modulation of the feed concentration has a direct influence on the fronts in zone III. The zero feed concentration at the beginning of the switching period involves a reduction of the wave height in zone III, which leads to a smaller propagation velocity of the wave of the more adsorbed component (black lines) than in the traditional process with constant feed concentration. Then, the feed concentration is increased and the opposite variation occurs, however not to the same extent, so that it does not balance the previous effect. Moreover, the front velocity modification is enhanced by the fact that at the end of every switching period the feed port is moved by one column in the direction of the fluid, so that the wave located in zone III is placed in zone II, just to the left of the feed node. Therefore the global effect of the modulation of the feed concentration is the decrease of the wave velocity propagation and the improvement of the raffinate purity.

Table 2 Comparison of OCP1 results with the classical SMB process and standard ModiCon.

	Classical Process	Standard ModiCon	OCP1
10^3 *PR*[m^3/(m^3·s)]	1.690	2.769	3.313
Pur_{Raf} [%]	85.00	96.06	85.00
% Improvement	--	[a] 64%	[a]96,[b]20

[a] Compared to classical SMB process
[b] Compared to standard ModiCon

The results of OCP1 are compared in Table 2 to those obtained through the optimization of the classical SMB process (ρ=1) and the standard ModiCon procedure. The classical process operated with an optimal feed concentration of 0.3052%vol led to PR=$1.690 \cdot 10^{-3}$ m^3/(m^3·s) and 85% purity, while the recommended ModiCon two fixed length steps profile (Schramm et al., 2003a) resulted in a PR value of $2.769 \cdot 10^{-3}$ m^3/(m^3·s) with a 96.06% purity. The productivity using a four-step profile as proposed by Schramm et al. (2003b) is considerably smaller ($1.3845 \cdot 10^{-3}$ m^3/(m^3·s)), but corresponds to a very high purity (>99.999%). Note that the reported results do not include a re-optimization of the internal flow rates, via a regulation method as in Schramm et al. (2003b).

To complete the study of OCP1, it is interesting to modify the minimum required value of the raffinate purity in order to see the impact of this constraint. For different purities, the optimal feed concentration profiles only differ in the time of switching between the zero and maximum feed concentration. Thus, this time of switching becomes the unique degree of freedom in the two-step dynamic optimization problem.

Figure 5 presents the evolution of the process productivity and the raffinate purity as functions of the time of switching. It is apparent from this Figure that the methodology proposed in this work allows the computation of the optimal solution independently of the desired purity value, whereas the two-step ModiCon solution provides a single reachable solution. This ModiCon solution would be suboptimal (for $Pur_{Raf} < 96.06\%$), infeasible (for $Pur_{Raf} > 96.06\%$) and optimal only when the required purity is 96.06%. This emphasizes how adding flexibility in the solution of the optimal problem results in added flexibility for the process operation since a specific time of switching is related to a purity and a productivity.

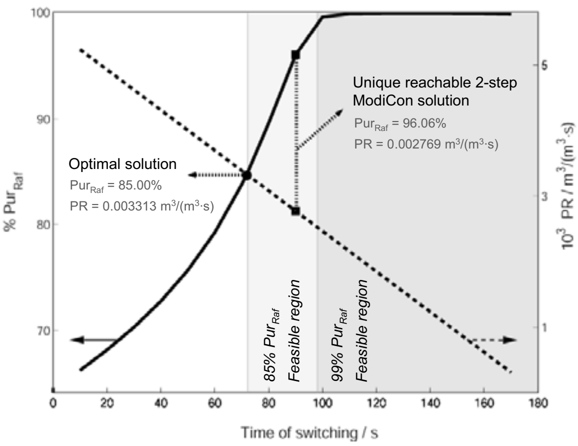

Fig 5. Evolution of the raffinate purity and the productivity versus the time of switching (OCP1 with two feed steps).

4.2. Case Study 2 (OCP2).

In this case study, the feed flow rate is considered as an additional control variable. Therefore the following decision variables are now calculated: the first step duration, the two steps heights for the feed concentration and the feed flow rate.

Using DE a process productivity of $3.363 \cdot 10^{-3}$ m^3/(m^3·s) is obtained. This maximum PR corresponds to a zero feed concentration at the beginning of the switching time followed by a saturated concentration (1%vol) in the second part of the period (as in the best results of OCP1). However, in this case, the first feed concentration step has a shorter duration (t = 47.0s) to promote higher productivity, while the value of the optimal feed flow rate is smaller (14.58 ml/min) in order to fulfil the imposed constraint. This optimal feed flow rate contributes to achieve the desired purity as it involves a smaller value of the internal flow rate Q_3, modifying the triangle of Storti in such a way that the new optimal point allows a higher overall concentration. Thus, the necessary duration of the zero-concentration step is smaller, so as the reduction of the wave height in zone III (Figure 6). This consequently increases the duration of the second feed concentration step, which leads to a higher process productivity (maximum value). Nevertheless, the overall increase of productivity as compared to OCP1 is only 1.5%; see Table 3.

Table 3 Comparison of OCP2 results with the classical SMB process and standard ModiCon. For active constraint (85%).

	Classical Process	Standard ModiCon	OCP2
10^3 *PR*[m^3/(m^3·s)]	2.691	3.211	3.363
Q_{FEED} [ml/min]	14.19	20.57	14.58
% Improvement	--	[a] 19%	[a]25,[b]5

[a] Compared to classical SMB process
[b] Compared to standard ModiCon

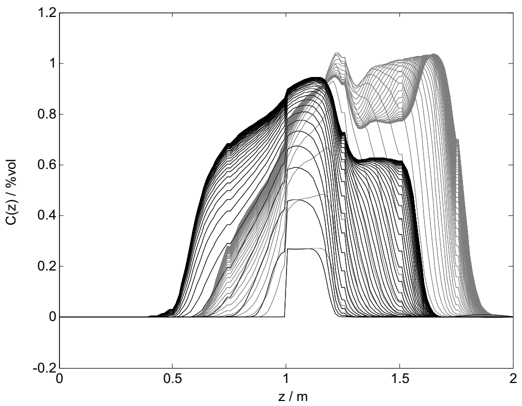

Fig 6. Spatial evolution of the concentrations for OCP2.

Finally several new cases were consider in order to check the influence of parameter perturbations on the solution. The results show that the optimal PR is not particularly sensitive to parameter variations since changes up to 10% affect in no more than a 5% the values of PR

5. CONCLUSIONS

In this work we stated and solved two open-loop OCP related to the chromatographic SMB process. For their solution, we applied a control vector parameterization approach in combination with state of the art IVP and global and local NLP solvers.

The use of robust optimization techniques allows for the computation of the optimal operating conditions, i.e. feed concentration, flow rate, switching time, etc, with maximum flexibility simultaneously considering practical constraints.

The capabilities of this methodology were illustrated through the solution of the proposed problems. The results obtained show that significant improvements, as compared to existing procedures, can be obtained using the computed optimal policies. Note that these operating conditions cannot be calculated via any of the classical approaches.

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