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Time-optimal Diafiltration in the Presence of Membrane Fouling

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Abstract: This paper is a preliminary study that deals with time-optimal control of a batch membrane diafiltration processes where fouling of the equipped membrane is pronounced. We account for the membrane fouling by its dynamic model where the pore blocking mechanism applies. It is assumed that due to the deposit of foulants, the radius of membrane pores decreases and the part of membrane surface becomes unavailable for the filtration. We apply Pontryagin's minimum principle to solve the time-optimal control problem in an analytical fashion. It is found that the analytical approach enables to fix the control structure into sequence of arcs. It is further shown that once the sequence of control arcs is fixed, the optimal solution is determined by identification of switching times between the control arcs using a simple numerical technique. The method is demonstrated by its application to some of the most commonly used models of diafiltration processes.

Keywords: diafiltration, optimal control, limiting flux, Pontryagin's minimum principle, membrane fouling, pore blocking model

1. INTRODUCTION

Diafiltration (DF) is a unique membrane process for separation of two or more solutes in a solution. It found many applications in pharmaceutical, chemical and biotechnological industry (Jönsson and Trägårdh, 1990). The governing principle of separation is based on the molecular size differences of the solutes which pass through the perm-selective membrane with different rate. The process is usually designed to increase the concentration of the valuable product (high molecular weight component, macro-solute) and to decrease the concentration of impurities (low molecular weight components, micro-solutes).

In this work, we study batch DF which operates under constant pressure and temperature. The control of the process is achieved via addition of a solute-free solvent (diluant, normally water) into the system in order to influence the solutes concentrations and to achieve the desired separation degree. Several studies show that the use of different diluant addition profiles may result in different operational savings (Foley, 1999). For example, one can achieve time-optimal separation of the solutes.

There have been several works devoted for optimization of diluant addition in DF process. These either optimize the switching times between the arbitrarily predefined operation modes, such as concentration (C) or constant-volume diafiltration (CVD) modes (Foley, 1999), or more sophisticated analytical and numerical approaches are exploited (Ng et al., 1976; Takači et al., 2009). Our recent study (Paulen et al., 2012) showed application of Pontryagin's minimum principle to address the minimum time problems of common DF processes in completely analytical way. None of the DF optimization studies mentioned in this paragraph neither other ones, present currently in the literature, deal with the membrane fouling phenomena.

Aging of membrane by its fouling stands for one of the major obstacles for wider application of membrane separation technology in the industry. Due to the fouling, operational expenditures rise as a consequence of membrane replacement and (complete or partial) cleaning (Fane, 1997). Nonetheless, the use of partially fouled membrane inhibits the filtration due to the smaller effective membrane area being available for filtered solution to penetrate. Consequently, processing time increases in batch operations such as batch DF. This study is motivated by the possibility of running the batch plant time-optimally in the presence of

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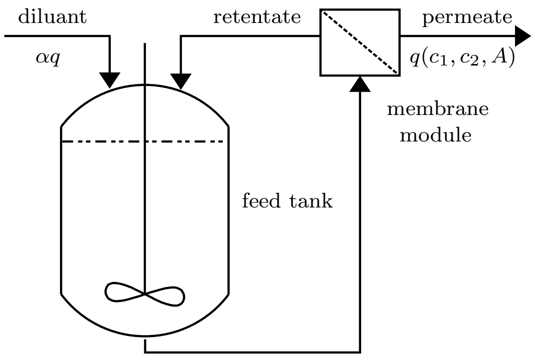

Fig. 1. Schematic representation of a generalized diafiltration process.

fouling and thus achieving minimum fouling operation of the batch.

In this work, we consider batch DF process where utilized membrane provides perfect rejection of macro-solutes and absolute permeability for micro-solute. We consider fouling to be present due to the blocking of the membrane pores. We employ the fouling model developed by Bolton et al. (2006). The minimum time optimization problem is resolved via application of Pontryagin's minimum principle.

The paper is organized as follows. We briefly explain the DF process (Section 2), we define the studied optimal control problem (Section 3), and we provide the definition of the optimal operation (Section 4). Next, we apply the results on two case studies from open literature and we compare the obtained results with traditionally used control approaches.

2. PROCESS DESCRIPTION

A schematic representation of a general batch DF process is shown in Fig. 1. The filtered solution consists of solvent and two solutes, macro-solute and micro-solute. The filtered solution is brought to the membrane from the feed tank. The membrane is designed to retain the macro-solute and to allow the micro-solute to pass through. The permeate, that leaves the system at flowrate q, is often found to be a function of concentrations of both solutes and effective membrane area. The retentate stream is taken back to the feed tank. The process control is achieved by adjusting the flowrate of diluant into the feed tank. The control variable α is defined as a ratio between the inflow of diluant into the feed tank and the outflow of the permeate q. There are several types of control strategies which consist of sequences of operation modes. These modes differ by the rate of diluant utilization:

- concentration (C) mode when $\alpha = 0$,
- variable-volume diafiltration (VVD) when $\alpha \in (0, 1)$,
- constant-volume diafiltration (CVD) when $\alpha = 1$.
- instantaneous pure dilution (D) when $\alpha \to \infty$.

Optimality of arbitrarily preselected, traditional control strategies is questionable. Here the goal is to identify the optimal control strategy which does not necessarily result in traditionally used combination of modes (e.g. C-CVD).

Considering membrane with perfect rejection to macro-solute and absolute permeability to micro-solute, the mass balance for each solute can be written as

$$\frac{\mathrm{d}c_1}{\mathrm{d}t} = \frac{c_1^2 q}{c_{1,0}V_0}(1-\alpha), \qquad c_1(0) = c_{1,0}, \tag{1}$$

$$\frac{\mathrm{d}c_2}{\mathrm{d}t} = -\frac{c_1 c_2 q}{c_{1,0}V_0}\alpha, \qquad c_2(0) = c_{2,0}, \tag{2}$$

where c_1 and c_2 represent the concentration of macro and micro-solute, respectively, V_0 stands for initial volume of the processed solution, and $q(c_1, c_2, A)$ denotes the permeate flowrate such that

$$q(c_1, c_2, A) = AJ(c_1, c_2), \tag{3}$$

where A represents an effective membrane area and $J(\cdot)$ stands for the permeate flux subject to unit membrane area and is generally a function of both concentrations.

In this work, we investigate the process where fouling of the membrane occurs due to blocking of the membrane pores. This phenomenon was previously reported by Makardij et al. (1999) to be the main cause of fouling in ultrafiltration of milk. Blockage of the membrane pores affects the effective membrane area. We use a model suggested and validated by Bolton et al. (2006) which reads as follows

$$\frac{\mathrm{d}A}{\mathrm{d}t} = -A_0\frac{K_b}{q_0}q = -\frac{K_b}{J_0}AJ, \quad A(0) = A_0, \tag{4}$$

where K_b represents the fouling rate and

$$q_0 = q(c_{1,0}, c_{2,0}, A_0) = A_0 J(c_{1,0}, c_{2,0}) = A_0 J_0, \tag{5}$$

with q_0 and J_0 being initial fluxes when effective membrane area A_0 is available.

3. PROCESS OPTIMIZATION

The objective of the optimization is to find such time-dependent function $\alpha(t)$ which drives the process from initial to final concentrations in minimum time. The mathematical formulation of this dynamic optimization problem is as follows

$$\min_{\alpha(t)} \int_0^{t_\mathrm{f}} 1\,\mathrm{d}t, \tag{6a}$$

s.t.

$$\frac{\mathrm{d}c_1}{\mathrm{d}t} = \frac{c_1^2 AJ}{c_{1,0}V_0}(1-\alpha), \quad c_1(0) = c_{1,0}, \quad c_1(t_\mathrm{f}) = c_{1,\mathrm{f}}, \tag{6b}$$

$$\frac{\mathrm{d}c_2}{\mathrm{d}t} = -\frac{c_1 c_2 AJ}{c_{1,0}V_0}\alpha, \quad c_2(0) = c_{2,0}, \quad c_2(t_\mathrm{f}) = c_{2,\mathrm{f}}, \tag{6c}$$

$$\frac{\mathrm{d}A}{\mathrm{d}t} = -\frac{K_b}{J_0}AJ, \qquad A(0) = A_0, \tag{6d}$$

$$\alpha \in [0, \infty). \tag{6e}$$

This optimization problem can be solved using various methods of dynamic optimization. As the process equations are affine in control, the optimal control profile might exhibit discontinuous behavior with several arcs, input-saturated and singular ones. Consequently, it is not advisable to use numerical optimal control (hence arbitrary control discretization) as the possibility of singular arc might deteriorate the performance of such scheme and the physical insight w.r.t. the truly optimal process might be lost.

In case the optimization problem is treated by means of numerical optimization, a suitable parametrization (piece–wise polynomial approximation of control profile, in general) has to be found out. Resulting problem of nonlinear programming (NLP) is then resolved having the knots of control profile and the time intervals as optimized variables (Goh and Teo, 1988). In order to avoid these issues, we make use of Pontryagin's minimum principle (Pontryagin et al., 1962; Bryson, Jr. and Ho, 1975) to solve the problem (6) analytically. We can rearrange (6b)–(6d) as

$$\dot{\boldsymbol{x}} = \boldsymbol{f}(\boldsymbol{x}) + \boldsymbol{g}(\boldsymbol{x})\alpha, \tag{7}$$

where $\boldsymbol{x} = (c_1, c_2, A)^T$. The Hamiltonian function can be then written as

$$\begin{aligned} H(\boldsymbol{x}, \alpha, \boldsymbol{\lambda}) &= 1 + \boldsymbol{f}^T(\boldsymbol{x})\boldsymbol{\lambda} + \boldsymbol{g}^T(\boldsymbol{x})\boldsymbol{\lambda}\alpha \\ &= H_0(\boldsymbol{x}, \boldsymbol{\lambda}) + H_\alpha(\boldsymbol{x}, \boldsymbol{\lambda})\alpha, \end{aligned} \tag{8}$$

where $\boldsymbol{\lambda} = (\lambda_1, \lambda_2, \lambda_3)^T$ is the vector of adjoint variables which are defined from

$$\dot{\boldsymbol{\lambda}} = -\frac{\partial H}{\partial \boldsymbol{x}} = -(\boldsymbol{f}_x + \boldsymbol{g}_x\alpha)\boldsymbol{\lambda}, \tag{9}$$

where

$$\boldsymbol{f}_x(\boldsymbol{x}) = \frac{\partial \boldsymbol{f}^T(\boldsymbol{x})}{\partial \boldsymbol{x}}, \quad \boldsymbol{g}_x(\boldsymbol{x}) = \frac{\partial \boldsymbol{g}^T(\boldsymbol{x})}{\partial \boldsymbol{x}}. \tag{10}$$

According to Pontryagin's minimum principle the optimal solution to (6) minimizes the Hamiltonian function. Since Hamiltonian is affine in α, its minimum is attained with α on its boundaries (bang-bang control) as

$$\alpha = \begin{cases} 0 & \text{if } H_\alpha > 0, \\ \infty & \text{if } H_\alpha < 0. \end{cases} \tag{11}$$

If $H_\alpha = 0$ then the Hamiltonian is singular and does not depend on α. We use the fact that condition $H_\alpha = 0$ implies the derivatives of H_α w.r.t. time to be equal to zero. Then we obtain a set of equations linear in $\boldsymbol{\lambda}$

$$H_\alpha(\boldsymbol{x}, \boldsymbol{\lambda}) = \boldsymbol{g}^T\boldsymbol{\lambda} = 0, \tag{12a}$$

$$\dot{H}_\alpha(\boldsymbol{x}, \boldsymbol{\lambda}) = \boldsymbol{h}^T\boldsymbol{\lambda} = 0, \tag{12b}$$

$$\ddot{H}_\alpha(\boldsymbol{x}, \boldsymbol{\lambda}, \alpha) = (\boldsymbol{h}_x\boldsymbol{f} - \boldsymbol{f}_x\boldsymbol{h} + (\boldsymbol{h}_x\boldsymbol{g} - \boldsymbol{g}_x\boldsymbol{h})\alpha)\boldsymbol{\lambda} = 0, \tag{12c}$$

where

$$\boldsymbol{h}(\boldsymbol{x}) = \boldsymbol{g}_x\boldsymbol{f} - \boldsymbol{f}_x\boldsymbol{g}, \quad \boldsymbol{h}_x = \frac{\partial \boldsymbol{h}^T(\boldsymbol{x})}{\partial \boldsymbol{x}}. \tag{13}$$

We will also define

$$J_i = \frac{\partial J}{\partial c_i}, \qquad J_{ij} = \frac{\partial^2 J}{\partial c_i \partial c_j}, \qquad i, j = 1, 2. \tag{14}$$

4. OPTIMAL OPERATION

In this section, we state the main theoretical results and we define the time-optimal operation of general batch DF process.

4.1 Optimal Operation without Fouling

Let us first study the optimal operation in the case where no fouling occurs, i.e. $K_b = 0$, $\lambda_3 = 0$. Elimination of adjoint variables from the conditions (12a) and (12b) results in the expression for *singular surface* in the state space

$$S(c_1, c_2) = \det(\boldsymbol{g}, \boldsymbol{h}) = J + c_1J_1 + c_2J_2 = 0, \tag{15}$$

and it depends on concentrations only. The corresponding singular control that keeps the state on singular surface can be found either by elimination of adjoint variables from (12) or by differentiation of the singular surface (15) w.r.t. to time

$$\begin{aligned} \alpha_{\text{sing}}(c_1, c_2) &= \frac{\frac{\partial S}{\partial c_1}c_1}{\frac{\partial S}{\partial c_1}c_1 + \frac{\partial S}{\partial c_2}c_2} \\ &= \frac{c_1\left(2J_1 + c_1J_{11} + c_2J_{12}\right)}{\sum_{j=1}^{2} c_j\left(2J_j + \sum_{i=1}^{2} c_iJ_{ij}\right)}. \end{aligned} \tag{16}$$

The optimal operation consists of three arcs and it is defined by

(1) The control in first step is step is found from

$$\alpha = \begin{cases} 0 & \text{if } S(c_{1,0}, c_{2,0}) > 0, \\ \infty & \text{if } S(c_{1,0}, c_{2,0}) < 0. \end{cases} \tag{17}$$

It is applied until the condition $S(c_1, c_2) = 0$ is met.

(2) In the second step, the state reside on the singular surface with singular control from (16).

(3) The last step uses again either pure concentration or dilution mode until the final concentrations are reached.

The duration of the singular step is fully determined by the last step and the final conditions on concentrations. More details on these results can be found in our previous study (Paulen et al., 2012).

4.2 Optimal Operation with Fouling

When fouling behavior occurs, $K_b \neq 0$, $\lambda_3 \neq 0$. Therefore, it is no longer possible to obtain singular state surface from the conditions (12a) and (12b).

However, the expression for the singular control can still be derived by elimination of adjoint variables from (12). This step uses

$$\begin{vmatrix} \boldsymbol{g}^T \\ \boldsymbol{h}^T \\ \boldsymbol{h}_x\boldsymbol{f} - \boldsymbol{f}_x\boldsymbol{h} + (\boldsymbol{h}_x\boldsymbol{g} - \boldsymbol{g}_x\boldsymbol{h})\alpha \end{vmatrix} = 0, \tag{18}$$

which gives

$$\alpha_{\text{sing}}(c_1, c_2, A) = \frac{L_1}{L_2} - \frac{L_3}{L_2}K_b, \tag{19}$$

where

$$L_1 = 2c_1c_2J_1J_2 - c_1c_2J_{12}J + 2c_1^2J_1^2 - c_1^2J_{11}J, \tag{20}$$

$$\begin{aligned} L_2 = &2c_1^2J_1^2 + 2c_2^2J_2^2 - c_1^2J_{11}J - c_2^2J_{22}J - 2c_1c_2J_{12}J, \\ &+ 4c_1c_2J_1J_2 \end{aligned} \tag{21}$$

$$L_3 = \frac{V_0c_{1,0}J}{Ac_1J_0}(c_1J_1 + c_2J_2). \tag{22}$$

It is straightforward to show that $K_b = 0$ and (15) reduce this singular control to (16).

The optimal operation structure remains the same as in the previous section, i.e. three steps with control on the boundaries in the first and the last step. The middle step is characterized by singular control. Note that the control over all intervals of the three-step control strategy is completely characterized by PMP. The difference to the case without fouling is given by the fact that it is not anymore possible to decide when to switch between the individual phases. Therefore, we propose here to solve a small NLP problem that provides the lengths (time durations) of the first two control intervals. Such NLP is very easily solvable with only a few iterations and converges without difficulties.

5. CASE STUDIES

We apply the theoretical results derived in above section to study the optimal operation on two case studies. In order to allow for a fair comparison of the results obtained in both investigated case studies, we assume the initial effective membrane area to be $A_0 = 1\,\mathrm{m}^2$ and we choose the values of K_b such that these become inversely proportional to the amount of permeate processed for the concentration of macro-solute relative to the initial volume of the processed solution.

$$\Gamma = 1 - \frac{c_{1,0}}{c_{1,\mathrm{f}}}. \tag{23}$$

5.1 *Diafiltration at Limiting Flux*

We consider a membrane plant which operates under limiting flux conditions which is the common model in membrane separation (Aimar and Field, 1992). The permeate flux is given by

$$J(c_1) = k \ln \frac{c_{\mathrm{lim}}}{c_1}, \tag{24}$$

where k is the mass transfer coefficient and c_{lim} represent the limiting concentration for of macro-product. This example was treated in Jelemenský et al. (2013) without the pore blocking model. In this example we demonstrate the time-optimal operation on the case when $c_{\mathrm{lim}} = 319\,\mathrm{mol/m^3}$, $k = 0.0172\,\mathrm{m/h}$. The goal is to process 100 L of solution from initial point $c_{1,0} = 10\,\mathrm{mol/m^3}$, $c_{2,0} = 100\,\mathrm{mol/m^3}$ to final point $c_{1,\mathrm{f}} = 100\,\mathrm{mol/m^3}$, $c_{2,\mathrm{f}} = 1\,\mathrm{mol/m^3}$.

The initial and final concentrations determine the first and the third step of the optimal control. In the first step we use concentration mode ($\alpha = 0$). NLP problem will provide time interval length and optimal concentration of the macro-solute to switch to singular surface. As the last step is characterized by dilution mode ($\alpha \to \infty$), the optimal concentration to switch from the singular arc is fully determined from the final concentrations. The ratio of the concentrations at time of the switch should be equal the ratio of their final values. The optimal control in the singular arc (19) has the following form

$$\alpha_{\mathrm{sing}}(c_1, A) = 1 - \frac{V_0 c_{1,0}}{A c_1 J_0}\left(1 - \frac{2}{\ln \frac{c_1}{c_{\mathrm{lim}}} + 2}\right) K_b. \tag{25}$$

If there is no fouling ($K_b = 0$) then the singular control is equal to one.

In Fig. 2 we show the time-optimal operation for different values of fouling rate. The top figure plots state trajectories. We start at initial concentrations of both solutes (green circle) and finish at the red cross. The bottom figure shows the corresponding optimal control. We can observe that by increasing the value of fouling rate the processing time is increased. This behavior was expected as increased fouling decreases the effective membrane area.

Table 1 summarizes the comparison of final time in case of minimum time and traditionally used operation for different values of fouling rates. The traditionally used operation consists of concentration mode followed by constant-volume diafiltration mode (C-CVD) where the switching from C to CVD operation is performed at $c_1 = c_{1,\mathrm{f}}$. We can observe that in all cases the minimum time and traditionally used operation have similar final processing time, however, the difference (Δ) increases with the increase of K_b. In this case we can conclude that for small values of the fouling rate it is not necessary to use advanced control strategy but instead we can use the traditional operation.

Further we can also compare the optimal operations from Section 4.1 where model assumes no fouling. Clearly, this

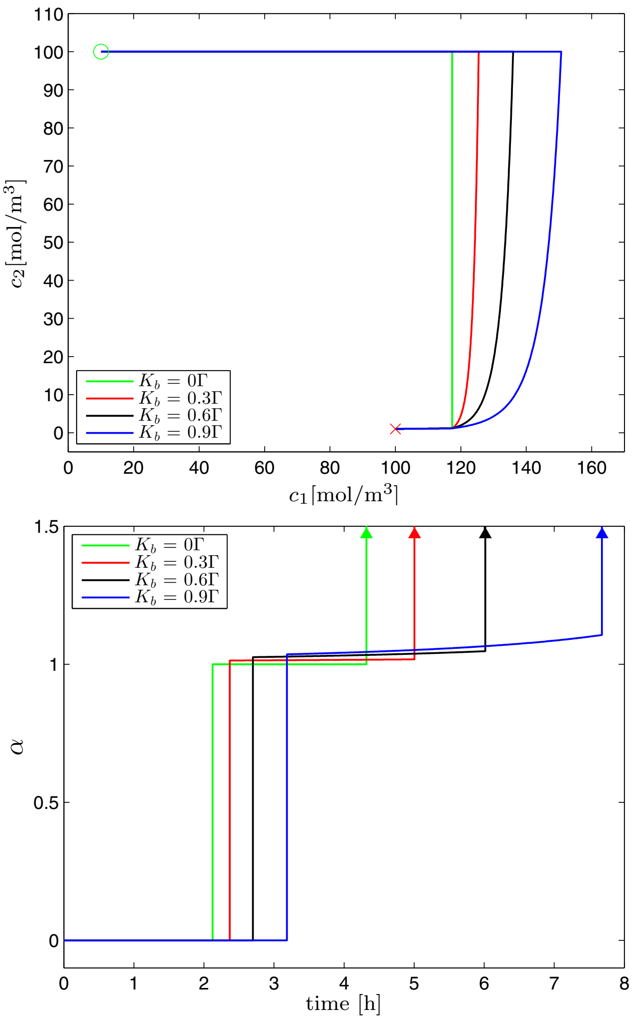

Fig. 2. Concentration state diagram and optimal control profiles for DF at limiting flux conditions with different fouling rates.

Table 1. Time-optimal operation of DF under limiting flux conditions compared with traditionally used operation with different values of fouling rate.

$K_b[10^{-4}\,\mathrm{s}^{-1}]$	minimum time t_f [h]	C-CVD t_f [h]	Δ[%]
0Γ	4.32	4.35	0.61
0.3Γ	5.01	5.06	1.05
0.6Γ	6.02	6.14	2.05
0.9Γ	7.68	8.06	4.73

strategy would be suboptimal but has an advantage that no NLP needs to be solved. We used the largest considered fouling rate $K_b = 0.9\Gamma$. The final processing time is 7.79 h whereas the optimal operation is 7.68 h (difference 1.41%). It is questionable in this case whether the optimal fouling control will be justified.

5.2 Separation of Lactose from Proteins

Here we consider the separation of lactose (with concentration c_2) from proteins (with concentration c_1). This problem was originally formulated in Rajagopalan and Cheryan (1991) where model of the permeate flux was obtained experimentally to be

$$J(c_1, c_2) = b_0 + b_1 \ln c_1 + b_2 \ln c_2 = 63.42 - 12.439 \ln c_1 - 7.836 \ln c_2. \quad (26)$$

The goal is to process 100 dL of solution in order to increase the concentration c_1 from $c_{1,0} = 3.3$ g/dL to $c_{1,f} = 9.04$ g/dL and to simultaneously decrease the concentration of lactose from $c_{2,0} = 5.5$ g/dL to $c_{2,f} = 0.64$ g/dL. Similarly to the first case study, the initial and final concentrations determine the first and the third step of the optimal control to be concentration ($\alpha = 0$) and dilution mode ($\alpha \to \infty$), respectively. Using (19) the control on singular surface is found to be

$$\alpha_{\text{sing}}(c_1, c_2, A) = \frac{b_1}{b_1 + b_2} - \frac{V_0 c_{1,0} J}{A c_1 J_0 (2b_1 + 2b_2 + J)} K_b. \quad (27)$$

Hence that this boils down to simple VVD operation when $K_b = 0$. The switching times to commence and to end the singular control are again determined by the resolution of a simple NLP.

In Fig. 3, we show the optimal control strategies for minimum time operation for different fouling rates (state diagram in top figure, the respective control actions in the bottom one). We can observe that the numerically determined singular surface (evolution of the concentrations under control (27)) is different for different values of blocking rate, so we can conclude that it depends significantly on the value of K_b. Furthermore we can observe that the increase of this value translates to longer processing time, as expected.

It can also be observed that the singular control profiles differ more significantly from any of the traditional operation modes as compared to the profiles obtained in previous case study. This observation is reflected and quantified in Table 2 which shows the comparison of final times for minimum time and C-CVD operations with different values of fouling rate. We can observe that the increased

Table 2. Time-optimal operation of DF for separation of lactose and proteins compared with traditionally used operation with different values of fouling rate.

$K_b[10^{-4}\,\text{s}^{-1}]$	minimum time t_f [h]	C-CVD t_f [h]	Δ[%]
0Γ	4.49	4.74	5.31
0.3Γ	5.12	5.52	7.26
0.6Γ	6.04	6.81	11.24
0.9Γ	7.52	9.69	22.38

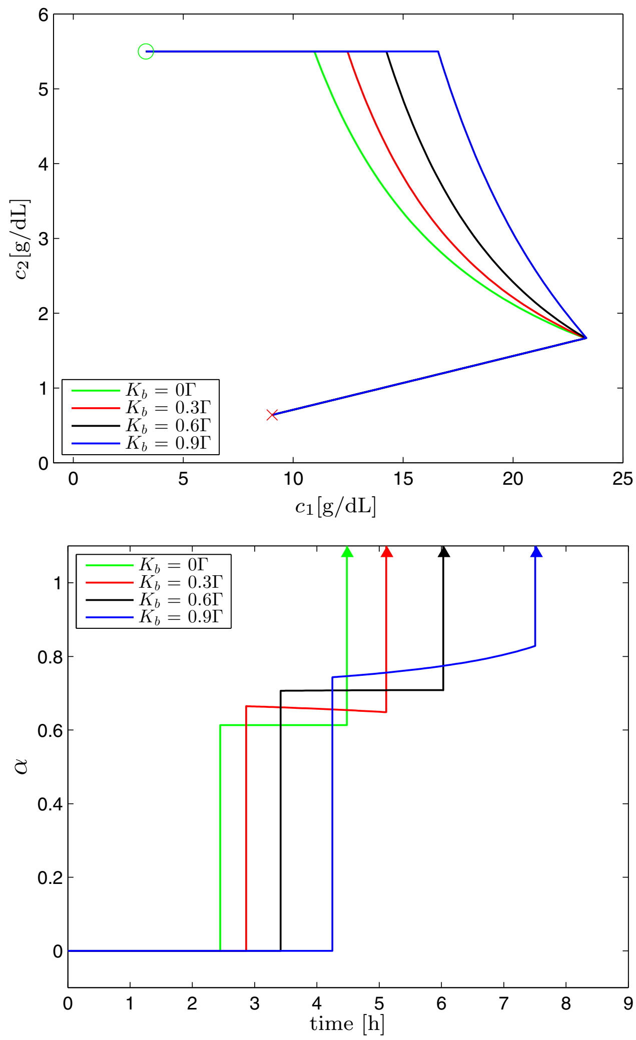

Fig. 3. Concentration state diagram and optimal control profiles for separation of lactose from proteins with different fouling rates.

value of K_b pronounces the differences in obtained processing times between minimum time and traditionally used operation. In this case we can conclude that by using advanced control strategy we can significantly reduce the production costs. Moreover, we can claim that with the increased complexity of the process model the differences between traditional and optimal control strategies will be amplified in terms of savings of processing time.

An interesting conclusions can be also made when the effective membrane area at the end of batch, $A(t_f)$, is evaluated. This is always found to be larger after performing the time-optimal control of the batch. As the value of processing time is closely coupled to the effective membrane area, this phenomenon can be expected. In Fig. 4, we represent the evolution of effective membrane area in 7 consecutive batches under minimum time and traditional (C-CVD) control where $K_b = 0.6\Gamma \times 10^{-4}\,\text{s}^{-1}$. We assume the cleaning of the membrane to be done after each batch and the efficiency of the cleaning to be 95% w.r.t. to the effective area being available at the beginning of the batch (taken from (Zhang and Liu, 2002)). We assume that the membrane life-cycle is 7 batches and the cleaning costs are

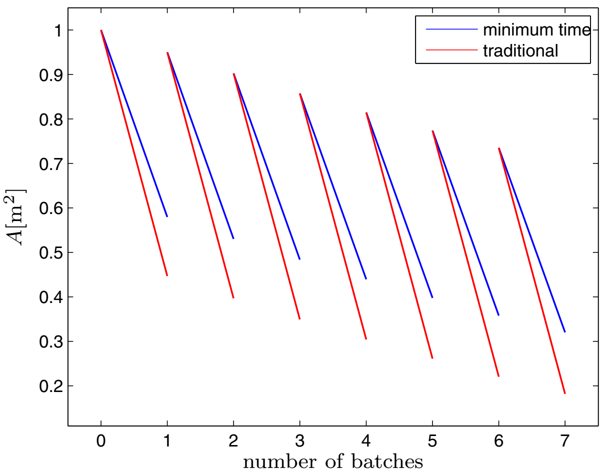

Fig. 4. Reduction of membrane area due to fouling during 7 batches for separation of lactose from proteins.

proportional to the level of fouling of the membrane. When the costs for cleaning are evaluated over the life-cycle of the membrane, we find the costs from cleaning the membrane after performing minimum time operation to be reduced by 27% in comparison with cleaning the membrane after traditional used operation being performed. This reveals additional advantages of using the time-optimal control strategy.

As in the previous case study, we can compare the processing time of the optimal operations from Section 4.1 where the model assumes no fouling. This strategy is found to be suboptimal but the difference to the truly optimal operation (given in Section 4.2) is rather small (1.8%) even when the highest considered fouling rate ($K_b = 0.9\Gamma \times 10^{-4}\,\text{s}^{-1}$) is considered. It is again questionable whether the optimal fouling control will be justified. This result has to be viewed in the light of the assumptions made for the process model, relatively simple flux model structures, and the sole mechanism of the membrane considered.

6. CONCLUSIONS

In this paper we studied the time-optimal control for diafiltration process in the presence of fouling. We formulated the optimal control problem and we solved this problem for the general batch DF plant using Pontryagin's minimum principle. The sequence of controls arcs was firstly determined and the simple numerical procedure was proposed for finding the switching times between the arcs.

We demonstrated the developed theory on two case studies where we compared the minimum time strategy with the traditionally used ones. The obtained results indicate that the traditional operation is sufficient with lower fouling rates and simple flux models. On the other hand, once fouling becomes a major issue or when the behavior of the process gets more complex, advanced control strategy reduces the processing time, production costs, and cleaning costs.

There are several fouling models described in the literature. This is a preliminary analysis based on a particular one. Further work will be needed to obtain more general results.

REFERENCES

Aimar, P. and Field, R. (1992). Limiting flux in membrane separations: A model based on the viscosity dependency of the mass transfer coefficient. *Chemical Engineering Science*, 47(3), 579–586.

Bolton, G.R., Boesch, A.W., and Lazzara, M.J. (2006). The effects of flow rate on membrane capacity: Development and application of adsortive membrane fouling models. *Journal of Membrane Science*, 279, 625–634. doi:10.1016/j.memsci.2005.12.057.

Bryson, Jr., A.E. and Ho, Y.C. (1975). *Applied Optimal Control*. Hemisphere Publishing Corporation.

Fane, A. (1997). *Control of concentration polarisation in membrane process by process operation strategies and module design*. Proceedings of the Symposium on Characterization of Polymers with Surface, Lappeenranta, Finland.

Foley, G. (1999). Minimisation of process time in ultrafiltration and continuous diafiltration: the effect of incomplete macrosolute rejection. *Journal of Membrane Science*, 163(1–2), 349–355.

Goh, C.J. and Teo, K.L. (1988). Control parameterization: a unified approach to optimal control problems with general constraints. *Automatica*, 24(1), 3–18.

Jelemenský, M., Paulen, R., Fikar, M., and Kovács, Z. (2013). Multi-objective optimal control of ultrafiltration/diafiltration processes. In *Proceedings of the European Control Conference*, 3384–3389. Zurich, Switzerland.

Jönsson, A.S. and Trägårdh, G. (1990). Ultrafiltration applications. *Desalination*, 77, 135 – 179. doi: 10.1016/0011-9164(90)85024-5. Proceedings of the Symposium on Membrane Technology.

Makardij, A., Chen, X., and Farid, M. (1999). Microfiltration and ultrafiltration of milk: Some aspects of fouling and cleaning. *Food and Bioproducts Processing*, 77(2), 107 – 113. doi:10.1205/096030899532394.

Ng, P., Lundblad, J., and Mitra, G. (1976). Optimization of Solute Separation by Diafiltration. *Separation Science and Technology*, 11(5), 499–502.

Paulen, R., Fikar, M., Foley, G., Kovács, Z., and Czermak, P. (2012). Optimal feeding strategy of diafiltration buffer in batch membrane processes. *Journal of Membrane Science*, 411-412, 160–172. doi: 10.1016/j.memsci.2012.04.028.

Pontryagin, L.S., Boltyanskii, V.G., Gamkrelidze, R.V., and Mishchenko, E.F. (1962). *The Mathematical Theory of Optimal Processes*. Wiley (Interscience), New York, New York.

Rajagopalan, N. and Cheryan, M. (1991). Process optimization in ultrafiltration: Flux-time considerations in the purification of macromolecules. *Chemical Engineering Communications*, 106(1), 57–69.

Takači, A., Žikić-Došenović, T., and Zavargó, Z. (2009). Mathematical model of variable volume diafiltration with time dependent water adding. *Engineering Computations: International Journal for Computer-Aided Engineering and Software*, 26(7), 857–867.

Zhang, G. and Liu, Z. (2002). Membrane fouling and cleaning in ultrafiltration of wastewater from banknote printing works. *Journal of Membrane Science*, 211, 235–249.