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Multi-objective optimal control of ultrafiltration/diafiltration processes

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Abstract— **This paper deals with multi-objective optimal operation of a general batch ultrafiltration/diafiltration (UF/DF) process. We consider the optimization criterion consisting of two parts as combination of two classical optimization problems, namely the minimum time problem and the problem of minimum diluant consumption. We apply Pontryagin's minimum principle for obtaining the necessary conditions for optimality. Based on these, we show how to obtain a Pareto front and how to derive multi-objective optimal operation for considered type of processes. The presented methodology will be applied to the most commonly used model in membrane filtration – the limiting flux case and to another experimentally derived membrane model.**

I. INTRODUCTION

Diafiltration is an effective membrane process for separation of two or more solutes from solution. When semipermeable ultrafiltration membrane is used, such process is traditionally exploited to increase the concentration of proteins in the solution and to decrease the concentration of lower molecular weight impurities within a specified level [9], [16].

In this work, we consider batch process which operates under constant pressure and temperature. We control the process by influencing the concentrations of solutes to achieve the desired separation of solutes. This can be done by adding the solute-free solvent (diluant, water in this case) into the feed tank. By a good choice of dynamic profile of adding the diluant we can achieve the desired final state (concentrations of solutes) in minimum time and/or with minimal cost of consumed diluant.

Several works have been devoted for optimization of adding the diluant during the diafiltration process. The earliest investigators [11] used limiting flux model of transmembrane (permeate) flow and derived optimal concentration to start constant-volume diafiltration (CVD) step. Since then, there have been a number of attempts to treat the time-optimal and minimum diluant problems of diafiltration. These either optimize switching times between arbitrarily predefined operational modes [2], [6], [19] or they numerically find approximations to optimal control [18], [5], [14]. In our previous studies [12], [13], we have shown an application of Pontryagin's minimum principle to minimum time and minimum diluant consumption problems and we have analytically derived respective optimal operations.

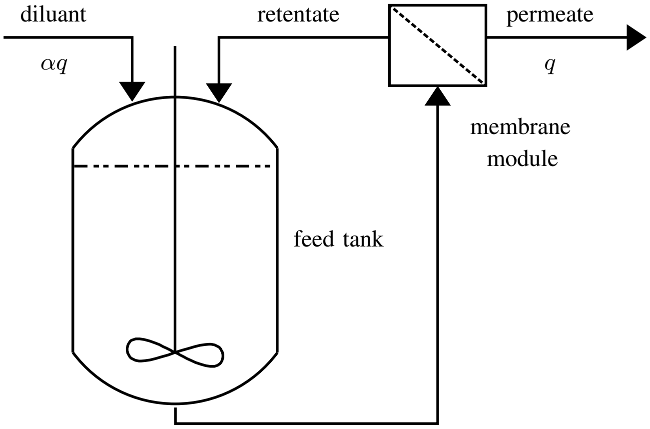

Fig. 1. Schematic representation of a generalized UF/DF process.

Economical operation of industrial plants, however, requires simultaneous minimization of process time (i.e. an equivalent to minimization of electricity and cooling costs) and consumption of diluant (commonly highly purified water). The motivation for this paper is to account for both of these cost factors that are traditionally handled separately. The relative importance of the cost contributions in the objective function is expressed by means of non-negative price coefficients which is a common way of formulating multi-objective optimization problems [3], [10].

The solution to treated multi-criterion optimization problem then provides information for the decision maker on economically optimal UF/DF process. In particular, the optimal operation of general UF/DF process can be easily evaluated once the unit price for electricity, cooling and diluant costs are known.

In the following, we introduce generalized UF/DF process model and we derive the optimality conditions for multi-objective optimal control problem. Based on these we provide a recipe for obtaining the optimal operation. Finally, we show the optimal operation for two selected case studies.

II. PROCESS DESCRIPTION

The schematic representation of diafiltration process is shown in Fig. 1. Filtered solution consists of solvent (water), micro-solute (lower molecular weight impurities) and macro-solute (higher molecular weight component, e.g. protein) which is passed from feed tank to ultrafiltration membrane module. The membrane module is designed to retain macro-solute and to allow the passage of micro-solute. Permeate stream leaves the system at a flowrate q which is often (under constant pressure and temperature conditions) a function of

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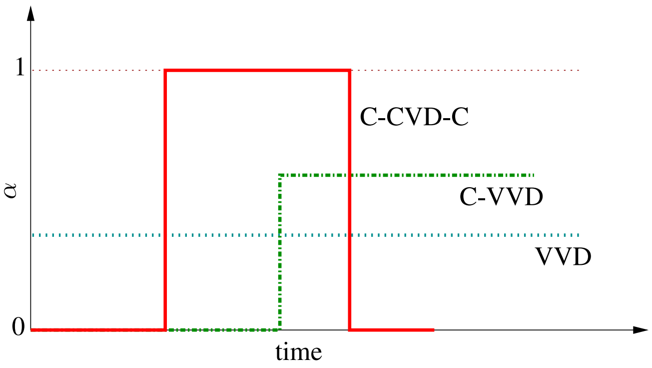

Fig. 2. Representation of traditional control strategies in terms of the α function.

concentrations of separated species. The retentate stream is introduced back into the feed tank. The process control is achieved by adjusting the flowrate of water to the feed tank. Control variable α is defined as a dimensionless ratio between the inflow of diluant to the feed tank and the outflow of permeate q.

We consider a process where a macro-solute is to be increased in concentration from $c_{1,0}$ to $c_{1,\mathrm{f}}$ and a micro-solute reduced in concentration from $c_{2,0}$ to $c_{2,\mathrm{f}}$. The standard three-step operation is to pre-concentrate, to diafiltrate with constant volume, and to optionally post-concentrate (C-CVD-C), defined in Fig. 2. This has been previously showed [14] to give time-optimal operation in some cases of diafiltration under limiting flux conditions (treated in Section V).

Another traditional diluant utilization approach, variable-volume diafiltration (VVD) possesses only 2 degrees of freedom (constant α value and process duration) and thus is very unlikely to achieve optimal operation. Suboptimality of VVD operation together with its modification, C-VVD, are to be investigated in this work.

We consider a solution with two species with concentrations c_1 and c_2. The balance of each solute can be written as,

$$\dot{c}_i = \frac{c_i q}{V}(R_i - \alpha), \quad c_i(0) = c_{i,0}, \quad i = 1,2 \tag{1}$$

where V is the retentate volume at time t. The rejection coefficient $R_i(c_1, c_2)$ is in general a function of both concentrations. The same holds for the permeate flowrate $q(c_1, c_2)$. The volume balance can be written as

$$\dot{V} = (\alpha - 1)q, \quad V(0) = V_0. \tag{2}$$

We assume that the utilized membrane is absolutely impermeable to macro-solute and perfectly permeable to impurities, i.e. $R_1 = 1$ and $R_2 = 0$. This assumption transforms the process model into the form

$$\dot{c}_1 = \frac{c_1^2 q}{c_{1,0} V_0}(1 - \alpha), \qquad c_1(0) = c_{1,0}, \tag{3}$$

$$\dot{c}_2 = -\frac{c_1 c_2 q}{c_{1,0} V_0}\alpha, \qquad c_2(0) = c_{2,0}. \tag{4}$$

III. Process optimization

The objective is to find the time dependent function $\alpha(t)$ which minimize the multi-objective function and drive the process from initial to final conditions. Mathematical formulation of this dynamic optimization problem is as follows

$$\min_{\alpha(t)} \int_0^{t_\mathrm{f}} (w_T + w_D \alpha q)\,\mathrm{d}t, \tag{5a}$$

s.t.

$$\dot{c}_1 = \frac{c_1^2 q}{c_{1,0} V_0}(1 - \alpha), \quad c_1(0) = c_{1,0},\ c_1(t_\mathrm{f}) = c_{1,\mathrm{f}}, \tag{5b}$$

$$\dot{c}_2 = -\frac{c_1 c_2 q}{c_{1,0} V_0}\alpha, \qquad c_2(0) = c_{2,0},\ c_2(t_\mathrm{f}) = c_{2,\mathrm{f}}, \tag{5c}$$

$$\alpha \in [0, \infty), \tag{5d}$$

where the coefficients $w_T \geq 0$ and $w_D \geq 0$ represent weights of final time (t_f) and total diluant consumption (V_w), respectively. The value of $\alpha = \infty$ represents, physically, a pure dilution step, i.e. pouring of a certain amount of diluant into the feed tank in one time instant. We note that such operation maintains constant ratio of concentrations c_1 and c_2.

We make use of Pontryagin's minimum principle [15], [4] to solve this problem. Multi-objective functional and process differential equations are affine in control. Due to this, Hamiltonian function takes control-affine form as well

$$\begin{aligned} H(\boldsymbol{x}, \alpha, \boldsymbol{\lambda}) &= w_T + w_D \alpha q + \lambda_1 \dot{c}_1 + \lambda_2 \dot{c}_2 \\ &= H_0(\boldsymbol{x}, \boldsymbol{\lambda}) + H_\alpha(\boldsymbol{x}, \boldsymbol{\lambda})\alpha, \end{aligned} \tag{6}$$

with $\boldsymbol{x} = (c_1, c_2)^T$ and vector of adjoint variables $\boldsymbol{\lambda} = (\lambda_1, \lambda_2)^T$. Necessary conditions for optimality as derived in Pontryagin's principle of minimum are then defined as

$$\alpha = \arg \min_{\alpha \in [0,\infty)} H(\boldsymbol{x}, \alpha, \boldsymbol{\lambda}), \tag{7a}$$

$$\dot{\boldsymbol{x}} = \frac{\partial H}{\partial \boldsymbol{\lambda}}, \quad \boldsymbol{x}(0) = \boldsymbol{x}_0, \quad \boldsymbol{x}(t_\mathrm{f}) = \boldsymbol{x}_\mathrm{f}, \tag{7b}$$

$$\dot{\boldsymbol{\lambda}} = -\frac{\partial H}{\partial \boldsymbol{x}}. \tag{7c}$$

Since Hamiltonian is linear in α, its minimum will be attained with α on its boundaries as

$$\alpha = \begin{cases} 0 & \text{if } H_\alpha > 0, \\ \infty & \text{if } H_\alpha < 0. \end{cases} \tag{8}$$

If $H_\alpha = 0$, the Hamiltonian is singular and does not depend on α. In this case, it may be possible to construct switching surface $S(\boldsymbol{x}) = 0$ corresponding to singular arc that depends on state variables only [8], [17]. We use the fact that the condition $H_\alpha = 0$ implies that time derivatives of H_α are equal to zero as well. We will make use of the following equations

$$H_\alpha(\boldsymbol{x}, \boldsymbol{\lambda}) = 0, \tag{9a}$$

$$\dot{H}_\alpha(\boldsymbol{x}, \boldsymbol{\lambda}) = 0, \tag{9b}$$

to eliminate the adjoint variables $\boldsymbol{\lambda}$.

IV. RESULTS

Based on the statements of previous section, we eliminated adjoint variables from conditions (9) by simple algebraic manipulations. The switching surface was then found in the following form

$$S(c_1, c_2) = w_T\left(q + c_1\frac{\partial q}{\partial c_1} + c_2\frac{\partial q}{\partial c_2}\right) + w_D q^2 = 0. \quad (10)$$

Singular control can then be found by additional differentiation of this condition with respect to time which gives

$$\dot{S}(c_1, c_2) = \frac{\partial S}{\partial c_1}\dot{c}_1 + \frac{\partial S}{\partial c_2}\dot{c}_2 = 0. \quad (11)$$

By using the process differential equations, this condition results in an expression for control when switching condition (defined by Eq. (10)) is active and it takes the form

$$\alpha(t) = \frac{\frac{\partial S}{\partial c_1}c_1}{\frac{\partial S}{\partial c_1}c_1 + \frac{\partial S}{\partial c_2}c_2}. \quad (12)$$

Once the weights w_T and w_D are fixed (i.e. closed form of the switching condition is known), the optimal operation can be stated as follows:

1) In the first step, we use pure dilution ($\alpha = \infty$) or pure ultrafiltration ($\alpha = 0$) until the optimal surface is reached $S(c_1, c_2) = 0$.
2) The second step is characterized as diafiltration on singular surface (10). Thus we use singular control (12).
3) The last step is either pure dilution ($\alpha = \infty$) or pure ultrafiltration ($\alpha = 0$) until the final concentrations are reached.

Optimal duration of the operation and the optimal water consumption are then found using respective time duration (Δt) and water consumption of these steps by

$$t_{\mathrm{f}}^* = \Delta t_1 + \Delta t_2 + \Delta t_3, \quad (13)$$

$$V_{\mathrm{w}}^* = V_{\mathrm{w},1} + V_{\mathrm{w},2} + V_{\mathrm{w},3}. \quad (14)$$

Any of the three steps can be missing from the optimal operation. This depends on particular process initial and final conditions. For instance, there might not exist a three-step operation which fulfill final conditions on concentrations. In such case, the second step is usually skipped from optimal operation and optimal control is saturated on constraints.

We note that the necessary conditions for optimality only suggest that the three steps can form a part of the optimal solution. Their number and sequence can only be deduced based on numerical simulations and insight in process operation.

V. CASE STUDIES

In this section, we show how to achieve the optimal operation for two cases studies of batch UF/DF processes. The first one is based on limiting flux theory a represents one of the most commonly used models. The second one was determined experimentally and describes separation of lactose from milk proteins. We will compare the proposed optimal operation with traditionally used control approaches.

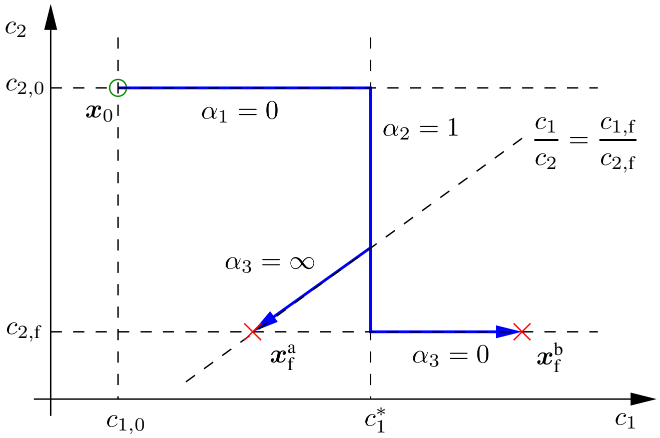

Fig. 3. Optimal operations of diafiltration with limiting flux model in concentration diagram with one initial point (○) and two endpoints (×).

A. Ultrafiltration/Diafiltration at Limiting Flux

We consider membrane plant which operates under limiting flux which is a classical modeling assumption used in membrane process engineering [1]. The permeate flow is given by

$$q(c_1) = kA \ln\frac{c_{\mathrm{lim}}}{c_1}, \quad (15)$$

where k is mass transfer coefficient, A represents membrane area, and c_{lim} stands for limiting concentration of macrosolute. The switching curve is constant it this case and can be derived from (10). The limiting cases are

- minimum time problem

$$c_1^* = \frac{c_{\mathrm{lim}}}{\mathrm{e}}, \quad (16)$$

- minimum diluant problem

$$c_1^* = c_{\mathrm{lim}}, \quad (17)$$

which agrees with the findings in [12]. Singular control is defined using Eq. (12) as constant-volume diafiltration, $\alpha = 1$.

We can now define the optimal operation in general. This is illustrated in Fig. 3. We consider an initial point $\boldsymbol{x}_0$ with concentrations $[c_{1,0}, c_{2,0}]$ where $c_{1,0} < c_1^*$ that is a usual case for practical applications. Once the concentration c_1^* is identified from (10), we distinguish two possible endpoints

a) $c_{1,\mathrm{f}} < c_1^*$ (denoted as $\boldsymbol{x}_{\mathrm{f}}^{\mathrm{a}}$ in Fig. 3)
b) $c_{1,\mathrm{f}} > c_1^*$ (denoted as $\boldsymbol{x}_{\mathrm{f}}^{\mathrm{b}}$ in Fig. 3)

with final concentrations which, by the problem definition, satisfy $c_{1,\mathrm{f}} > c_{1,0}$ and $c_{2,\mathrm{f}} < c_{2,0}$.

The optimal operation depends on initial and final conditions and on singular concentration c_1^* which is itself dependent on the choice of the coefficients w_T and w_D in (10). The optimal control structure (sequence of control actions) is either $\alpha = \{0, 1, \infty\}$ or $\alpha = \{0, 1, 0\}$.

An exception from this control structure can occur in the case of $c_1^* \geq c_{2,0}\, c_{1,\mathrm{f}}/c_{1,0}$ which means that the CVD step will be skipped from optimal operation and resulting optimal control structure will be $\alpha = \{0, \infty\}$.

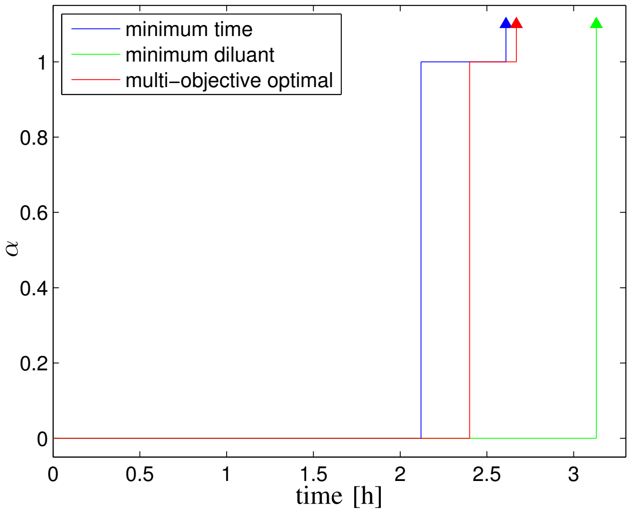

Fig. 4. Optimal control of diafiltration with limiting flux model for minimum time, minimum diluant, and multi-objective optimal operations.

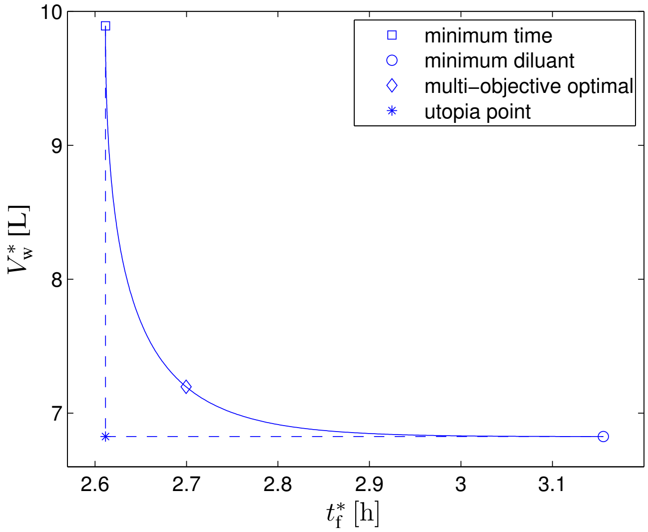

Fig. 5. Pareto front for UF/DF under limiting flux conditions.

TABLE I

MULTI-OBJECTIVE OPTIMAL OPERATION OF UF/DF UNDER LIMITING FLUX CONDITIONS COMPARED WITH MINIMUM TIME, MINIMUM DILUANT, AND TRADITIONALLY USED OPERATIONS.

UF/DF operation	t_{f} [h]	V_{w} [L]	δ^*
multi-objective optimal	2.67	7.2	0%
minimum time	2.61	9.9	10.6%
minimum diluant	3.13	6.8	9.9%
C-CVD (C-VVD)	2.62	11.5	18.0%
VVD	3.06	44.8	178.2%

We demonstrate the optimal operation procedure on the case when $c_{\mathrm{lim}} = 319\,\mathrm{mol/m^3}$, $k = 4.79 \times 10^{-6}\,\mathrm{m/s}$, and $A = 1\,\mathrm{m^2}$. The goal is to process 100 L of solution from initial point $[c_{1,0}, c_{2,0}] = [10\,\mathrm{mol/m^3}, 31.5\,\mathrm{mol/m^3}]$ to final point $[c_{1,\mathrm{f}}, c_{2,\mathrm{f}}] = [100\,\mathrm{mol/m^3}, 10\,\mathrm{mol/m^3}]$. Taking into account these parameters and conditions and using previously described general analysis, structure of multi-objective optimal operation of this process is either $\alpha = \{0, 1, \infty\}$ or $\alpha = \{0, \infty\}$ depending on the choice of coefficients w_T and w_D. Concretely, optimal operation is defined as $\alpha = \{0, 1, \infty\}$ with singular concentration which can be obtained from Eq. (10) for high values of ratio w_T/w_D. Decreasing this ratio of weighting coefficients maintains the same control structure until a switch in this control structure occurs when $c_1^* \geq c_{2,0}\, c_{1,\mathrm{f}}/c_{1,0} = 315\,\mathrm{mol\,m^{-3}}$ and optimal operation is $\alpha = \{0, \infty\}$.

Fig. 4 shows the optimal control for minimum time, minimum diluant, and multi-objective cases, respectively. We observe that in the minimum time case the CVD step ($\alpha = 1$) starts earlier and requires less time to reach the final point than in the multi-objective case. The minimum diluant operation skips the CVD step and the overall control strategy requires only two steps $\alpha = \{0, \infty\}$ to reach the final point. We assume that the last step (denoted by arrow) in all three cases takes no time.

Fig. 5 shows Pareto optimal values for individual parts of the objective function. This was obtained by changing coefficients w_T and w_D and by evaluating respective multi-objective optimal operation. We can notice the limiting cases minimum time ($w_T > 0$, $w_D = 0$, denoted by square) and minimum diluant ($w_T = 0, w_D > 0$, denoted by circle). The objective function contains contradictory terms, i.e. the amount of utilized water can only be reduced at the expense of increased processing time and vice versa. If these objectives were non-conflicting, optimal point would lie at the intersection of minimal values of t_{f}^* and V_{w}, an utopia point depicted in Fig. 5. For multi-objective optimization, we choose coefficients $w_T = 0.0567$ €/h and $w_D = 10$ €/m^3 and the Pareto optimal point is denoted for this choice as diamond in Fig. 5.

Table I shows comparisons of final time and amount of added water for several control scenarios. We also compare optimality loss δ^* of minimum time, minimum diluant and traditional control strategies with respect to multi-objective optimum. Both minimum time and minimum diluant possess optimality loss around 10%. There is 18% loss of optimality of C-CVD which according to [7] collides in this case with C-VVD strategy. Traditional VVD is the most optimality lossy operation which is in agreement with expectations.

B. Separation of Lactose from Proteins

We consider a process described in [16] where lactose is separated from milk proteins. The permeate flow was determined experimentally and can be written as follows

$$q(c_1, c_2) = b_0 + b_1 \ln c_1 + b_2 \ln c_2 \quad (18)$$

$$= 63.42 - 12.439 \ln c_1 - 7.836 \ln c_2, \quad (19)$$

where c_1 represents the concentration of proteins and c_2 represents the concentration of lactose. We will assume that this model is valid for the whole range of concentrations in question.

TABLE II

MULTI-OBJECTIVE OPTIMAL OPERATION OF UF/DF FOR SEPARATION OF A LACTOSE FROM PROTEINS COMPARED WITH MINIMUM TIME, MINIMUM DILUANT, AND TRADITIONALLY USED OPERATIONS.

UF/DF operation	t_f [h]	V_w [dL]	δ^*
multi-objective optimal	4.71	36.6	0%
minimum time	4.49	48.6	9.2%
minimum diluant	8.91	32.8	52.3%
C-CVD (C-VVD)	4.74	78.5	42.8%
VVD	5.38	135.5	109.1%

The optimal concentration curve is dependent on both concentrations

$$S(c_1, c_2) = w_T(b_1 + b_2 + q) + w_D q^2 = 0. \tag{20}$$

According to Eq. (12) we calculate the control on optimal surface

$$\alpha(t) = \alpha_s = \frac{b_1}{b_1 + b_2} = 0.61. \tag{21}$$

Our goal is to drive the concentrations from initial point $[c_{1,0}, c_{2,0}] = [3.3\,\mathrm{g/dL}, 5.5\,\mathrm{g/dL}]$ to final point $[c_{1,\mathrm{f}}, c_{2,\mathrm{f}}] = [9.04\,\mathrm{g/dL}, 0.64\,\mathrm{g/dL}]$ for $100\,\mathrm{dL}$ of solution. Once the coefficients w_T and w_D are fixed, the optimal operation uses three step strategy:

1) Starting at initial point (green circle) and concentrating with $\alpha = 0$ until the optimal surface is reached.
2) Staying on optimal surface with constant control α_s (21) (i.e. VVD step).
3) Pure dilution step until the final point is reached (red cross).

This is illustrated in Figs. 6, 7 for multi-objective optimal ($w_T = 0.0132$ €/h, $w_D = 0.001$ €/dL), minimum time ($w_T > 0, w_D = 0$), and minimum diluant ($w_T = 0, w_D > 0$) operations. All three operations require three step control strategy $\alpha = \{0, \alpha_s, \infty\}$.

Note that in the minimum diluant case the singular surface is reached at condition $q(c_1, c_2) = 0$ which theoretically gives infinite processing time. Therefore, we define a small positive precision ε for practically optimal minimum diluant operation with the cost $V_\mathrm{w}^*(1+\varepsilon)$. A value of $\varepsilon = 0.0025$ has been chosen. Such a change of operation is almost negligible with respect to the water consumption but has a large impact on the duration of optimal operation – it is reduced to $8.91\,\mathrm{h}$.

We can observe this *practical* minimum diluant problem solution in Fig. 8 where Pareto optimal values (Pareto front) are plotted. These were again found by changing the ratio w_T/w_D. The respective optimal operation can be found for each choice of these coefficients where we can use the fact that transmembrane flowrate is constant during the middle VVD step, as predicted by Eq. (20). In Fig. 8, we denoted respective points that signify multi-objective optimal, minimum time, and utopia optimal points.

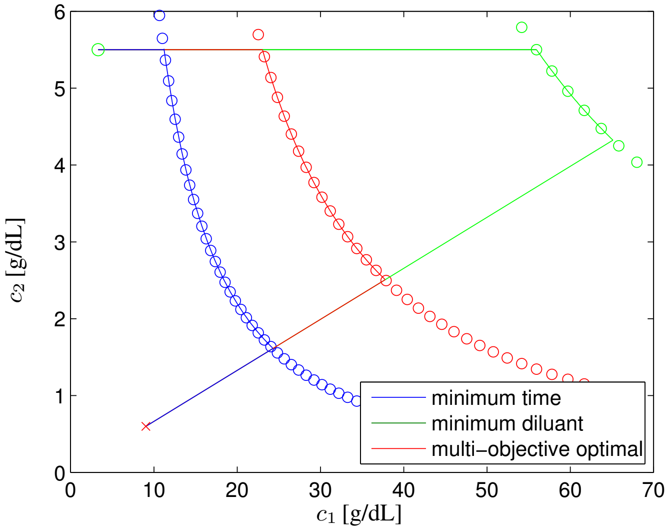

Fig. 6. Concentration state diagram for multi-objective optimal, minimum time, and minimum diluant strategies for separation of lactose from proteins. Singular surfaces for particular operations are denoted by circles.

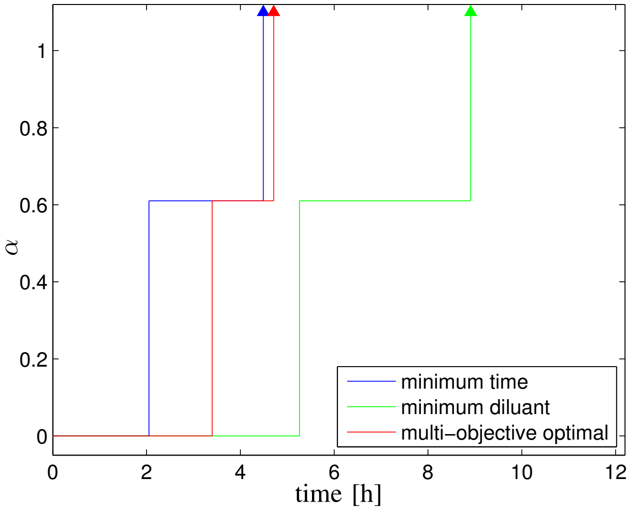

Fig. 7. Optimal control for multi-objective optimal, minimum time, and minimum diluant strategies for separation of lactose from proteins.

Tab. II shows values of processing times and of water consumption for multi-objective optimal, minimum time, minimum diluant, and traditional control approaches. Multi-objective optimal operation takes 4.71 hours of processing time and consumes 36.6 dL of water. Minimum time operation is, similarly to previous case, suboptimal by 9.2% in comparison with multi-objective optimal one. Both minimum diluant and C-CVD (which again collides with C-VVD operation) approaches result in more than 40% loss on optimality. When we compare VVD and C-CVD approaches, we notice that by C-CVD operation we reached the desired final state(s) in less time than by VVD and also the amount of added water was less then by VVD. This behavior was expected since C-CVD operation possesses more degrees of freedom.

VI. CONCLUSIONS

In this paper, we formulated multi-objective optimal control problem of general ultrafiltration/diafiltration process operation. The multi-criterion objective stands for the com-

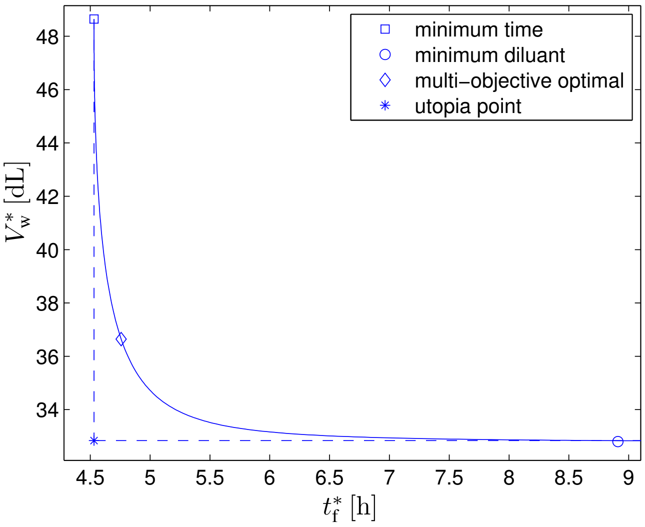

Fig. 8. Pareto front for separation of lactose from proteins.

bination of minimum time and minimum diluant problems, traditionally considered membrane filtration problems.

We derived conditions for optimality using Pontryagin's minimum principle. Then, we showed how these conditions can be used to determine multi-objective optimal operation of UF/DF processes. Although the conditions are only necessary, simulations and numerical dynamic optimization confirmed the optimal operation. This was demonstrated on two case studies.

The optimal operation usually takes three steps, except some cases which were discussed. The first and the last step is concentration or pure dilution. In the middle (second) step, various control strategies can occur potentially.

Our future work will consider more complex case studies (e.g. imperfect membrane rejection of macro-solutes), experimental verification in laboratory conditions, and robust control implementation capable to deal with model errors and plant uncertainties.

VII. ACKNOWLEDGMENTS

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