Development of a multi-objective coagulation system for long-term fouling control in dead-end ultrafiltration

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\textbf{A B S T R A C T}

In this paper, a multi-objective control system has been developed and experimentally tested. The multi-objective control system can be effectively used to control short-term fouling as well as long-term fouling. In an earlier study it was found that coagulant dosing in ultrafiltration can be used effectively to control the short-term fouling resistance during several sequential filtration cycles. To control long-term resistance increase during sequential chemical cleaning cycles, usually, in open-loop setting of the cleaning frequency or variables which influence the cleaning efficiency, such as, the cleaning time and the chemical composition are adjusted. Additional introductory experiments showed that changes in the coagulant dosing have a more pronounced effect on long-term fouling than changes in the usual applied variables. For this reason, it was decided to develop a closed-loop multi-objective controller where the coagulant dosing is used as the manipulated variable to accomplish both control objectives namely the fouling resistance over multiple filtration cycles (the short-term objective), as well as the irreversible fouling resistance over multiple chemical cleaning cycles (long-term objective). However, the controller is too slow to deal with temperature changes influencing the effectiveness of the coagulant dosing. To handle these influences a kind of gain scheduler should be included in the control algorithm.

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1. Introduction

Ultrafiltration (UF) is increasingly used as a technology for surface water purification. UF membranes have a high selectivity and became economically attractive during the last 15 years. However, membrane performance is influenced by fouling. For this reason frequent cleaning of the membrane is required. In the short term the membrane is cleaned by means of backwashing and in the long term the membrane is cleaned with cleaning chemicals. Although backwashing and chemical cleaning are useful methods to remove-fouling, the execution of such procedures is often based on rules of thumb and/or pilot plant studies.

In recent studies, attempts were made to model and validate the accumulation and removal of fouling during filtration and backwashing [1–7].

In our work, a black box modeling approach is chosen, as surface water composition is complex [13–15] and physico-chemical effects are difficult to describe. Black box models have excellent predictive properties and contain variables that can be used to control the process. Suitable models are required to calculate optimal production settings and/or trajectories. Subsequently, the optimal settings/trajectories should be realized by means of process controllers.

The authors have devised a hierarchical structure, representing all time- and production stages of an UF process for the purification of surface water. In Fig. 1 the hierarchical structure is depicted. The structure consists of three layers. For each layer, models and optimization routines were formulated.

In the lowest layer, short-term operations were modeled and optimized, such as filtration-, backwash- [8] and chemical cleaning settings [9,10]. In the middle layer, optimization over multiple filtration- and cleaning cycles was done [11], while in the upper layer the overall operational costs were balanced with the investment costs [12], on the basis of a membrane lifetime model. Optimization lead to optimal profiles for production settings, and these settings form the basis for the formulation of a control policy.

Optimal control of fouling in membrane filtration is not reported extensively in the literature [16,17]. In reference to the hierarchical optimization structure mentioned earlier, a coagulant controller...
was reported in [18], where the coagulant dosing – as a surface water pre-treatment step – was successfully employed in a feedback fouling control system. The set point trajectory of the controller is determined by means of dynamic optimization. This control system showed to be useful in controlling fouling over several filtration cycles. We note that in this context, short-term control is defined as control over several filtration cycles.

The objective of this paper is to extend the current inline control system, in which irreversible fouling in over several cleaning cycles is controlled by means of chemical cleaning. In this context long-term control is defined as control over several cleaning cycles. It is important to control fouling in the long-term, as the accumulation of irreversible fouling influences membrane lifetime in a negative way [19]. Irreversible fouling is in this context defined as fouling that cannot be removed by means of backwashing.

The irreversible membrane fouling resistance, accumulating in the long-term, may be controlled by adaptation of the open-loop settings for chemical cleaning (frequency, flushing, soaking, cleaning flux, cleaning agent concentration) using a feedback controller. The chemical cleaning model, in its current form is able to optimize such variables. However, experiments showed that the overall chemical cleaning effectiveness is strongly influenced by the coagulant dosing strategy of the preceded filtration sequence.

For this reason the chemical cleaning model should be extended with the coagulant dosing as additional variable, in order to determine dynamic control profiles. In contrast to the short-term control
strategy, where optimal trajectories of the manipulated variables are realized, we circumvented the shortcoming in the model by the formulation a long-term control objective that is determined by an optimal steady irreversible fouling level. The steady irreversible fouling level is determined by means of optimization at the membrane lifetime level.

Now the short-term, as well as long-term control objectives have to be realized by means of the coagulant dosing, an extended control system is developed, where the coagulant concentration is used, both, as manipulated variable to control the short-term fouling resistance as well as the long-term irreversible fouling resistance. It is noted that controlling two objectives with only one variable limits the operating region. However, implementation of this multi-objective control system shows to be effective for a pilot-plant processing surface water.

Two other options for closed-loop control worth mentioning are: (1) a cascade control system where the resistance profile of a filtration sequence is adapted to the achieved chemical cleaning effectiveness, or (2) a feedback control system where the chemical cleaning frequency (duration of the filtration sequence) is adapted to the achieved chemical cleaning effectiveness.

2. Materials and methods

The experiments were performed on a pilot plant scale filtration unit which is shown in Figs. 2 and 3. Two Norit-Xiga UFC SXL-225 FSFC modules with a membrane surface of 40 m² each and a cut-off of approximately 200 kDa were used. These modules consist of hollow fiber porous PES/PVP membranes with an internal diameter 0.8 mm and an effective length of approximately 1.5 m. The internal fiber volume is approximately 16.1, the additional dead volume of the system is estimated at 8.1.

Surface water taken from the Twente Canal was used as feed. This water contains, amongst others fatty acids, polysaccharides, proteins, high molecular weight organic structures and clay. Most of these substances are naturally present in surface water and originate from, for example, plant material. In addition, some municipal, industrial and agricultural contamination can be found. The feedwater was pre-filtered (200 μm) to prevent too large particles from entering the system. The feedwater was buffered in a continuously refreshed and well-stirred feed tank.

Filtration sequences were preceded by a chemical cleaning procedure. First a caustic cleaning with sodium hydroxide (pH 11) and sodium hypochlorite (100 ppm) was performed, secondly an acidic cleaning with hydrochloric acid (pH 2) was performed. During filtration, coagulant was dosed to the feed water (Quadrafloc PUS, ViVoChem BV). The coagulant dosing was controlled by flow ratio control on a dosing pump (FrC in Fig. 2). The mixing point is just before the filtration pump.

In Fig. 2, the pressure sensors are indicated by PT, the temperature sensors by TT and the flow transmitters by FT. Based on these measurements, the values of the viscosity and resistance are calculated by the control software. The initial value of the filtration resistance is obtained by calculating the average resistance during the first 50 s of the filtration phase. Subsequently, the adaptation of the coagulant dosing is calculated and the dosing setpoint is adapted.

3. Closed-loop control

The design of a control system consists of the following steps: definition of control objectives, selection of manipulated and measured variables, selection of the control configuration and finally
controller design. An introduction to control systems can be found in Stephanopoulos [20].

3.1. Control objectives

As discussed in the introduction, there are two types of control objectives that should be realized within ultrafiltration of surface water. The short-term control objective, where fouling levels are controlled during the filtration cycle and the long-term control objective, where fouling levels are controlled over several chemical cleaning cycles.

Fig. 4 shows how reversible and irreversible resistance evolves over multiple filtration and chemical cleaning cycles. Where \( R_{OF,D}(n_F) \) is the desired resistance at the end of a filtration sequence and where \( R_{OC,D}(n_C) \) is the desired resistance at the end of a chemical cleaning phase, \( n_F \) and \( n_C \) denote the filtration cycle number and the chemical cleaning cycle number, respectively.

3.2. Manipulated and controlled variables

To realize the control objectives, they should first be quantified. The resistance is a good measure for the amount of fouling present in the system and will serve as the controlled variable. The resistance is the sum of the membrane resistance \( R_M \) and a fouling resistance \( R_f \). Darcy’s law relates the resistance to the flux \( J \), the trans-membrane pressure \( \Delta P \) and the viscosity \( \mu \):

\[
R_M + R_f = \frac{\Delta P}{\mu J}
\] (1)

The objective of the coagulant controller (short term) is to track a trajectory of desired initial filtration resistances. The objective of the chemical cleaning controller (long-term) is to keep the resistance after chemical cleaning at a certain value.

4. Initial control experiments

In principle we want to realize the short-term and long-term objectives by two independent feedback controllers. Previous work showed that the coagulant dosing influences the cleaning efficiency of the backwash [18]; the short-term objective can be realized using the coagulant dosing \( C_F \).

The long-term objective can be realized by manipulating the chemical cleaning settings, such as the cleaning agent concentration, cleaning time or cleaning frequency. The chemical cleaning time is chosen as manipulated variable because it is most easily changed in terms of capacities of the pilot setup.

A control scheme where the cleaning time \( t_C \) is applied as manipulated variable to realize the long-term objective and where the coagulant dosing \( C_F \) is used to realize the short-term objective is illustrated in Fig. 5.

It should be mentioned that there could be interaction between the two controllers; the coagulant dosing applied during the filtration sequence may influence the effect that the chemical cleaning time may have on the cleaning sequence.

In the two sections below, we will introduce the short-term and long-term controllers and evaluate their behaviour

4.1. Short-term control

The short-term controller that tracks a desired initial filtration resistance trajectory was developed in earlier work [18]. An empirical relationship to describe the resistance trajectory can be given as

\[
R_{OF,D}(V_F) = R_{OC}(n_C) + \alpha_V V_F + R_i (1 - e^{-V_F/V_{eq}})
\] (2)

where \( R_{OF,D}(V_F) \) is the desired resistance trajectory as function of the specific volume produced \( (V_F) \). \( R_{OC}(n_C) \) is the resistance after the \( n_C \)-th chemical cleaning phase, \( \alpha_V \) is the final slope, \( R_i \) is the gain of exponential rise and \( V_{eq} \) is the characteristic volume. A deviation from the desired initial filtration resistance trajectory curve indicates that the final value is most likely going to deviate from the desired final value as well. For this reason, a difference between the desired initial filtration resistance and the measured initial filtration resistance should result in an adaptation of the coagulant dosing. The difference between the desired and realized initial filtration resistance is the control error:

\[
\epsilon_F(n_F) = R_{OF}(n_F) - R_{OF,D}(V_F(n_F))
\] (3)

with \( n_F \) the filtration cycle number, \( R_{OF,D}(V_F(n_F)) \) the desired initial filtration resistance trajectory and \( R_{OF}(n_F) \) the measured initial resistance.

The algorithm that determines how the information obtained from the process (the control error) is used to adapt the manipulated variable is called the controller. Because a trajectory for the initial filtration resistance is tracked, the coagulant dosing \( C_F \) is adapted once every filtration, at the moment at which the initial resistance is estimated.

When the manipulated variable is constrained, it is more convenient to use the velocity form, which expresses the change of the
Fig. 6. Sequence of chemical cleaning cycles.

In the following eight chemical cleaning cycles (indicated by II), the coagulant controller settings were adjusted. The realized resistance is now much closer to the desired resistance profiles. But the effect of chemical cleaning is now much stronger, and the resistance at the beginning of a chemical cleaning cycle is below its setpoint. The chemical cleaning controller becomes also out of control and sets the cleaning time to its minimum value $t_C = 5$ min.

In the last two runs (indicated by III) the desired fouling profile is chosen more steeply, to see if increased fouling will lead to an increased cleaning time. However, the chemical cleaning controller keeps the cleaning time at its minimum value of 5 minutes, as the coagulant dosing is still high enough to enable the chemical cleaning to be fully effective. The coagulant controller settings of Eq. (2) are shown in Table 1.

The experimental results of Fig. 6 show that the chemical cleaning effectiveness is not strongly influenced by the cleaning time, whereas the effectiveness appears to be sensitive to the coagulant dosing. In Fig. 7 the chemical cleaning effectiveness is plotted as a function of the applied coagulant dosing for both cleaning times that the cleaning controller calculated (5 and 100 min). Chemical cleaning effectiveness is in this context defined as

$$\eta = \frac{R_{OF,D}(n_C) - R_{OF,D}(n_C - 1)}{R_{OF,D}(n_C) - R_{OF,D}(n_C - 1)}$$

The figure clearly shows that cleaning time hardly influences the cleaning effectiveness, but that the applied coagulant dosing has a significant impact.

### Table 1
Settings for the coagulant controller.

<table>
<thead>
<tr>
<th></th>
<th>I</th>
<th>II</th>
<th>III</th>
</tr>
</thead>
<tbody>
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<td>$R_c$ (m$^{-1}$)</td>
<td>$7 \times 10^{11}$</td>
<td>0</td>
<td>$3 \times 10^{11}$</td>
</tr>
<tr>
<td>$R_f$ (m$^{-1}$)</td>
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<td>$1 \times 10^{12}$</td>
<td>$1 \times 10^{12}$</td>
</tr>
<tr>
<td>$V_C$ (m)</td>
<td>0.1</td>
<td>-</td>
<td>0.1</td>
</tr>
</tbody>
</table>

In the following eight chemical cleaning cycles (indicated by II), the coagulant controller settings were adjusted. The realized resistance is now much closer to the desired resistance profiles. But the effect of chemical cleaning is now much stronger, and the resistance at the beginning of a chemical cleaning cycle is below its setpoint. The chemical cleaning controller becomes also out of control and sets the cleaning time to its minimum value $t_C = 5$ min.

In the last two runs (indicated by III) the desired fouling profile is chosen more steeply, to see if increased fouling will lead to an increased cleaning time. However, the chemical cleaning controller keeps the cleaning time at its minimum value of 5 minutes, as the coagulant dosing is still high enough to enable the chemical cleaning to be fully effective. The coagulant controller settings of Eq. (2) are shown in Table 1.

4.2. **Long-term control**

The objective of the long-term controller is to control the irreversible filtration resistance after a chemical cleaning phase. The control error can be defined as

$$\varepsilon_C(n_C) = R_{OC}(n_C) - R_{OC,D}(n_C)$$

where $R_{OC}(n_C)$ is the measured resistance after chemical cleaning and $R_{OC,D}(n_C)$ is the desired resistance after chemical cleaning.

A discrete PI controller in velocity form is given as

$$t_C(n_C + 1) = t_C(n_C) + K_C \left( 1 + \frac{1}{n_{CI}} \right) \varepsilon_C(n_C) - \varepsilon_C(n_C - 1)$$

where $t_C$ is the cleaning time, $K_C$ is the proportional controller gain and $n_{CI}$ is the integration interval. The bounds can be given as

$$t_{C,lb} < t_C(n_C) < t_{C,ub}$$

Fig. 6 shows the results. In the upper figure the desired and realized resistances are shown, and in the lower plot the coagulant dosing is shown. During the first 15 chemical cleaning cycles (indicated by I), the settings of the coagulant controller were chosen incorrectly, leading to a fast increase of the resistance. The resistance increased to its maximum allowed value of $3 \times 10^{12}$ (1/m), subsequently a chemical cleaning procedure was performed. The chemical cleaning controller now calculates a new cleaning time, but as the resistance at the beginning of a chemical cleaning cycle is far above its set point (the dashed line), the chemical cleaning controller becomes out of control and sets the cleaning time to its maximum value $t_C = 100$ min.
5. Multi-objective control

This observation lead to the concept of multi-objective control, where the coagulant dosing is used, both to realize the short-term objective as well as the long-term objective.

5.1. Controller configuration

Usually, the control of the long-term resistance confines itself to the open-loop (manual) adjustment of the chemical cleaning settings of the membrane unit. Our secondary aim is to achieve a closed-loop control of a desired resistance trajectory ranging the entire lifetime of a membrane unit.

The results from the previous chapter indicate that both objectives are strongly influenced by the coagulant concentration. This observation leads to the concept of multi-objective control where the coagulant dosing is used to achieve both control objectives:

(1) the short-term objective, in which the initial resistance trajectory during the sequential filtration cycles $R_{0F,D}(V_F)$ is controlled and,

(2) the long-term objective, in which the initial irreversible resistance of the sequential cleaning cycles $R_{0C,D}(n_C)$ is controlled.

Fig. 4 shows the trajectories of the filtration resistance of the short-term set point $R_{0F,D}(V_F)$ and the long-term set point $R_{0C,D}(n_C)$. Note that according to 2, the desired short-term resistance is defined relatively to the last realized long-term resistance.

Therefore, both set-up trajectories are uncoupled and can be aimed for independently. The optimal course of the desired short-term trajectory of $R_{0F,D}(V_F)$ is given before in 2.

The trajectory of the desired long-term trajectory of $R_{0C,D}(n_C)$ is, for the time-being, chosen arbitrarily. In a later stage this can be derived by dynamic optimization over the life-time of the membrane.

However, by choosing one manipulated variable for both objectives, it is physically impossible to meet both trajectories independently.

It is important yet to do not exceed one of them (see Fig. 4). Therefore, the largest necessary positive correction obtains priority.

In the chosen configuration, two controllers calculate a value for the coagulant dosing and the maximum of the two values is applied. This concept is illustrated in Fig. 8.

The first controller minimizes the control error formulated in Eq. (3), using Eq. (5) and the second controller minimizes the control
error formulated in Eq. (6). With the control error given in Eq. (6) the following control algorithm is used:

\[ C_F(n_C + 1) = C_F(n_C) + K_C \left( \frac{1}{n_C} \right) \varepsilon(n_C) - \varepsilon(n_C - 1) \]  

(10)

and the bounds can be given by

\[ 0 < C_F(n_C) < C_{F,ub} \]  

(11)

5.2. Results

Fig. 9 shows the results of the multi-objective controller. In the upper figure the desired an realized resistance are shown, in the middle plot the coagulant dosing is shown and in the lower plot the temperature evolution is shown.

In the first 20 chemical cleaning cycles (11 days), the setpoint for the irreversible resistance is set at 1.4 \times 10^{12} (1/m). The realized resistance after chemical cleaning, as indicated by the circles is below the desired resistance (dashed line). Also the realized resistance profiles during the filtration sequence (dots) are below the desired profiles (lines). The initial coagulant dosing was set at 1 ppm, which showed to be too high. The multi-objective controller adapts the coagulant dosing by decreasing its value steadily. The adaptation leads to better realized profiles for the resistance during the filtration sequence, but the irreversible fouling resistance after chemical cleaning is still below its setpoint.

After the first 20 chemical cleaning cycles, the setpoint for the irreversible fouling resistance is decreased to a more realistic value of 1.1 \times 10^{12} (1/m). Better results are now obtained for the filtration sequence fouling profile as well as for the irreversible fouling resistance after chemical cleaning.

The lower part of Fig. 9 shows the temperature of the feed water during the experiments. As temperature is increasing, the required coagulant dosing is decreasing. The required coagulant dosing is related to the temperature. Since adaptation at the chemical cleaning cycle level is slow, the controller is not able to adapt to changes in the temperature, which occur on the same timescale (day to week), as a result of the day–night cycle and weather effects.

5.3. Gain scheduling

The coagulant dosing is more effective at high temperatures. In Fig. 10 the filtration sequence average temperature is plotted versus the filtration sequence average coagulant concentration. The line in the figure represents an empirical approximation of the relationship between the temperature and the required coagulant dose. It is assumed that the relationship between the effective coagulant concentration and the temperature has an exponential form, given by

\[ C_T = C_{T,D} e^{-k_T(T-T_0)} \]  

(12)

For the data of Fig. 10 \( k = 0.23 \) and \( T_0 = 20 \degree C \).

This empirical relationship can be used to expand the control strategy with a gain scheduler, to correct the coagulant dosing for temperature changes. It should be noted that in this study the temperature correction by means of gain scheduling has not yet been implemented.

Implementation of a gain scheduler to cope with temperature changes that influence the effect of coagulant would improve a faster realization of the control objective.

6. Conclusions

Independent feedback controllers to realize short-term and long-term objectives in dead-end ultrafiltration are not suitable to control irreversible fouling. However, multi-objective control is suitable for controlling both the backwash efficiency and the chemical cleaning effectiveness. Due to the time interval at long-term control takes place, the long-term controller part of the multi-objective control cannot adapt to changes in the temperature. In order to cope with temperature changes a gain scheduler correction in the coagulant dosing is required.

Acknowledgements

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Appendix A. Nomenclature

- \( C_F \): coagulant concentration (ppm)
- \( C_{F,D} \): coagulant concentration (ppm)
- \( C_{F,lb} \): lower bound coagulant concentration (ppm)
- \( C_{F,ub} \): upper bound coagulant concentration (ppm)
- \( C_T \): effective coagulant concentration (ppm)
- \( J \): flux (m/s)
- \( K_C \): long-term controller gain (ppm m)
- \( K \): short-term controller gain (ppm m)
- \( k_T \): empirical constant (1/ppm)
- \( n_C \): number of chemical cleaning cycles
- \( n_F \): number of filtration cycles
- \( n_C,I \): long-term controller integration interval
- \( n_I \): short-term controller integration interval
- \( \Delta P \): trans-membrane pressure (Pa)
- \( R_{0,F} \): measured resistance at the beginning of a chemical cleaning cycle (1/m)
- \( R_{0,C,D} \): desired resistance at the beginning of a chemical cleaning cycle (1/m)
- \( R_{0,F,D} \): desired resistance at the end of a chemical cleaning cycle (1/m)
- \( R_F \): fouling resistance (1/m)
- \( R_M \): membrane resistance (1/m)
- \( R_I \): gain of exponential rise (1/m)
References


