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Optimal Design of Membrane Processes: a Problem of Choices Between Process Layout, Operating Conditions and Adopted Control System

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The development of membrane processes as a technology for environmental treatment applications and in particular for the purification of wastewater streams has significantly increased in the last decades. Fouling on membranes appears to be one of the main technical limit of this technology. This phenomenon causes the unavoidable deposition of particles on the membrane surface, building a resistive growing layer to permeability. Sensible fouling of the membrane leads to a significant reduction of the performances, a decrease of the operating life and, as a consequence, the increase of the operational costs due to the replacement or cleaning of the exhausted membrane modules. The presence of the fouling phenomena makes the proper design and control of membrane systems a difficult task.

Optimal design of the membrane processes will be here discussed. The procedure requires to determine the optimal process layout given the input data and target requirements. At the end, the required membrane area is calculated. This latter property is strictly dependant of the adopted operating conditions, most importantly by the adopted value of transmembrane pressure (TMP). Moreover, it depends if the value of TMP remain fixed as a function of time or is variable (as in case of fixed permeate flow rates). Therefore, the optimal design of the system may occur only if the adopted control strategy is defined a priori. As a consequence, design choices of the membrane process layout, operating condition and adopted control system are strictly dependant, and connections between these different aspects should not be neglected during the engineering and P&I development stage of membrane systems.

This paper will start from the theory of the boundary flux, in order to describe a novel design approach to membrane systems. Parallel to this, the development of an advanced control system, that allows to limit fouling formation during operation, is presented. The advanced control system relies on a suitable simulation software capable to predict the boundary flux, that changes the controller's set-points accordingly. Finally, the paper will merge all elements together, and report about the optimal design of membrane processes equipped with the advanced membrane process control system; validation of the proposed approach will be based on the use of a custom simulation model in ASPEN HYSYS and by experiments on lab scale.

1. Introduction

Membrane technologies have gained great importance in water and wastewater treatment applications due to its wide range of operation, ease to scale-up and high versatility. Membranes exhibits both high productivity and selectivity values towards pollutants, and therefore high efficiencies of water treatment.

Environmental hazardous effluents may be treated to an aqueous stream, reaching the requirements for a municipal sewer system discharge, superficial aquifer release or industrial reuse. As stated by Schroetter and Boskaya-Scroetter, in 2010 about 60 Mm^3 of wastewater are treated worldwide every day, and almost 50% by membrane processes such as microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). In the next 10 years, an annual growth of 10% of this capacity is foreseen, and in respect to this, most probably membrane technology will gain more and more in importance.

The main drawback of membranes appears to be membrane fouling, that requires to be avoided or at least inhibited. This phenomenon does not permit to assure the performances of membranes over a long period of time once it starts to form. Starting with the last decade, research and expertise is focused on understanding and avoiding membrane fouling, and covers modelling (Ortiz Jerez et al., 2008), reporting (Daufin et al., 2001), development of new membrane materials (Lu et al., 2002), suggestion of proper operating conditions (Stoller and Chianese, 2007), management and control of the process (Stoller, 2016), proper pretreatment processes Ochando Pulido et al., 2016) and cleaning procedures (Ochando-Pulido, 2015). The result of actual know-how is that fouling cannot be avoided at all and will exist to a different extend in every membrane operation. If the fouling formation is small, it might be neglected when compared to other phenomena, such as aging.

On contrary, is sensible fouling occurs, the phenomena must be considered and might represent a bottle neck to overcome, in order to guarantee a reliable separation process for long term operations. The latter case is most common in case of wastewater treatment.

In this case, for a given system, the input parameters are:

1. Capacity, that is the flow rate of water to be purified
2. Recovery, that is the amount of water to recover as purified stream from the initial volume
3. Selectivity, that is the amount of pollutants to be removed in order to obtain a suitable purified water stream

It is possible to observe that fouling do not enter as an initial input parameter, at least not directly. In the past, many times this aspect was completely neglected or, in best case, this problem was overcomed by over-design a forfeit or based on the experience of the process designers. Despite the adoption of these techniques, failures were not avoided.

Therefore, an additional input parameter must be introduced in the design of the process, that is:

4. Membrane longevity, which is a minimum number of operation cycles where membranes are capable to exhibit performances that guarantees the input targets 1 to 3.

In this insight, it appears mandatory to perform design checks on membrane fouling. Options to inhibit fouling are the modelling and suggestion of proper operating conditions: the boundary flux concept and its determination seems to be one of the best method to identify process conditions capable to limit the formation of irreversible fouling (Stoller and Ochando Pulido, 2015). On the other hand, reversible and semi-reversible fouling will always trigger, leading to a performance loss as a function of time between washing periods. This latter phenomenon has a great importance on the definition of the process capacity, since the minimum required capacity must be guaranteed at the end of operation.

Moreover, many times, boundary flux values are very low and not very attractive from an economic point of view. Therefore, proper pretreatment processes are mandatory: since operation is limited by the boundary flux values, one possible strategy may involve the sensible increase of its value. This can be performed by proper pretreatment of the feed stream (pretreatment tailoring).

Membrane processes may be controlled mainly by maintaining fixed a pressure (TMP) or a permeate flow rate value. The first case is of easy implementation, but the ever changing permeate flux values during operation makes this approach unattractive. The second case requires pressure adjustments in order to be accomplished, and represent the common control scheme for membrane processes. In this paper, the control system will rely on this latter control strategy (permeate flow rate set-point).

Management and control of the process appears to be key to reliability of this technology. Once the boundary flux value is determined and the relevant changes as a function of time are well defined, a control system should be implemented in order to operate the system correctly.

The development of advanced control systems for membrane processes based on the boundary flux concept and relevant equations were introduced in the framework of previous membrane technology works, and here only limited to those used and briefly summarized (Stoller and Ochando Pulido, 2015):

$$-dm/dt = \alpha \text{ if } J_p<J_b \tag{1}$$

$$J_p(t) = m(t)\ TMP \tag{2}$$

$$F_p(t) = A\ J_p(t) \tag{3}$$

where J_p is the permeate flux, TMP the transmembrane pressure, J_b the boundary flux, m is the membrane permeability, α the sub-boundary fouling rate index, $\Delta w\%$ the cleaning efficiency (and thus indication of membrane aging), Y the recovery value, F_p the permeate flow rate. All other relationships may be derived or integrated starting from these.

2. Process design and control in absence of fouling

In this first case study, fouling will be neglected or simply is neglectable. There are very few processes where this approximation may apply, mostly in case of critical flux based systems. As ease of an example, olive mill wash wastewater is one of those systems (Stoller and Chianese, 2007).

In this paper, typical input parameters in case of the treatment of olive mill wastewaters are assumed as in Table 1:

Table 1: Process design input parameters

Design parameter	Value
Capacity, F*	1 $m^3 h^{-1}$
Recovery, Y*	90%
Selectivity (on COD), R*	60%
Longevity, N*	1000 d

Washing is performed at the end of day, lasting a time T equal to 18h of operation.

On the other hand, the membrane choice will affect together with the feedstock, the performances of the system. The main constraint to consider in a proper membrane type identification process is selectivity. In our case, the Desal Osmonics DK - SW nanofiltration membrane is capable to achieve the desired purification result. Therefore, we will assume that this design parameter results fully accomplished.

Typical input values for the boundary flux equation set for this membrane are reported in Table 2, obtained adopting as a pretreatment ultrafiltration:

Table 2: Membrane properties

Membrane characteristics	Value
Pure water permeability	1,07 $l\ h^{-1} m^{-2} bar^{-1}$
Osmotic pressure	0 bar
Selectivity (on COD)	70%
Boundary flux	12,18 $l\ h^{-1} m^{-2}$
Boundary TMP	7 bar
α	130 10^{-4} $l\ h^{-2} m^{-2} bar^{-1}$
$\Delta w\%$	0,0005 % h^{-1}

Since no fouling will occur, the constraint on the desired longevity of the membranes is apparently useless. In this case, the value of α is assumed to be equal to 0. On the other hand, even with absence of fouling, membrane aging lowers the membrane performances as a function of time to a small extend, and this must be considered ($\Delta w\%$).

At this point, it is possible to perform the calculations for our (quite simple) example (referred as case study A) to evaluate the required membrane area A to fulfil the process targets, as reported in Table 3:

Table 3: Calculations for case study A

Step	Calculation	Equation	Result
1	No fouling		$\alpha = 0$
2	Permeate flux loss due to fouling	$-dm/dt = 0$	m = const
3	Permeate flux loss due to aging	$\Delta W\% = 1/100\ \Delta w\%\ T\ N^*$	$\Delta W\% = 0,09$
4	Project capacity due to flux losses	$F^{**} = F^*\ Y^*\ (1 + \Delta W\%)$	$F^{**} = 981\ l\ h^{-1}$
5	Determination of permeate flux	$J_p = J_b$	$J_p = 12,18\ l\ h^{-1}\ m^{-2}$
6	Determination of membrane area	$A = F^{**}\ J_p^{-1}$	$A = 80,54\ m^2$

As noticed, calculations are very easy and quickly performed. Once finished, many times an over-design coefficient of 10-15% was applied in order to be safe from process failures. But again, this approach was not sufficient in many cases where fouling triggers.

3. Process design and control in presence of fouling

In this case study, called “B”, the same process design input parameters and membrane as in the case study A will be used (Table 1 and Table 2).

In this case, α cannot be assumed equal to 0 and must be considered during calculations. In order to perform the calculations without a simulation model (although strongly suggested), two assumptions will be made:

- Since both α and $\Delta w\%$ will reduce the permeate flux values as a function of time, in order to guarantee the project targets, the suggested approach is to look at the last operation cycle, that is for N equal to 1000. If this latter cycle meets the requirements, all previous cycles automatically will reach them.
- The boundary flux value is assumed to be constant as a function of COD. This may appear to be a strong condition, but is as much true if the given value is referred to the typical end of operation COD value (and this is the case of the data reported in Table 2).

Again, we may perform the required calculations as reported in Table 4:

Table 4: Calculations for case study B

Step	Calculation	Equation	Result
1	Permeate flux loss due to fouling (last cycle)	$\Delta J_p = \alpha\ T\ TMP_b$	$\Delta J_p = 1,63\ l\ h^{-1}\ m^{-2}$
2	Permeate flux loss due to aging (all cycles)	$\Delta W\% = 1/100\ \Delta w\%\ T\ N^*$	$\Delta W\% = 0,09$
3	Project capacity due to flux losses (all cycles)	$F^{**} = F^*\ Y^*\ (1 + \Delta W\%)$	$F^{**} = 981\ l\ h^{-1}$
4	Determination of permeate flux (last cycle)	$J_p(T) = J_b$	$J_p(T) = 12,18\ l\ h^{-1}\ m^{-2}$
5	Determination of membrane area (last cycle)	$A = F^{**}\ (J_p^{-1}(T) - \Delta J_p)$	$A = 92,98\ m^2$

It can be noticed that the required membrane area is about 16% higher than that calculated in case study A, at the limit of the overdesign applied to the process a forfeit. Given that both processes are properly controlled, that is to work below boundary flux conditions all the time.

4. The control system

The previous calculations were possible since many parameters remain constant, the most important being α and $\Delta w\%$, respectively.

In a previous work it was shown that these parameters will change sensibly and irreversibly as soon as superboundary conditions were used (Stoller, 2011). Even operating 20 minutes the system in superboundary conditions will lead to irreversible fouling, and a consequent increase about 30% of these parameters.

A suitable control system capable to check operation in subboundary flux conditions is therefore mandatory and part of the overall high longevity strategy of the used membrane modules. Without this approach, membranes must be substituted more frequently, thus increasing the operating cost to such a level where the overall process results, form an economic point of view, not attractive and advantageous. This is especially true in wastewater treatment applications.

The proposed control system is here shortly reported, and consists in a simulation model capable to evaluate and predict the boundary flux value profiles as a function of time (and COD) implemented in Hysys (AspenTech).

In Figure 1, the scheme of the simulation model as shown in HYSYS GUI is pictured.

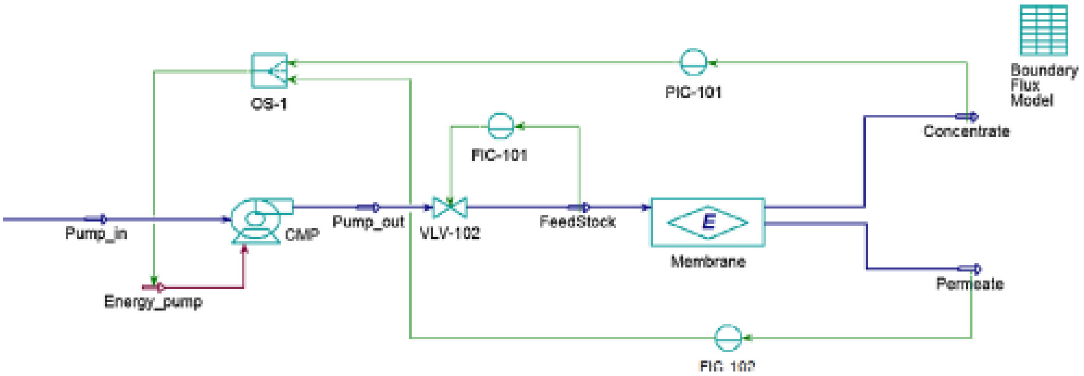

Figure 1: Simulation of the membrane process and the advanced control system

The advanced control system is capable to:

- Determine the operating parameters to reach end of operation in sub-boundary flux conditions given a desired permeate flow rate set-point by adopting given initial input values.
- Check the constancy of the input parameters values given initially to the model during operation. If confirmed, the control system will follow the set-point settings. If the check fails, the model will evaluate again the operating conditions, and if these are not compatible to the given set-point, override the setting to best possible performances.

For example, this happens in the case reported below. The missing operating time was about 8,5h, and the check on the model parameters (in particular on α) failed. The simulation model calculated a new boundary flux plot as a function of time, and estimated that a safe operation was possible only below 9,00 l h^{-1} m^{-2} and not at 12,18 l h^{-1} m^{-2} anymore.
Moreover, the simulation code calculated that in this case, an additional operating time of 3h was required to reach the desired Y* value: therefore, end of operation was shifted to 11,5h.
Without all these adjustments, it might be noticed that operating at previous boundary flux values or at adjusted ones without considering the additional amount of operating time, that is at 12,18 l h^{-1} m^{-2} (previous value) and 9,15 l h^{-1} m^{-2} (value at 8,5h), respectively, would lead to super-boundary flux conditions and, as a consequence, to severe irreversible performance loss of the process.

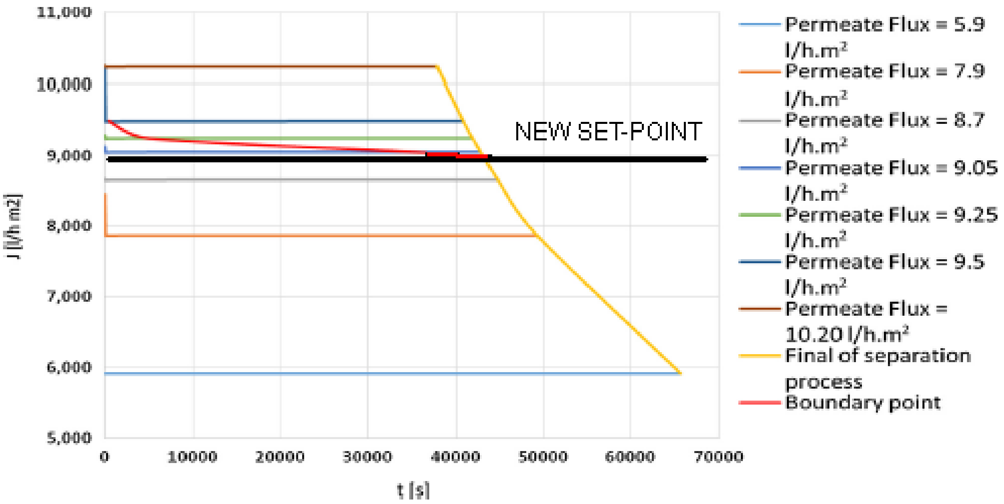

Figure 2: Determination of the new set-point after a failed check on the model parameters

5. Conclusions

In this paper, design concepts of membrane processes affected by fouling issues are given and highlighted.
First of all, proper design requires to know, beside boundary flux data, at least the value of α. In many works, this important data is missing, since this parameter will affect directly the required overdesign to overcome fouling constraints in the long run.
Finally, the importance of an advanced control system is mandatory, since the application of such a controller is the only way to guarantee the constancy of important values such as α and Δw%. The approach relies on a guarantee to never overcome boundary flux conditions, but wants also to optimize operating conditions as high as possible in order to use all the available membrane area. The control system should therefore by of inferring and predictive type, to be capable to determine by real time measurable parameters the actual situation and to calculate the outcome of the operation till end.
The developed control system in HYSYS appears to have all these properties and capabilities.

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