About the Development of Advanced Membrane Process Control Systems

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This paper focuses on the design and development of advanced control systems to use on either batch or continuous, new or existing membrane process plants.

In the last decade, membrane technologies resulted to be very appealing and shows positive market trends. One main drawback is membrane fouling, which affects productivity, selectivity and longevity of the process, which leads to both technical and economical failures; proper membrane process design and control is a difficult task to accomplish. This leads to overdesign the plant capacities by process engineers, making this technology less reliable and convenient.

Nowadays membrane processes are controlled by a constant permeate flow rate or constant applied operating pressure. These simple control strategy approaches are sufficient to operate the processes, but do not distinguish different fouling operating regions, and therefore do not avoid process failures due to fouling. Fouling may be described by the boundary flux theory in a convenient way, separating low-fouling operations from high-fouling ones.

The paper reports about the development of an advanced membrane process control system based on the boundary flux concept. The developed advanced control strategy by the use of a simulation software, capable to predict boundary flux values by measurement of some key parameters, provides suitable set-point values to the feedback controllers in order to work at or below the boundary flux. As a consequence, the membrane process is always operated far from irreversible fouling issues. The developed approach was then successfully validated by experiments on lab scale.

1. Introduction

In the last decade membrane technologies gain market and are used as stand-alone, integrated or substitutive processes. The technology is very appealing, both from technical and economic point of view, and has many advantages if compared to conventional technologies. On the other side, one main drawback is membrane fouling, which represents a main constraint holding membrane technologies away from a definitive maturation. Nowadays, proper membrane process design can be a difficult task to accomplish when fouling is present and must be considered. The designer normally should consider the project variables concerning productivity and selectivity and follow these targets; in the presence of fouling, additional parameters, in particular the longevity of the membrane modules and the constancy of the permeate fluxes as a function of time must be considered. Fouling leads to a reduction of the permeate flux rate and parallel to this, may lead to sensibly shorten the life time of membrane modules. The presence of fouling, and the consequent reduction of permeate fluxes as a function of time, forces the designer to over-design the membrane plant in order to guarantee sufficient operating autonomy to conduct the process for a certain period of time at or above the permeate project values (Saad, 2005). In most cases the over-design is performed a forfeit or by past experience of the designer, starting only from the knowledge of the permeate project value, without considering in detail the entity and nature of fouling. Some examples of existing overdesigned membrane plants are reported in...
One method to increase the reliability of a process is to adopt stable control systems. In the case of most membrane processes, this is performed by simple control strategies, that do not include knowledge and control of the system. Moreover, most control systems rely on the time control of washing procedures (Ruano et al., 2013), when fouling just triggered, or the control of anti-fouling procedures (Xu et al., 2014), such as the correct dosage of chemicals or air bubbles. Those systems do react on fouling issues, but do not include active strategies to avoid them. The complete lack of advanced control systems in membrane technologies, capable to avoid fouling issues, limits the reliability of the technology and represent one key problem to be solved in order to permit further maturation of membrane applications. In order to achieve this result, the fouling behaviour of the system must be defined a priori. For liquid-liquid separation processes, Field et al. introduced the concept of critical flux for microfiltration, stating that there is a permeate flux below which fouling is not promptly observed (Field et al., 1995). Afterwards, it was possible to identify critical flux values on ultrafiltration ("UF") and nanofiltration ("NF") membranes systems, too (Mänttäri and Nystöm, 2000). Nowadays, the critical flux concept is well accepted by both scientists and engineers as a powerful membrane process optimization tool as long critical fluxes apply [7]. In case of most real waste water streams Le Clech et al. noticed that operations below the critical flux may not be sufficient in order to have zero fouling rates (Le Clech et al., 2006). Moreover, the measurement of critical fluxes was often not possible, and in order to overcome this problem, the identification of "apparent" critical points were used. Therefore it appears that membrane systems treating real waste water streams do not exhibit a critical flux in strict way. To overcome this limitation in the definition of critical flux, in a recent paper, Field and Pearce introduced for the first time the concept of the threshold flux (Field and Pierce, 2011). Summarizing briefly the concept, the threshold flux is the flux that divides a low fouling region, characterized by a nearly constant rate of fouling, from a high fouling region, where flux dependant high fouling rates can be observed. The main drawback of this concept is that the determination of threshold flux values cannot be theoretically predicted, but only experimentally determined by time consuming experiments at a certain time moment. The unavoidable development of fouling will irremediably change the relevant threshold flux value, too. The author actively performed much research in the last years by applying both concepts on different systems, such as olive mill, tannery and tomato industry wastewater streams. Finally, both critical and threshold flux concepts, that share many common aspects, were merged by the principal investigator into a new concept, that is the boundary flux, and was introduced in year 2014 (Stoller and Ochando-Pulido, 2014). The introduction of the new boundary flux concept does not extend by addition of new theory or knowledge the critical and threshold flux concepts. The important point is that the knowledge of real-time boundary flux values is a key factor to design a stable control system for membrane processes, since operation in sub-boundary flux conditions avoids irreversible fouling and thus premature technical (and economical) failures. The boundary flux values are sensibly influenced by those parameters affecting the critical and threshold flux, that is: hydrodynamics, temperature, membrane properties, time and feedstock characteristics. Concerning the first three points much has been reported in literature and the effects are nowadays well known. Moreover, it is relatively simple to maintain these parameters constant during membrane operation once fixed. Concerning time, the boundary flux values are as a function of this parameter, changing mainly due to the variable feedstock characteristics and the membrane actual health. This factor sensibly increases the difficulty to follow the boundary curve throughout operation, since theoretically boundary flux measurements must be frequently performed in order to accurately define the actual status of the process. Concerning the feedstock characteristics, Stoller and Ochando-Pulido performed some research on this topic by using olive mill wastewater streams, both exiting the 2-phase with (Ochando-Pulido et al., 2014) and without (Ochando-Pulido and Stoller, 2014) and the 3-phase olive oil production processes, again with (Stoller et al., 2014) and without (Stoller and Ochando-Pulido, 2012) pretreatment processes. A good design of the pretreatment processes (so-called pretreatment tailoring) beforehand the membrane section appears to sensibly increase the value of Jb, such as using photocatalysis with magnetic nanocore (Ruzmanova et al., 2013a) or doped nanotannia (Ruzmanova et al., 2013b), or regulated at different dosages (Stoller et al., 2013ab). Moreover, for control purposes, it appears mandatory that the key parameters were measured on-line, in-situ and real-time. Together with operating conditions measurement, an advanced control system, based on a simulation software capable to predict the actual boundary flux value, is capable to control the process in sub-boundary conditions. This advanced control system is put behind conventional control systems, and defines real-time proper set-points to optimize the process with insight of productivity, selectivity and longevity by avoiding irreversible fouling.
2. Basic control systems of membrane processes

Mainly two main membrane processes control strategies are adopted worldwide, that is controlling the permeate flow rate by changing the applied pressure value by a regulation valve on the retentate line, or controlling directly the constancy of the applied pressure.

The advantage of the constant permeate flow rate control strategy is the possibility to maintain the productivity of the membrane plant always in line with the project values. Moreover, the application requires the use of a simple proportional-integral (PI) type control system. The problem of this strategy is to define correctly the permeate flow rate set-point of the controlled system. Since boundary flux values will change as a function of time and of the pollutant concentration in the feedstock, it is not correct to assume as set-point value the boundary flux rate value at beginning of the operation. In particular, this problem would have higher impact when applied to batch membrane processes. In order to overcome this problem, a simulation software is required to estimate the boundary flux value at the end of the operation.

The advantage of the constant operating pressure value control strategy is the simplicity, but the productivity of the membrane plant may not be in line with the expected project values. The problem of this strategy is to define correctly the operating pressure set-point $P_P$ of the controlled system: the osmotic pressure will change as a function of time and of the pollutant concentration in the feedstock, and needs to be added to the TMP value; again, a simulation software is required to follow the changes during operation. In particular, this problem would have higher impact when applied to batch membrane processes.

The advantages and disadvantages of applying the control of the permeate flow rate (FC) or of the operating pressure (PC) to both continuous and batch membrane processes are hereafter briefly listed, partly in accordance with previous observations from Vyas et al. (2002): the membrane area requirements in case of batch processes are lower for PC strategies, in case of constant feedstock, if compared to FC strategies. The gap reduces sensibly as soon as $K_P$ changes accordingly to feedstock concentration variations. By the same terms, the advantage still holds for continuous processes. The control of the process is performed in both cases by a PI type controller. In case of changing feedstock at the start of operation or batch processes, advanced control is necessary: PC strategies requires the indirect measurement of the initial osmotic pressure of the feedstock, and this is in most cases not an easy task to accomplish; FC strategies requires the simulation software to calculate a new value for FF. In both cases the purpose it to give a different set-point to the PI type control systems.

In Figure 1, the two basic control strategies are reported and the relevant block diagram is shown.

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![Figure 1: Scheme and block diagrams for the basic control systems of FC (left) and PC (right) strategies.](image-url)
3. The boundary flux equations

The boundary flux divides the operation of membranes in two different regions: a lower one, where no or a small, constant amount of fouling triggers, and a higher one, where fouling builds up very quickly. The relevant equations may be written as:

\[
\frac{dm}{dt} = -\alpha; J_p(t) \leq J_b \quad (1)
\]
\[
\frac{dm}{dt} = -\alpha + \beta (J_b(t) - J_b); J_p(t) > J_b \quad (2)
\]

where \(\alpha\), expressed in \(L \cdot h^{-1} \cdot m^{-2} \cdot \text{bar}^{-1}\), is the sub-boundary fouling rate index and \(\beta\), expressed in \(h^{-1} \cdot m^{-2} \cdot \text{bar}^{-1}\), is the super-boundary fouling rate index.

The boundary flux value itself is a function of many parameters, mainly affected by hydrodynamics (Re number), temperature (T) and the solute concentration (KP). In this work the key parameter KP will be the COD value of the feedstock. If the other parameters remain constant, the relationships can be summarized separately in the following set of equations:

\[
J_b(\text{Re}) = J_b(\text{Re} \to \infty) (1 - e^{-A \text{Re}}) \quad (3)
\]
\[
J_b(T) = AT T^2 + BT T + CT \quad (4)
\]
\[
J_b(KP,t) = w P_b(0) - \alpha t P_b(0) - [w p_1 - \alpha p_1 t + m_1 P_b(0)] KP + m_1 p_1 KP^2 \quad (5)
\]

With \(A, A_T, B_T\) and \(C_T\) fitting parameters. It is interesting to notice that only the last one is a function of time as well, even if KP remain constant during operation. On contrary, by proper control systems, both Re and T can be maintained constant by fixing their set-point. In eq.(5) \(w, m_1\) and \(p_1\) are fitting parameters of the respective equations fitting the membrane permeability \(m\) and the osmotic pressure \(\pi\) to KP:

\[
m(KP,t) = w - m_1 KP \quad (6)
\]
\[
\pi(KP) = p_1 KP \quad (7)
\]

and where \(P_b(0)\) is the applied operating pressure at the boundary flux conditions at the start of operation, in detail:

\[
P_b(0) = \text{TMP}_b + \pi(KP)|_{t=0} \quad (8)
\]

The pure water permeability, that is \(w\), may be a function of time depending from the amount of irreversible fouling formed over on the membrane.

The definition of a value for \(J_b\) permits to determine the range of validity of eq.(1), here of interest for membrane process design purposes.

The model is completed by two equations concerning permeate fluxes and selectivity, that is:

\[
J_p(t) = J_p(0) - \alpha \text{TMP}(t) t; J_p(0+t) \leq J_b \quad (9)
\]
\[
R(KP) = \sigma \text{TMP} (\text{TMP} + \gamma)^{-1} \quad (10)
\]

where \(\sigma\) is the reflection coefficient and \(\gamma\) a fitting parameter.

4. Advanced control system

Eqs(3-10) define the value of \(J_b\) once values of KP, T, t, Re are measured and the other fitting parameters are known. In the hypothesis to have all the available data to run the model, the proposed equations are suitable for advanced control.

If the membrane area \(A\) and the total operating time \(\tau\) is fixed, the set-point of the two control strategies may be defined as follows:

For FC strategy: \(F_p^{SP} = 0.9 J_b(\tau) A\) \quad (11)
For PC strategy: \(P^{SP} = 0.9 P_b\) \quad (12)

where 0.9 is an applied safety factor. In order to evaluate \(J_b(\tau)\), following relationship can be used if the final value of the desired recovery factor \(Y^*\) is fixed:
KP(t) = KP(0) \left( 1 - Y' + Y' R(TMP_b) \right) \left( 1 - Y' \right)^{-1} \tag{13}

Figure 2 shows the relevant block diagrams.

5. Results and discussion

The two control strategies were applied to a batch membrane system well known in terms of data and parameters and reported elsewhere, and hereafter briefly summarized in Table 1 (Stoller, 2013). The feedstock was a pretreated olive mill wastewater stream (COD equal to 20000 mg/l), the used ultrafiltration membrane was supplied by GE Water (Model GM).

In Figure 3 the results were shown.

In case of both strategies, the beginning polarization of the membrane is not taken into account by the model, thus permeability and pressure data points were slightly off curve before their stabilization.

Batch membrane processes are not easy to control since as a function of time and operation, KP changes accordingly due to the concentrate recycle and increase of solute concentration. From these preliminary results, a good control of the batch membrane process at the fixed \( F_p^{SP} \) or \( P^{SP} \) values, respectively, was observed and the suggested approach in this paper appears to be validated.

Both processes were operated to the end, and after this no fouling was observed.

6. Conclusions

Basic control system for membrane processes may not prove to be sufficient in those cases severe fouling issues are triggered, as a consequence of a manual setting of the set-point values to those of the project. The problem is that the boundary flux values will change as a function of time, and fixed set-point values may overcome boundary flux conditions during operation. Advanced control systems are desirable and should provide suitable set-point values to the control system in order to perform the operation always below the actual boundary flux value, avoiding irreversible fouling operating conditions.

The model reported in the paper was capable to provide suitable set-point values for both constant permeate flux and constant TMP control strategies leading to the end of operation of a wastewater batch membrane treatment process, without formation of irreversible fouling. The success of the method relies on the determination of some parameters and the continuous measurement of others. If properly set up, the proposed approach may fit to other membrane batch systems, providing advanced process control with the target to avoid irreversible fouling formation.
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