Flux-step method for the assessment of operational conditions in a submerged membrane bioreactor

Ezio Ranieri, Vito Goffredo, Mariachiara Campanella and Michael W. Falk

ABSTRACT

A flux-step method was used for monitoring the pressure variation in a solids separation membrane at different operating conditions. A submerged membrane bioreactor pilot plant, used during the short-term tests, was used to purify actual restaurant wastewater. The influence of membrane backwash and relaxation on the variation of pressure variation was also evaluated. In order to reduce the deposition of irreversible fouling, the authors modified the literature-supported filtration to backwash cycling with filtration and relaxation cycling. The trials maintained a constant filtration to relaxation ratio that was in line with optimal filtration to backwashing ratios found in the literature. The relaxation cycling between two constant flux-steps effectively counteracted membrane fouling and the excessive decrease in average pressure, and it results in a lower waste of energy and water than a backwashing strategy.

Key words | flux-step method, fouling, membrane biofouling cleaning protocol, submerged membrane bioreactor, wastewater

INTRODUCTION

It is well documented that membrane bioreactors (MBRs) produce better effluent quality than conventional wastewater treatment processes. Trussell et al. (2000) demonstrated that a well-operated activated sludge process produces an effluent characterized by suspended solids ≤10 mg/L, turbidity ≤10 NTU and biochemical oxygen demand ≤ 10 mg/L, while the MBR effluent typically contains suspended solids ≤2 mg/L (non-detect), turbidity ≤0.2 NTU and biochemical oxygen demand ≤ 2 mg/L (non-detect). MBR systems combine a biological process with a membrane to separate liquids from solids. MBRs are widely used for municipal and industrial waste treatment (Seo et al. 1997; Ahn et al. 1999; Stephenson et al. 2000). This combination leads to many advantages, such as the elimination of clarifiers, ability to operate at higher mixed liquor suspended solids (MLSS) concentrations, longer sludge ages, improved effluent water quality, and a compact footprint. Such conditions also lead to less sludge production, with a consequent reduction in sludge treatment and disposal costs, and a lower reactor volume. The presence of the membrane allows the growth of microorganisms which are more acclimated to high molecular weight and/or recalcitrant compounds and have the ability to degrade them. For these reasons, MBRs also present better removal efficiency of micropollutants, persistent organic pollutants and slowly biodegradable pollutants (Bernhard et al. 2006; Petrella et al. 2013), so reducing the potential for formation of disinfection by-products (Ranieri & Świetlik 2010).

One of the most important disadvantages of an MBR system is the deposition of submicron particles on the membrane surfaces (abiotic and biotic fouling); the different amount of these foulant deposits is also related to permeate flux (Guglielmi et al. 2007). This phenomenon leads to increase in hydraulic resistance, with the decline of permeate flux during a constant-pressure process, and of the suction pressure in constant-flux conditions. Membrane fouling remains one of the major obstacles to widespread MBR installations (Kimura et al. 2007; Akamatsu et al. 2010). Additionally, membrane fouling in MBRs potentially decreases membrane life-span and increases energy, chemical, and labor demands (Kim & Lee 2014).

There are two distinct forms of fouling, reversible and irreversible. Reversible fouling can be resolved by cleaning protocols and routine maintenance, whereas irreversible fouling can only be removed by a chemical cleaning with products that restore the initial permeability (Judd 2006). Determining the best cleaning protocol and optimal
permeate flow are extremely important for maintaining reversible fouling. Even if a control module based on a real-time comparison of permeability trends against a reference trend were used, the high number of variables that influence membrane fouling complicates the design of such a knowledge-based control module (Yigit et al. 2008; Comas et al. 2010; Drews 2010). These considerations support developing an operating protocol to maintain permeate quality and flow while reducing energy consumption.

The introduction of backwashing during an MBR filtration process has the potential to remove the majority of the foulant layer on the membrane surface (Smith et al. 2005; Van Lienden et al. 2010). The backwashing frequency is a vital parameter for successful long-term MBR operations because it reduces the number of intensive chemical cleaning operations, minimizing the extent of fouling. Maintaining a rigorous control strategy can reduce the reversible foulant layer propensity and in turn reduce the degree of irreversible fouling (Smith et al. 2006).

The particle matrix deposited on the membrane surface is composed of biological flocs formed by a large range of living micro-organisms along with soluble and colloidal compounds, but the concentrations of mixed liquor suspended solids and extra-polymer substances are linked to the operating conditions. Given the multitude of parameters, fouling is unpredictable and difficult to control (Le-Clech et al. 2005).

A short-term flux-step method (on the order of hours) has been successfully applied, continuously monitoring pressure changes at various operating conditions while maintaining all other parameters (MLSS concentration, solids residence time, feed wastewater composition).

Flux-stepping is the most popular method used to estimate critical flux. Wu et al. (2008) found that the increase of step length or height causes a decrease in critical flux. The use of smaller steps facilitates a more accurate determination of critical flux, but it comes at a cost in the form of time by changing the fouling propensity. Guglielmi et al. (2007) verified the reliability of short-term flux-stepping by determining the sustainability time in a large pilot plant. Critical flux determination techniques by flux-step method could be divided into two methods: (a) common short-term flux-step method, with increase in flux or permeate suction rate, and monitoring the corresponding pressure; (b) improved flux-step method in which the membrane is operated at higher flux, followed by a lower flux for an equal duration before the next higher flux level (Navaratna & Jegatheesan 2011).

Flux-stepping method has been used by several authors (Yigit et al. 2008; Kim et al. 2011; Van den Broeck et al. 2012) to determine preferred MBR operational conditions. Yigit et al. (2008) used a flux-step method characterized by variable operational conditions to establish the impact of different backwash scenarios on fouling in an MBR pilot plant that aerobically treated domestic wastewater. Flux-step method was also employed to understand the optimum membrane aeration time by modifying the blower aeration intensity to determine the relationship between the applied flux and MBR run time (Guglielmi et al. 2007).

The flux-step methods’ results change not only with operational conditions, but also with the MLSS. Navaratna & Jegatheesan (2011) demonstrated that the increase of suction pressure over time was higher in tests conducted at higher MLSS concentrations. The method was also used in the same study to verify the variation of permeate chemical oxygen demand concentration at different fluxes.

The operating protocols used in the present study were characterized by different durations of the constant flux-steps and by different cleaning protocols, with the introduction of relaxation or backwashing periods. The tests use an incremental flux increase for a fixed duration per increment. This strategy is preferred over the constant pressure-step method because the convective flow of solute towards the membrane is kept constant during the run, which facilitates improved control of the flow of material deposition on the membrane surface (Defrance & Jaffrin 1999). The results in terms of pressure variation (∆P), rate of pressure variation (dP/dt) and average pressure are closely related to step duration, step height, initial state of the membrane (new/backwashed), feed characteristics and system hydraulics.

The aim of the paper is to understand the relationship between these parameters and the system operating conditions, assessing the best operating protocol and the optimal permeate flow to be taken during the long-term real run. The analysis of the state of the art has shown other authors have already widely studied the relationship between backwashing duration and frequency and fouling. The study analyzes the possibility to reduce the waste of water and energy due to the backwashing operations, by replacing them with relaxation periods characterized by the same frequency and duration.

**MATERIALS AND METHODS**

**MBR**

The restaurant Sala Azzurra in Mottola (Italy) had a conventional suspended growth bioreactor. The system was
retrofitted with two submerged hollow-fiber membranes (model MBR4.5FR – Water Code membrane technology Co., Ltd) and a continuously operating blower. Upstream of the biological tank there was a screening phase, a 30 m³ equalization tank and a 10 m³ pre-denitrification tank with a continuously operating submerged mixer. The nitrification-oxidation tank was also equipped with an submerged air diffuser at the base of the tank, operating under conditions of constant airflow. The biological reactor worked with a MLSS concentration of 4,000 to 5,000 mg/L. Flux filtration was carried out using a suction pump (P1). Another pump (P2) was used to pump mains water through the membranes during each backwash cycle. The permeation rate was steadily increased and the pressure change continuously monitored by a pressure transducer in the permeate line and linked to the control system which records these values on a computer to manually and/or automatically control the system components (Figure 1).

The nominal pore size of each membrane (0.750 m length and 0.500 m width) was 0.1 μm with a total standard filter area of 9 m² and a recommended permeate flow between 0.8 and 1.5 m³/day for the membrane unit. The recommended vacuum should be less than 0.01 Mpa to allow the filtration phase and greater than 0.03 Mpa to avoid the excessive obstruction of the pores. The characteristics of the submerged MBR are shown in Table 1.

**Operating protocol**

The experimental set-up parameters were based on a combination of values developed during the calibration phase and those in the literature (Visvanathan et al. 1997; Larrea et al. 2002; Smith et al. 2006; Trigo et al. 2006; Irizar & Gracia 2008; Wu et al. 2008; Monclús et al. 2010) and the specific features of the submerged membranes, such as the recommended values for the maximum and minimum suction pressure and the recommended permeate flow. The hypothesized operating protocol assumed 9 min of filtration with 1 min of relaxation of the membranes and an operating pressure of 100 mbar to allow the ‘out-to-in’ passage of the water through the fibers. During each run, pressures must be in the range of 300 to 300 mbar. A pressure less than the minimum was attributed to clogged pores, while pressure greater than the maximum value result in ruptured membranes.

To avoid excessive fouling caused by biotic foulants, a cleaning protocol was developed with fixed backwashing frequencies (1 min of backwash water every 120 min of filtration and 30 min of backwash with sodium hypochlorite every month). The membranes were also backwashed with a pressure below 300 mbar. The short-term tests facilitated a rapid confirmation (hours) of test parameters prior to any long-term runs.

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**Figure 1** | Pilot plant and system controls.
Table 1 | Membrane characteristics

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Units</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Length × width</td>
<td>mm</td>
<td>750 × 500</td>
</tr>
<tr>
<td>Standard filter area</td>
<td>m²</td>
<td>4.5</td>
</tr>
<tr>
<td>Recommended flow</td>
<td>m³/day</td>
<td>0.8–1.5</td>
</tr>
<tr>
<td>Design flow for unit area</td>
<td>L/h-m²</td>
<td>10–20</td>
</tr>
<tr>
<td>Material of membrane</td>
<td>unitless</td>
<td>Reinforced PVDF</td>
</tr>
<tr>
<td>Capillary diameter (I/O)</td>
<td>mm</td>
<td>1.0–2.4</td>
</tr>
<tr>
<td>Longitudinal breaking force</td>
<td>N</td>
<td>&gt;200</td>
</tr>
<tr>
<td>Type of operation</td>
<td>unitless</td>
<td>Outside–In (vacuum type)</td>
</tr>
<tr>
<td>Molecular weight cut-off</td>
<td>Dalton</td>
<td>500 K</td>
</tr>
<tr>
<td>(pore size = 0.1 μm)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Recommended vacuum</td>
<td>Mpa</td>
<td>–0.01 to –0.03</td>
</tr>
<tr>
<td>Temperature</td>
<td>°C</td>
<td>5–45</td>
</tr>
<tr>
<td>pH</td>
<td>s.u.</td>
<td>2–12</td>
</tr>
<tr>
<td>Continuous chlorine resistant</td>
<td>ppm</td>
<td>500 ppm</td>
</tr>
<tr>
<td>N-Hex resistant</td>
<td>mg/L</td>
<td>&lt;3</td>
</tr>
</tbody>
</table>

The study involved a series of experiments conducted changing the step length (from 10 to 30 min). Tests were divided into cycles characterized by different operating conditions: in the first cycle, flux-steps with the same step height (5 L/m²·h) at different lengths (10, 15 and 30 min) were carried out, while the second and third cycles consisted, respectively, in the repetition of two cycles of flux-stepping with the same step length (10 min) and height (5 L/m²·h) (with or without the interposition of 5 min of relaxation or backwashing), and in the introduction of 1 min of relaxation or backwashing after every single flux-step. The last backwashing duration was based on case studies that met the following criteria: treated domestic wastewater, used a similar membrane, and attempted to develop an optimal filtration/backwashing ratio with both steady-state and dynamic fouling models (Yigit et al. 2008; Kim et al. 2011; Van den Broeck et al. 2012). Table 2 summarizes the conditions of the experiments conducted in the context of the different test cycles.

The continuous flux-step method, characterized by a different step height and length and proposed in the trials under cycle number 1, has been carried out in order to understand the value of the apparent sustainable flux, while the second cycle of tests consisted of the interposition of a quite long cleaning period (5 min) between two continuous flux-steppings with the aim of testing the effect of this operation on long filtration periods. With the last two tests (Test #7 and Test #8), the ability of frequent relaxation periods to counteract the negative effects due to the fouling has been tested in comparison with backwashing intervals having the same duration and frequency.

Experiments characterized by six flux increments and as many decreases in the same conditions were carried out in such a way as to compare the pressures registered in the ascending and descending phases (Figure 2).

Data processing

For each flux-step, two pressure values were reported: the initial pressure (P_i), defined as the pressure following the step sudden increase in flux, and the final pressure (P_f), defined as the pressure at the end of the step. These values were taken from the trend line of the pressures, because of the large variability of pressure even at constant flow.

<table>
<thead>
<tr>
<th>Cycle #1 (flux-step method with different step durations)</th>
<th>Test</th>
<th>Step height = 5 L/m²·h, step duration = 10 min</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>#1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>#2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>#3</td>
<td></td>
</tr>
<tr>
<td>Cycle #2 (two flux-stepping cycles)</td>
<td>Test</td>
<td>Two flux-stepping cycles without interposition of</td>
</tr>
<tr>
<td></td>
<td>#4</td>
<td>cleaning operation</td>
</tr>
<tr>
<td></td>
<td>#5</td>
<td>Two flux-stepping cycles with interposition of 5 min of</td>
</tr>
<tr>
<td></td>
<td>#6</td>
<td>relaxation</td>
</tr>
<tr>
<td>Cycle #3 (flux-step method with cleaning protocol)</td>
<td>Test</td>
<td>Step height = 5 L/m²·h, step duration = 15 min, 1 min of</td>
</tr>
<tr>
<td></td>
<td>#7</td>
<td>membrane relaxation after every flux increment</td>
</tr>
<tr>
<td></td>
<td>#8</td>
<td>backwashing after every flux increment</td>
</tr>
</tbody>
</table>

Figure 2 | Example of ascending and descending phases.
From these two values, other parameters could be defined: the initial pressure decrease $\Delta P$ the rate of pressure decrease $dP/dt$ and the average pressure $P_{ave}$.

\[
\Delta P = P^n_f - P^{n-1}_f \tag{1}
\]

\[
\frac{dP}{dt} = \frac{P^n_f - P^n_i}{t^n_f - t^n_i} \tag{2}
\]

\[
P_{ave} = \frac{P^n_f - P^n_i}{2} \tag{3}
\]

These parameters are graphically summarized in Figure 3.

**RESULTS AND DISCUSSION**

It was observed that in each cycle of tests the pressure values obtained during the ascending phase were greater than the corresponding values recorded during the descending phase, unlike the results obtained during the study carried out by Van der Marel et al. (2009). This observation indicates the formation of an initial fouling layer and that the membrane was not fully recovered from fouling during the descending phase (Navaratna & Jegatheesan 2011).

All the trials were characterized by values of the pressure rate ($dP/dt$) that were never equal to 0. This is contrary to a number of publications (Tardieu et al. 1998; Madaeni et al. 1999) in which stable operation (or constant membrane permeability) has been reported.

The first cycle (flux-steps with the same step height at different lengths) was carried out to understand the relationship between pressures and constant flux–step duration. The results obtained in terms of $\Delta P$ for 5 and 15 min step durations were similar and relatively constant during the test. In contrast, increasing the step length to 30 min resulted in a decrease in $\Delta P$ at flux values above 25 L/m$^2$·h (Figure 4). The step length variation also influenced the $dP/dt$ values. In fact, the results yielded showed an increase in pressure rate with the flux which was more pronounced in the 15 min test (Figure 5). These results were comparable with those obtained by Le-Clech et al. (2005) who observed that increasing the step length from 5 to 120 min produced a significant variation in $\Delta P$ at flux values above 15 L/m$^2$·h. In this case speaking about the so-called sustainable flux is not possible, which represents the flux value at which the fouling rate is operationally and economically acceptable.
for MBR operation (Wang et al. 2008), because such a value can only be assessed in long-term trials; the value of 15 L/m²·h obtained during the presented study might be identified in a more accurate way as an apparent sustainable flux that comes from short-term trials (Drews 2010). It is interesting to note that studies conducted with the same type of membrane and wastewater (Navaratna & Jegatheesan 2011) lead to a very similar value of apparent sustainable flux, while this value changes significantly with differences in the membrane characteristics and history and different chemical composition of the wastewater (Wu et al. 2008).

An increase of average flux would be expected to provide a corresponding increase in fouling rate. Although the data obtained confirm this hypothesis, other studies (Wen et al. 2004; Brookes et al. 2006) reported a decreasing trend of fouling rate with flux. This would suggest that cake compression is not the only mechanism affecting the fouling rate once the local flux is higher than the dominant foulant critical flux (Guglielmi et al. 2007).

The importance of relaxation and backwashing on fouling potential was confirmed by performing flux-step tests characterized by the introduction of relaxation and backwashing phases with different frequencies and durations. The repetition of two cycles of flux-stepping with the same step length (10 min) and height (5 L/m²·h), performed with the interposition of 5 min of relaxation or backwashing, resulted in no differences when consecutive cycles were carried out without intermediate cleaning or relaxation. This is also in accordance with the study carried out by Yigit et al. (2009) who observed that the negative impact of filtration duration on the membrane resistance was more pronounced than the positive impact due to the increase of the backwash duration. In other words, fouling control could be improved by introducing more frequent backwashing in order to decrease the filtration duration. Instead the introduction of 1 min of backwashing after every single flux-step caused a significant variation in the pressure parameters (ΔP and dP/dt) compared to the results obtained during the test.
characterized by the same step length and height but without a cleaning protocol. An increase in backwashing and relaxation frequency phases caused a great increase in ΔP and dP/dt with the flux. In these cases, the parameters also appeared higher than the continuous experiment for flux values over 15 L/m²·h (Figures 6 and 7). These findings suggest that as the interval between backwashing or relaxation is extended, the foulant layer on the membrane compacts and becomes increasingly difficult to remove during the backwash cycle, causing the transition from reversible to irreversible fouling (Smith et al. 2006). Lower instantaneous flux was suggested to be applied to obtain better effect and, compared with relaxation, backwashing did not exhibit a more positive effect on fouling control. By applying intermediate relaxation, fouling appears to be almost completely reversible, indicating that cake layer formation is the dominant fouling mechanism. It is possible to provide a cleaning protocol made by relaxation periods with high frequency, avoiding in this way the waste of energy due to the frequent use of backwashing and maintaining unchanged the performances of the system.

These last two tests show that also with the introduction of backwashing/relaxation phases there is an increase of the suction pressure registered during constant-flux conditions, which becomes more important with the increase of the flux. However, the introduction of these intermediate cleaning phases at the end of every step allows the effect of the fouling layer on the membrane surface to be reduced. For this reason, compared to the values obtained during the first cycle of tests, the absolute value of the initial pressure Pi recorded in each constant flux-step considerably decreased because the interposition of a cleaning protocol made each flux-step almost independent from the previous one (the intermediate relaxation/backwashing removed the reversible fouling and reduced fouling history). This caused an increase in the values of ΔP that also reached a positive value, with Pi at the flux of 30 L/m²·h higher than the Pf reached when maintaining a constant flux of 25 L/m²·h for 15 min. Figure 8 shows the variation of pressures obtained by introducing 1 min of relaxation after every step.

This observation demonstrates that the introduction of an intermediate relaxation phase is able to reduce the irreversible component of fouling and can also decrease the influence of fouling history.

CONCLUSIONS

- The increase in step length and height causes a greater pressure variability with the flux.

Figure 6 | Effect of backwashing and membrane relaxation on ΔP.

Figure 7 | Effect of backwashing and membrane relaxation on dP/dt.
The optimal permeate fluxes obtained by introducing 1 min of relaxation.

- Pressures decrease over time with a constant flux, as an effect of fouling.
- The introduction of 1 min of membrane relaxation with the blower turned on, after 15 min of filtration, is able to remove the so-called ‘reversible’ component of the foulant layer.
- The optimal permeate flow of the system is 15 L/m²·h, because, when the plant works under this value of flux, the registered suction pressures remain almost constant even if the other operating conditions (step height and length) change. This value can be identified as the so-called apparent sustainable flux of the system.

The study has shown that reducing the backwashing frequency and replacing the cleaning operation step with relaxation periods represents an opportunity to improve MBR operational costs. Replacing the backwash step with relaxation at the same frequency and duration suggests the possibility to reduce water wasting and energy demand (limited to the blower presence), maintaining MBR performance for submerged hollow-fiber modules. The introduction of intermediate relaxation has reduced the influence of fouling history with a significant decrease in the fouling rate, showing almost no fouling rate hysteresis at equal fluxes in the ascending and the descending phase.

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