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The role of blocking and cake filtration in MBR fouling

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Abstract

Membrane fouling in a side stream biomass separation MBR pilot plant was investigated. Constant flux filtration (18–72 l/m^{2}h) was employed. Air was continuously supplied to the MBR system with the feed (sludge) to flush the membrane surface, and backwashing was applied every 5–10 min for 8 s to control membrane fouling. Although the duration of pore blocking was generally short (completed in 8 s at a flux of 52 l/m^{2}h), blocking resistance (mainly irreversible blocking resistance) was the main cause of membrane fouling. However, the resistance of the filter cake also played an important role, particularly when the backwashing interval was extended to 10 min. In terms of fouling reversibility, blocking resistance was not completely reversible by backwashing, especially at higher fluxes (e.g. 69 l/m^{2}h), and frequent chemical cleaning (once every week at 40 l/m^{2}h) was required. However, cake filtration was easily reversible via a combination of backwashing and sludge/air flushing of the membrane surface. Finally, a simple method to identify both irreversible and reversible blocking resistance and filter cake resistance was proposed.

Keywords: Membrane bioreactor; Fouling; Resistance; Blocking; Cake filtration

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1. Introduction

Feed water characteristics strongly affect membrane fouling in all membrane filtration systems. In membrane bioreactors (MBR), the feed to the membrane module is activated sludge, and its composition is very complicated. Generally, it comprises biological flocs, dispersed bacterial cells, protozoa, rotifers, and organic and inorganic compounds either introduced from raw wastewater or produced during biomass growth and decay. Bacteria are the major fraction of activated sludge, and they play the most important role in organic matter degradation and biological nutrient removal. Protozoa and rotifers act as effluent polishers. Protozoa consume dispersed bacteria that have not flocculated, while rotifers consume small biological floc particles that have not flocculated [1,2]. However, most microorganisms in activated sludge are flocculate into microbial flocs.

There are contradictions in literature about the major foulants in MBR systems. Wisniewski and Grasmick [3], Defrance et al. [4] and Bouhabila et al. [5] studied the resistance fractions caused by solutes, colloids and suspended solids respectively and drew different conclusions. Wisniewski and Grasmick reported that solutes were the major foulant (solutes 52%, colloids 25%, suspended solids 23%). Defrance et al. reported that suspended solids were the major foulant (solutes 5%, colloids 30%, suspended solids 65%), and more recently Bouhabila et al. reported that colloids were the major foulant (solutes 26%, colloids 50%, suspended solids 24%). Their different results may be due to the differences in biological operating conditions, membrane morphology, filtration conditions (especially hydrodynamics) and separation methods.

Foulant sizes may strongly affect fouling mechanisms in membrane filtration systems. If foulants have comparable or smaller sizes than the membrane pores, pore blocking may occur. However, if foulants are generally much larger than the membrane pores, they cannot enter the pores and a cake layer may be formed on the membrane surface. Most membranes (MF/UF) used in MBR have pore diameters between 0.01 μm–0.1 μm, which is within the colloidal range. However, most foulants (microbial flocs) in MBR are much larger than the membrane pore size (10–50 μm) [4,6,7], and are thus too big to enter and pass the membrane pores. This is the basis of the high rejection of suspended solids in MBR and cake formation on the membrane surface. However, colloidal and soluble matters have sizes comparable to the membrane pores and their rejection by the membrane may be poor. Moreover, the mechanisms of rejection and fouling are mainly due to adsorption on the cake layer or deposition within the pores of the membrane [8].

In many cases, the accumulation of a filter cake is the principal fouling mechanism in MBR. Lee et al. [9] reported that membrane resistance, cake resistance, blocking and irreversible fouling resistance contributed 12%, 80% and 8%, respectively to the total resistance of a submerged MBR (MLSS 3000 mg/l, 0.1 μm Mitsubishi UF). Chang and Lee [10] reported that cake resistance was the major contributor to the resistance of membrane coupled activated sludge systems, especially under low sludge age conditions. However, colloidal and soluble foulants were also important, as they cause pore blocking and irreversible fouling due to their small size. Bouhabila et al. [5] observed that the supernatant of MLSS had 20–30 times higher specific resistance than the sludge suspension, which illustrated the high fouling potential of soluble and colloidal fractions in activated sludge.

In this research, firstly blocking (R_b) and cake resistance (R_c) in a side-stream MBR were quantified for a range of fluxes and backwashing intervals. Secondly, a simple method to identify both irreversible and reversible blocking resistance and filter cake resistance was proposed. Finally, based on the rate of increase of irreversible resistance (R_{ir}), a simple method was developed to predict chemical cleaning frequencies.

2. Materials and methods

2.1. MBR set-up

A side-stream MBR pilot plant, located in Beverwijk the Netherlands, was fed with wastewater from the Beverwijk Zaanstreek wastewater treatment plant, and comprised mainly domestic wastewater. The raw wastewater was passed through a 0.5 mm micro-screen to remove hair, debris, rags and sand etc. Thereafter, the prescreened water entered the aeration tank where organic matter, nitrogen and phosphorous were removed. The effluent of the aeration tank was pumped to four ultrafiltration (UF) membrane modules for biomass separation. Permeate from the UF system was stored in a clean-water tank and the concentrate sludge of the UF system was returned to the aeration tank (Fig. 1).

2.2. Membrane module operation and cleaning

The ultrafiltration membrane modules were supplied by X-Flow. Each UF module comprised 600 vertical membrane tubes, with the length of 3 m and an internal diameter of 5.2 mm. The average pore size of the PVDF membrane was 0.03 µm. 20 m^3/h of activated sludge from the aeration tank was fed to the bottom of the UF module (inside-outside configuration), and 20 m^3/h air (at 0.56 bar) was supplied concurrently with the sludge to continuously flush the membrane surface. Permeate was collected at the outside of membrane module by a vacuum pump, while the concentrate flow (biomass), rejected by the UF membrane was returned to the aeration tank. The UF module was operated at a constant gross flux of 18–72 l/m^2h.

The liquid/air two-phase flow in the UF module was identified as "slug flow", with alternating bullet-shaped bubbles of gas surrounded by a thin liquid annulus and interspersed with slugs of liquid containing small bubbles of gas, to maximise shear force sloughing the membrane surface [11]. The UF membranes were also periodically (every 5 or 10 min) backwashed for 8 s by pumping a fraction of permeate back through the membrane. The backwashing pressure was around 0.8–0.9 bar. Additional air (for 4 s) was supplied by an air compressor to the feed water during backwashing to flush the inside of the tubes and enhance the removal of accumulated foulants during backwashing. When severe membrane fouling occurred, the UF membranes were chemically cleaned (using NaOCl and citric acid).

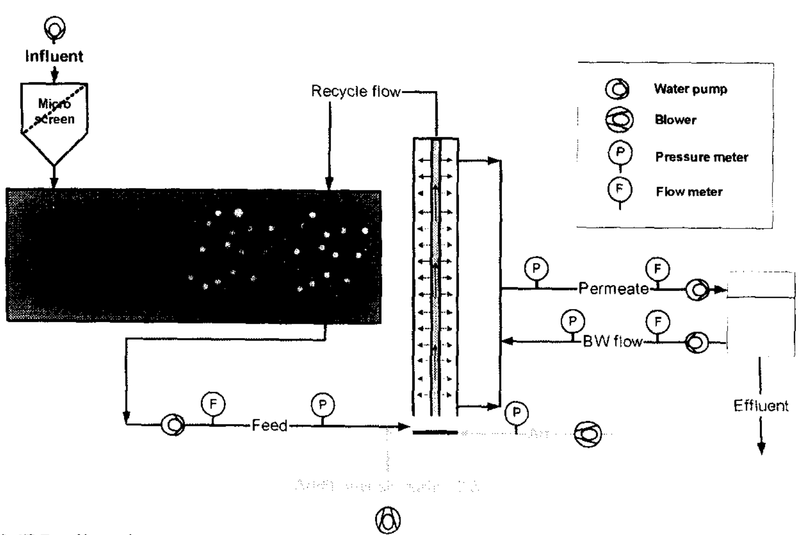

Fig. 1. Schematic of MBR pilot plant.

2.3. Data collection

In order to detect the rapid increase in transmembrane pressure (TMP) at the start of each filtration cycle, a very high sampling frequency (i.e. once per 0.2 s) was applied. Using a computer, the following data were recorded automatically every 0.2 s: time, temperature, feed flow, permeate flow, feed pressure, permeate pressure, BW flow, and backwashing (BW) feed pressure.

3. Results and discussion

3.1. The effect of flux on the shape of TMP vs. t *curve*

At a flux of 52 l/m²h, pore blocking, with a simultaneous rapid increase in TMP, was completed within 8 s (Fig. 2), and was attributed to the presence of organic solutes and colloids in the feed water (activated sludge). A typical low load activated sludge process has about 28 mg/l soluble COD (with a diameter less than 0.45 μm), which either originated from the raw wastewater (10 mg/l) or was produced during biomass metabolic processes (soluble microbial produces, SMP, 18 mg/l) [12]. The soluble and colloidal organic matter are comparable in size to the membrane pores (0.03 μm) and may be responsible for blocking the pore entrance (complete blocking). Furthermore, very small colloids can enter membrane pores and absorb on the walls (standard blocking).

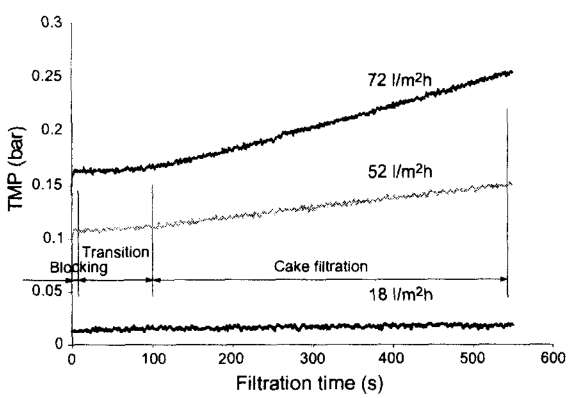

Fig. 2. Steps of membrane fouling in MBR.

The transition stage (8–100 s) following the blocking stage was characterised by a slower increase in TMP (Fig. 2). Finally, cake filtration dominated after 100 s (100–600 s) with a linear increase in TMP. Compression of the cake layer and depth filtration were not observed, which may be due to the relatively low TMP (up to 0.13 bar) and flux (52 l/m²h) applied in these tests. In addition, the fraction of colloids and solutes was small compared to the total solids in the feed sludge (28 mg COD/l in 10,000 mg/l total solids), and thus the contribution of colloids and solutes to depth filtration was probably negligible compared with the resistance of the filter cake, which comprised ca. 10,000 mg/l total solids (mainly bioflocs).

It is interesting to observe that, in Fig. 2, pore blocking was hardly visible at fluxes below 18 l/m²h, This was probably due to a combination of two factors: (i) less permeate was produced at 18 l/m²h, and consequently few foulants were deposited, and (ii) the particle permeation velocity towards the membrane was very low at 18 l/m²h (5.0×10^{-6} m/s). Therefore, the back transport velocity generated by shear force on the membrane surface was sufficient (6.4×10^{-6} m/s for 1 μm spherical particles) to limit the deposition of even small foulants [13]. However, at higher fluxes e.g. 72 l/m²h, the rate of blocking was significantly higher due to the increased permeation velocity (2.0×10^{-5} m/s). Consequently, suspended and colloidal foulants were rapidly deposited on the membrane, as the back transport velocity was much lower than the permeation velocity (i.e. 6.4×10^{-6} m/s for 1 μm spherical particles vs. 2.0×10^{-5} m/s).

3.2. Quantifying pore blocking and cake filtration

Pore blocking and cake filtration were quantified from Fig. 2 by calculating the initial resistance immediately after backwashing (R_{bw}, the starting point of a filtration cycle), the reversible blocking resistance (R_{reb}, the observed initial rapid increase in TMP), the cake resistance (R_c, the moderate increase in TMP dominating the rest filtration period) and the total resistance (R_{tot}, the resistance at the end of a filtration cycle). Within a 300 s filtration cycle, the initial resistance immediately after backwashing (R_{bw}) accounted for 69–85% of the total resistance; reversible blocking resistance (R_{reb}) accounted for 0–14% and the cake resistance (R_c) accounted for 15–21% within the flux range 18–72 l/m²h (Fig. 3).

The initial resistance immediately after backwashing (R_{bw}) increased with increasing flux (Fig. 3, from 2.2×10^{11} 1/m at 18 l/m²h to 4.5×10^{11} 1/m at 72 l/m²h), and may be attributed to the following: Firstly, when the gross flux increased from 18 l/m²h to 72 l/m²h, more permeate was produced and more foulants including macro organic colloids and solutes, which may contribute to irreversible fouling, were deposited on the membrane. Secondly, backwashing was probably less effective at high fluxes as some foulants may not be well removed and remain in/on the membrane after backwashing.

Cake resistance (R_c) was low for all fluxes and was attributed to the following reasons. Firstly, backwashing was applied frequently (once every 5 min), which removed the cake layer before it became too thick. Secondly, the crossflow velocity (1 m/s mean crossflow velocity) and sludge/air slug flow enhanced the back transport of foulants towards the bulk solution.

A simple method is proposed in Fig. 4 to illustrate the blocking/cake and reversibility of membrane fouling during operation. The true resistance of a fouled membrane was classified into membrane resistance (R_m), blocking resistance (R_b) and cake resistance (R_c) (left side of Fig. 4). The membrane resistance (R_m) can be estimated by measuring the clean water flux of a new membrane. However, from the TMP vs. t curve (Fig. 2), only the initial resistance immediately after backwashing (R_{bw}), reversible blocking resistance (R_{reb}) and observed cake resistance (R_{oc}) could be

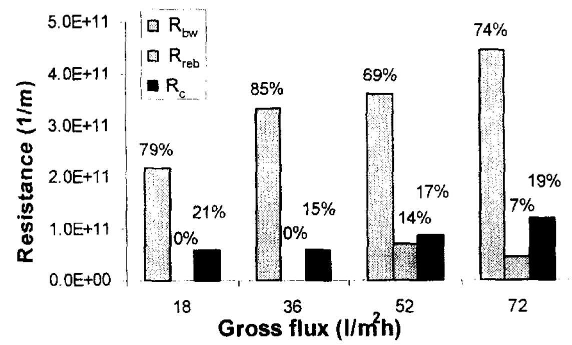

Fig. 3. Resistance due to blocking and cake as a function of gross flux.

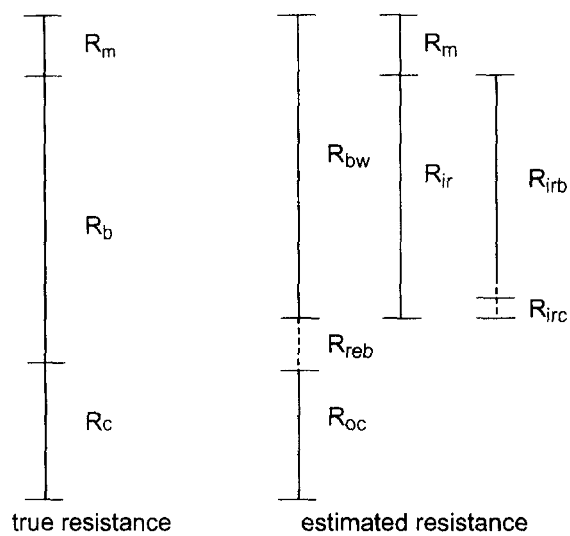

Fig. 4. Estimating blocking and cake resistance in a filtration cycle.

identified (right side of Fig. 4). Actually, the initial resistance immediately after backwashing (R_{bw}) included not only the clean membrane resistance (R_m), but also the resistance due to irreversible fouling (R_{ir}), i.e. fouling which could not be removed by simple hydraulic backwashing. The irreversible fouling (R_{ir}) was mainly due to the irreversible blocking resistance (R_{irb}) accumulated from previous filtration cycles. However the magnitude of irreversible cake resistance (R_{irc}) was marginal compared with the total irreversible fouling (R_{ir}) resistance, since the accumulated cake was easy to remove by a combination of backwashing and crossflushing. Thus, the observed cake resistance (R_{oc}) was very close to the true cake resistance (R_c).

If membrane resistance (R_m) and irreversible cake resistance (R_{irc}) is negligible, which may be especially true for fouled membranes, the sum of the initial resistance immediately after backwashing and the reversible blocking resistance (R_{bw} + R_{reb}) may give a reasonable estimation of the total blocking resistance (R_b). As a result, considering the large portion of (R_{bw} + R_{reb}) in Fig. 3, blocking resistance (R_b), especially irreversible blocking resistance (R_{irb}), resulting from blocking and adsorption of macro organic colloids/solutes during previous filtration cycles, was identified as the major resistance in this MBR pilot plant. Similar findings were reported by Bouhabila et al. [5].

3.3. The effect of backwashing interval

Obviously, cleaning frequency (e.g. backwashing) may affect the extent of pore blocking and cake filtration in membrane filtration systems. In Fig. 5, two different backwashing intervals are compared (BW once every 300 s and once every 600 s). The extension of the backwashing interval from 300 s to 600 s increased cake resistance (R_c) dramatically from 8.6×10^{10} 1/m to 3.6×10^{11} 1/m (17%–45% of total resistance), however both the initial resistance immediately after backwashing (R_{bw}) (mainly irreversible blocking resistance) and the reversible blocking resistance (R_{reb}) showed only a marginal increase (3.6–3.8×10^{11} 1/m and 6.9–7.1×10^{11} 1/m, respectively).

The fact that the initial resistance immediately after backwashing (R_{bw}) and the reversible blocking resistance (R_{reb}) were similar indicated that there was hardly any difference in backwashing efficiency for a backwashing intervals of 300 s and 600 s, which was probably due to the moderate flux of 52 l/m^{2}h. On the contrary, much higher cake resistance was observed at a backwashing interval of 600 s compared with 300 s (3.6×10^{11} compared with 8.6×10^{10} 1/m), and was attributed to the build-up of a thicker cake layer when the backwashing interval was extended. Therefore, in addition to pore blocking, cake filtration also played an important role when the backwashing interval was extended (longer filtration cycle). However, the cake was reversible and could be successfully removed by backwashing and sludge/air crossflushing.

3.4. Predicting chemical cleaning frequency

Backwashing is not always effective in removing all foulants. Resistance that cannot be

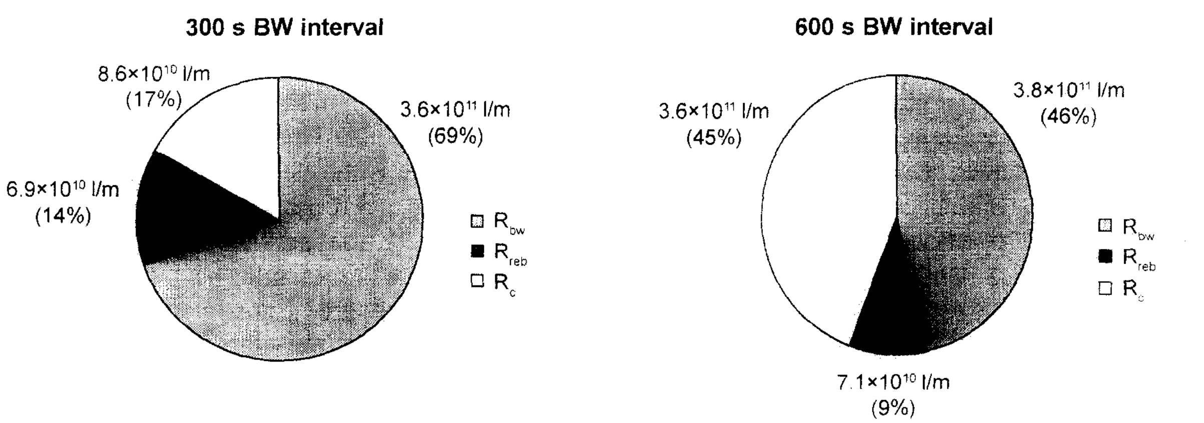

Fig. 5. Comparison blocking and cake resistance with two backwashing intervals (gross flux 52 l/m^{2}h).

removed by hydraulic cleaning is generally defined as irreversible fouling and usually requires intensive cleaning, for example with chemicals. Chemical cleaning frequencies depend on the accumulation rate of irreversible resistance. In general, extensive tests for a long period of operation have to be carried out to estimate the rate of increase of irreversible resistance. Herewith, a simple method was developed using consecutive filtration/backwashing cycles at gross fluxes of 41 and 69 l/m^{2}h (90 min for a total of 9 cycles) to predict chemical cleaning frequency.

The initial resistance immediately after backwashing (R_{bw}) at a gross flux of 41 and 69 l/m^{2}h are plotted in Fig. 6. The resistance after backwashing (R_{bw}) increased with time (number of cycles) due to irreversible fouling. The rate of increase in R_{bw} was estimated from the slope of the curve assuming a linear relation between R_{bw} and filtration time (cycle number). For both fluxes, the initial resistance after backwashing (R_{bw} of a clean membrane) was assumed to be 3×10^{11} 1/m (relative clean membrane after chemical cleaning) and the maximum allowable R_{bw} was assumed to be 2×10^{12} 1/m (corresponding TMP was 0.3 and 0.5 bar for fluxes of 41 and 69 l/m^{2}h respectively). As a result, 3.5 d would be required to reach the maximum allowable R_{bw} at a gross flux of 41 l/m^{2}h; and 1.2 d at a gross flux of 69 l/m^{2}h (details summarised in Table 1). The actual chemical cleaning frequency of this pilot plant was once per week at a gross flux of 40 l/m^{2}h.

Table 1
Prediction of chemical cleaning frequency

Gross flux, l/m^{2}h	41	69
Rate of increase of initial membrane resistance, 1/(m·min)	3.4×10^8	1.0×10^9
Initial resistance after backwashing (R_{bw} of a clean membrane)	3×10^{11}	3×10^{11}
Maximum allowable R_{bw}, 1/m	2×10^{12}	2×10^{12}
Filtration time (chemical cleaning interval), d	3.5	1.2

The deviation between the prediction and actual data was probably due to the limited duration of the pilot tests and the assumption of a linear increase in initial resistance immediately after backwashing (R_{bw}) with filtration cycle/time. According to Table 1, 68% increase in gross flux (41–69 l/m^{2}h) almost tripled the chemical cleaning frequency (from once per 3.5 d to 1.2 d), which indicated the critical role of flux in the reversibility of membrane fouling. The increase in irreversible fouling at 69 l/m^{2}h was not only due to the increased amount of foulants forced onto the membrane, but also due to the reduced effectiveness of backwashing (too short duration or too low backwashing flux). Compared with the chemical cleaning frequency of other MBR systems in Table 2 (from once per week up to no chemical cleaning within 371 d), this pilot plant

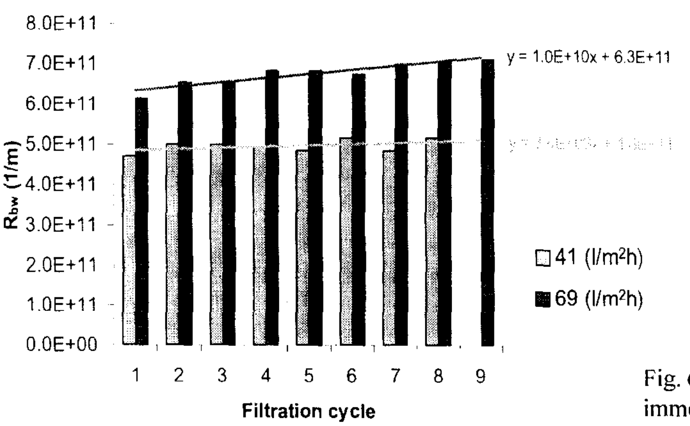

Fig. 6. Estimation of the rate of increase in initial resistance immediately after backwashing.

Table 2
Chemical cleaning frequency data in MBR systems [5, 14–17]

Source	Flux, l/(m^{2}h)	Configuration	Backwashing	Chemical cleaning frequency
Murakami et al. [14]	16.7	Submerged	No	No CC within 140 d
Ueda and Hata [15]	19.6	Submerged	No	No CC within 371 d
Bouhabila et al. [5]	7–27	Submerged	Yes	20 d
Côté et al. [16]	35	Submerged	Yes	Once per week maintenance CC
Cicek et al. [17]	n.a.	Side stream	n.a.	Once per week

required frequent chemical cleaning, which indicated severe irreversible fouling. Irreversible fouling in MBR systems may be due to the adsorption of macro organic matter (solutes and small colloids of a size comparable or less than that of the membrane pores). The high crossflow velocity (1 m/s) and liquid/air slug flow in this membrane module may have generated excess turbulence, causing large flocs to break and consequently, more small flocs and macro organic matter were produced. Therefore, the shear stress in membrane modules should be optimised, so that on the one hand it does not cause bioflocs to break, and on the other hand, it is sufficient to prevent rapid deposition of large amounts of foulants on the membrane surface. Optimisation of the biology may reduce the amount of solutes and macro organic matter entering the membrane system, and consequently reduce pore blocking and irreversible fouling.

4. Conclusions and recommendations

- At a flux of 52 l/m^{2}h, pore blocking was completed in 8 s, and was attributed to organic solutes and colloids in the feed water (activated sludge). The transition stage (8–100 s) following the blocking stage was characterised by a slower increase in TMP. Finally, cake filtration dominated after 100 s (100–600 s) with a linear increase in TMP.
- Pore blocking and cake filtration (at longer backwashing intervals) played important roles in membrane fouling. However, pore blocking was not completely reversible by backwashing and may partially remain contributing to irreversible fouling, and increase the initial resistance immediately after backwashing (R_{bw}). Backwashing was more successful in the removal of the accumulated filter cake.
- A simple method to identify both irreversible and reversible blocking resistance and filter cake resistance was proposed. Blocking resistance (R_b), especially irreversible blocking resistance (R_{irb}), resulting from blocking and adsorption of macro organic colloids/solutes during previous filtration cycles, was identified as the major resistance in this MBR pilot plant.
- The prediction of chemical cleaning frequency was not very accurate, but it indicated the critical role of flux in irreversible fouling and chemical cleaning frequency.

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