

Accepted Manuscript

Title: Membrane life estimation in full-scale immersed membrane bioreactors

Authors: D.F. Ayala, V. Ferre, S.J. Judd

PII: S0376-7388(11)00180-3
DOI: doi:10.1016/j.memsci.2011.03.013
Reference: MEMSCI 10586

To appear in: *Journal of Membrane Science*

Received date: 25-1-2011
Revised date: 3-3-2011
Accepted date: 7-3-2011



Please cite this article as: D.F. Ayala, V. Ferre, S.J. Judd, Membrane life estimation in full-scale immersed membrane bioreactors, *Journal of Membrane Science* (2010), doi:10.1016/j.memsci.2011.03.013

This is a PDF file of an unedited manuscript that has been accepted for publication. As a service to our customers we are providing this early version of the manuscript. The manuscript will undergo copyediting, typesetting, and review of the resulting proof before it is published in its final form. Please note that during the production process errors may be discovered which could affect the content, and all legal disclaimers that apply to the journal pertain.

Membrane life estimation in full-scale immersed membrane bioreactors

D.F. Ayala^a, V. Ferre^a, S.J. Judd^b

^aKubota Membrane Europe, W6 8JA, London, UK

^bCentre for Water Science, Cranfield University Bedfordshire, UK; corresponding author, s.j.judd@cranfield.ac.uk

Abstract

Limited quantitative available information exists regarding the robustness of membranes used in immersed membrane bioreactor (iMBR) applications, and no information on the relative contribution of the two main membrane failure modes of permeability and integrity loss. Such information is crucial given the significance of membrane replacement costs to the viability and sustainability of iMBRs.

Measurements of membrane permeability and integrity have been made on flat sheet (FS) membrane cartridge sampled from six existing full-scale iMBRs. The recovered membrane permeability (i.e. the permeability recorded following chemical cleaning) revealed all plants operating for less than six years to have a clean water flux greater than that of the virgin membrane cartridge. A lower permeability was recorded only for the oldest plant (eight years of operation), and this was attributed to high levels of phosphate scale fouling. Assigning a linear trend between membrane permeability loss and operation time indicated the recovered membrane permeability to reach a threshold minimum value for virgin membrane cartridge after ~7 years of operation.

The membrane cartridge integrity, as measured by the strength of the weld at its perimeter, correlated with the total volume of water permeated per cartridge, the total weight of NaOCl cleaning reagent to which the panel has been exposed, and the mean volume of water permeated per cartridge between chemical cleans. In this case, the linear trend indicated a membrane life equating to ~900 m³ per panel or exposure to a total of 1.3 kg NaOCl before the threshold minimum welding strength for membrane cartridge was reached. As with the permeability data, only for the oldest plant was the measured mean membrane integrity below the threshold.

The data indicate membrane life for FS iMBRs to exceed six years, on the basis of both permeability and integrity, and may ultimately be limited only by the irreversible deposition of inorganic scale, rather than operation time.

Keywords:

Flat sheet membrane; membrane bioreactor; integrity; membrane life; inorganic scaling

Abbreviations and notation

ADF	Average Daily Flow (m^3/d)
ABS	Acrylonitrile-butadiene-styrene (copolymer)
CIP	Clean in place
CWF	Clean water flow (rate)
FS	Flat sheet (membrane)
HRT	Hydraulic retention time (h)
iMBR	Immerse membrane bioreactor
J'_{cw-0}	Clean water flux before CIP, corrected to 20°C - CWF test (LMH)
J'_{cw-i}	Clean water flux after CIP i, corrected to 20°C - CWF test (LMH)
J'_{cw-v}	Clean water flux (minimum) standard for virgin membrane corrected to 20°C - CWF test (LMH).
LMH	$\text{L} (\text{m}^2 \text{h})^{-1}$
PDF	Peak daily flow
PVC	Polyvinyl chloride
PST	Peel strength test
r	Pearson coefficient
SAD_m	Specific aeration demand per membrane surface (Nm^3/m^2)
SAD_p	Specific aeration demand per m^3 of permeate (Nm^3/m^3)
SRT	Solids retention time (d)
TMP	Trans-membrane pressure (bar)
WwTP	Wastewater treatment plant
$\Delta J'_{cw-i}$	Flux recovery after CIP i – CWF test (%)
σ	Membrane weld strength, PST (N/mm)
σ_{ms}	Minimum recommended weld strength, PST (N/mm)

1 Introduction

Immersed membrane bioreactor (iMBR) technology forms the basis of many industrial and municipal wastewater treatment installations where a high quality effluent is demanded. However, its more widespread implementation, and its sustainability as a viable wastewater reuse technology, is limited by its relatively high costs [1]. These relate primarily to energy demand, and aeration energy in particular [2], and membrane replacement [3]. Given that the technology itself is only ~20 years old, long-term operational experience - and membrane performance data in particular - is limited and is, in case, rarely reported in detail for the more established installations, i.e. more than 4-5 years old.

From anecdotal evidence regarding membrane replacement [1, 3-4], it appears that FS membrane robustness is generally promising. The most established municipal flat sheet (FS) iMBR plant globally - Porlock in the UK - had only 6.4% of its original cartridges replaced after 10 years of operation [4]. The majority of the failures at this site were due to preventable external factors, such as handling errors. At the same site, the average permeability of the membranes declined almost insignificantly over the ten year period, the initial permeability being recovered by chemical cleaning.

Whilst the above study is illuminating, it refers only to a single plant which has operated under relatively benign conditions, i.e. a low mean flux and high membrane

aeration rate. There is no publically-accessible wide-ranging study encompassing a number of installations which could establish whether the trend reported for Porlock extends across other MBR-based municipal WwTPs. Crucially, there is no indication as to which operating parameters may play a significant role in determining membrane life.

The aim of the current study was thus to determine the impact of plant operation and maintenance characteristics, as well as plant age per se, on the key physical membrane characteristics of membrane permeability and integrity for a range of established MBR installations. Membrane integrity was determined using a bespoke test of the membrane seal strength, permeability from a conventional constant head method. As with the plant at Porlock, all membranes were of the same type (*Kubota 510* cartridges), the membrane being based on a hydrophilicised polyethylene microfilter of 0.4 μm and the plate of rigid ABS plastic [1].

2 Materials and methods

2.1 Plant characteristics

All six full-scale municipal FS iMBR installations selected employed the same flat sheet (FS) cartridges (*Kubota 510*), these being 0.8 m^2 in area and having dimensions of 1 m length x 0.5 m x width [1]. Operation and maintenance (O&M) data from these plants (Table 1) were obtained through the completion of a questionnaire by the plant operators. A sample of five membrane cartridges from each plant were taken and each examined for integrity, using a proprietary peel strength test (PST), and permeability using the clean water flow rate test (CWF). Data were then statistically analysed by Pearson correlation and linear regression analyses.

Table 1: MBR plants, general information

The mean operating flux across all six plants ranged from 16 to 26 LMH, and the specific aeration demand SAD_m according to the manufacturer's design specifications of 0.53 and 0.75 $\text{Nm}^3/(\text{m}^2\text{h})$ for double or single deck modules respectively [1]. Chemical cleaning with sodium hypochlorite solutions –between 15 and 30g of NaOCl per cartridge - had been performed for all the MBRs studied at periods between 2 and 6 months. For Plants 1 and 2, supplementary hydrochloric acid cleaning (0.9%) was performed every 3 months. For Plants 3 and 4, citric acid (0.8% - 1%) was used annually and Plants 5 and 6 had been operated without acid cleaning.

2.1.1 Clean water flow rate test

The CWF rate test was used to determine the clean water flux ($J'_{\text{cw-i}}$) through a single membrane cartridge at a constant pressure of 0.049 bar, with data temperature-adjusted to 20°C according to the standard viscosity correction equation. Membrane cartridges were tested before and after a series of chemical cleans performed with sodium hypochlorite, citric acid and hydrochloric acid solutions using the bespoke CWF equipment (Fig. 1). Values for the solution concentration and cleaning duration used

for the tests (Table 2) were typical of those normally employed for cleaning in place (CIP) of FS iMBRs. Chemical cleaning of the individual cartridges were performed by disconnecting the permeate tube and injecting the 3L of cleaning solution directly into the cartridge under gravity, thereby simulating the full-scale procedure.

Figure 1: CWF rate test illustration

Table 2: CIP procedure for CWF test

The clean water flux recovery ($\Delta J'_{cw-i}$) after each CIP is then defined as:

$$J'_{cw-i} = \frac{J'_{cw-i}}{J'_{cw-0}}$$

where $\Delta J'_{cw-i}$ is the % flux recovery after CIP i (where $i = 1, 2$ or 3), J'_{cw-i} the measured flux after CIP i , and J'_{cw-0} the flux prior to the CIP.

2.1.2 Peel strength test (PST) of seals

The membrane cartridges comprise two membrane sheets welded to each side of a central ABS plate. The PST is a simple proprietary destructive method used to assess the mechanical strength of the weld, which is widely known to represent the weakest structural component of the membrane cartridge [1]. The test employs a digital force gauge and a bespoke motorized tensile strength testing device, with the tensile force being applied orthogonally to the ABS plate. The PST is a simple destructive test where the force at which the weld yields is determined in N/mm. Four regions of the membrane panel were tested: two at the top and two at the sides on either side of the panel.

3 Results and discussion

3.1.1 Membrane replacement from mechanical failure

The calculated percentage of membranes replaced per year of operation in each plant was found to vary from 0% to 4.6%. Four of the six MBR installations had replacement cartridges, with the most common cause being damage by welding sparks during installation which accounted for 60% of the total membranes replaced (Fig. 2). Of the other causes (membrane cartridge weld failure, membrane breakage due to solids bypassing screens, mechanical failure of ABS plate, PVC manifold or hydraulic connection of the membranes, and other unknown causes), only 38% could be attributed to failure of the membrane cartridge itself since failure due to bypassing of screens is clearly a failure of the system engineering rather than the membrane cartridge. Of this figure 34% was due to membrane panel welding failure.

Figure 2: Membrane replacement causes

Overall, the average proportion of membrane cartridges replaced per year for reasons other than welding sparks was 0.2%, the median number being 0.03%. Cartridge replacement directly due to the compromising of membrane or cartridge integrity through normal operation was therefore small. The limited, and largely anecdotal, available published membrane life data suggests that extensive membrane replacement in MBRs is required mainly where membrane maintenance is insufficiently rigorous, but is also influenced by the feedwater characteristics, cleaning protocols and pre-treatment efficacy [5], and so by factors contributing to irreversible or irrecoverable fouling [1, 6-7]. It may also be influenced by inorganic scaling, since this may not always be suppressed through conventional chemical cleaning [8]. However, whilst it might be expected for irrecoverable fouling to be the primary contributor to ultimate membrane replacement, this was not the case for any of the plants surveyed in the current study, corroborating previous findings [3-4].

3.1.2 Membrane permeability

Figure 3 shows the flux recovery ($\Delta J'_{cw-i}$) after each CIP as determined by the CWF test. The observed mean value of $\Delta J'_{cw-3}$ (i.e. the total flux recovery after the completion of CIP 3) was 193%. In every case, the measured flux recovery was higher for the NaOCl clean (CIP 1) than for the two acid cleans combined (CIP 2 + CIP 3), the ratio values ranging from 1.89:1 to 4.77:1 (2.87:1 on average). This is presumed to roughly indicate the relative importance of organic over inorganic fouling.

Figure 3: Flux recovery following consecutive chemical cleans (CWF test)

Overall membrane permeability loss from virgin conditions was assumed to be reflected in ratio of the CWF measurement following CIP 3 (J'_{cw-3}) and that of the threshold minimum flux for a virgin membrane cartridge (J'_{cw-v}). For all but one of the plants the CIP recovered the membrane permeability to a level above that of the threshold minimum value (Fig. 4), the exception being Plant 4. Since J'_{cw-v} represents the minimum value considered acceptable for use, as stipulated by the supplier, it is not unusual for the measured permeability to exceed this value.

Plant 4 represents the oldest of the plants surveyed (8 years in operation) and treats a feedwater comprising 70% municipal and 30% dairy wastewater. Visual inspection of the cartridges indicated irrecoverable scaling from deposited ferric salts, used for chemical phosphorus removal at this plant and imparting a characteristic red colour which was retained even after CIP 3. This plant also provided the lowest CIP 1:CIP 2,3 flux recovery ratio of 1.89, indicating the relative significance of inorganic fouling. Given that no acid cleanings were performed for the first few years of plant operation at this site, the significant irrecoverable fouling is unsurprising. Insidious scaling of this nature is not readily eliminated through conventional chemical cleaning [8].

Figure 4: Clean water flux results (CWF test)

The impact of membrane operation time, and plant design and operating parameters on recovered membrane permeability ratio (i.e. J'_{cw-3}/J'_{cw-v}) was appraised. Key parameters considered included total membrane filtration time, total permeate discharged per cartridge, permeate discharged between chemical cleans, permeate discharged between physical cleans (i.e. between relaxation periods), mean flux, mean flux per time in operation, specific aeration demand (SAD_m , SAD_p), total membrane air provided, membrane tank MLSS concentration, solids retention time (SRT), chemical cleaning frequency, phosphorus removal, and events such as clogging episodes. For the most part, no correlation was evident other than for those parameters indicated below.

A correlation of J'_{cw-3}/J'_{cw-v} against operation time and total permeate volume indicates an approximately linear decline with age. A Pearson correlation test indicated that for the municipal MBRs there is a significant negative linear correlation between clean membrane flux and the membrane operation time ($r_{xy}=-0.87$) (Fig. 5). However, the decline is shallow, with only a 6% annual decline in baseline permeability suggesting that the membranes may operate for around seven years before the permeability of the chemically cleaned membrane decreases to that of the virgin membrane cartridge. Given that these data refer to the standard chemical clean shown in Table 2, it is likely that the membrane life could be further extended using a more aggressive chemical clean.

Data suggest that chemical precipitation of phosphorus may negatively impact on membrane permeability. The mean J'_{cw-3}/J'_{cw-v} ratio of those MBRs operating with chemical removal of phosphorus was slightly lower, at 110%, than the mean value for the remaining plants (124%). However, since one of the plants employing chemical P removal was also the oldest this may not be a significant finding. On the other hand, phosphorus scales are known to be tenacious [8] - probably demanding more regular acid cleaning to control fouling than that used for the two plants concerned.

Figure 5: Membrane age vs. flux of the chemically cleaned membrane (CWF test)

3.1.3 Membrane weld strength

The results of the PST for the 6 plants are shown in Figure 6. The weld strength (σ , in N/mm) is normalised against the minimum strength recommended by the manufacturer (σ_{ms}). Results indicate that all but one of the plants had an average σ value greater than the σ_{ms} , but that two of the six plants yielded some individual σ values below this threshold. Whilst the sample is small and the data scatter for each individual cartridge broad, there is some indication of a correlation, with a Pearson coefficient value r of 0.91, between welding strength and total permeate produced per cartridge (Fig. 7). No correlation was evident between σ and either the flux or membrane operation time, implying that the weld strength may relate more specifically to usage than simply operation time. According to this analysis, approximately 900 m³ of water per cartridge can be filtered before the average seal strength decreases to below the

threshold value. This equates to 6.4 years of operation at a mean net flux of 20 LMH, or 8.5 years at 15 LMH.

Figure 6: Results from the PST for the different MBRs

Figure 7: Total treated water per cartridge vs. membrane welding strength

A strong negative correlation ($r = -0.84$) was also found between σ and the calculated total weight of NaOCl per cartridge used for cleaning (Fig. 8). Chemical oxidants such as NaOCl are known to cause delamination of the membrane at higher concentrations [9], and are likely to contribute to membrane deterioration generally at high concentrations. It has also been shown, for FS membranes, that frequent (monthly) chemical cleaning with more dilute NaOCl solutions sustains higher mean membrane permeabilities and employs 30% less reagent than less frequent (quarterly) cleaning using higher-strength reagents [10]. It would appear that such an approach, i.e. the use of maintenance cleaning, may not only improve permeability but also be less deleterious to membrane integrity. This is supported by the correlation, of similar strength ($r = -0.88$) to that of Figure 8, between weld strength and the average filtrate volume between chemical cleans (Fig. 9), though this may simply be an extension of Figure 7. As with the permeability data, whilst other parameters were analysed no other clear correlations with weld strength were evident.

Figure 8: Total weight of NaOCl per cartridge dosed over the life of the plant vs. membrane welding strength

Figure 9: Weld strength vs. average permeate volume per panel per CIP

4 Conclusions

A study of membrane cartridges taken from six operating full-scale MBRs revealed no statistically significant impact from any key operation and maintenance parameter on the permeability and integrity of sampled membrane cartridges. Parameters which may have been expected to have an influence, including flux, MLSS concentration and the incorporation of acid chemical cleaning, were not demonstrated to influence membrane cartridge performance with respect to mechanical robustness and chemically recovered membrane permeability. Of the correlations that were produced:

- a. The recovered membrane permeability (i.e. the membrane permeability recorded following chemical cleaning) for all plants operating for less than six years was found to exceed the clean water permeability of the virgin material. Only the oldest plant (eight years operation) provided a membrane permeability below the threshold, and this was attributed to high levels of fouling by phosphate scale deposition. A linear correlation between membrane permeability loss and operation

time was determined, indicating that the average period of operation for the cleaned membrane permeability to reach the threshold value was around seven years.

- b. The membrane cartridge integrity, as determined by the strength of the welding at its perimeter (this being the weakest part of the cartridge) appears to correlate with the total volume of water permeated per panel, the total weight of NaOCl to which the cartridge has been exposed, and/or the mean volume of water permeated per cartridge between chemical cleans. According to linear regression analysis, the membrane weld strength may deteriorate after 900 m³ per cartridge of permeate, or after 1.3 kg NaOCl dosed (per cartridge), to below the threshold minimum welding strength for a virgin membrane cartridge set by the manufacturers. As with the permeability data, only for the oldest plant was the mean membrane integrity below the threshold, and this plant was also subject to inorganic scaling.

According to the integrity data, membrane life for flat sheet MBRs based on integrity and permeability exceeds six years for operation at a mean net flux of 20 LMH. This would seem to be somewhat lower than the membrane life reported for other plants, and may reflect the extent of irreversible deposition of inorganic scale, rather than membrane operation time itself. Clearly, given the significant contribution of membrane replacement to overall MBR operating costs, it is important to explore this further by applying the same tests to existing MBR installations which (a) have been operating for more than 6 years and/or (b) have been subject to inorganic scaling to establish the relative influence of operation time and inorganic scaling on membrane life.

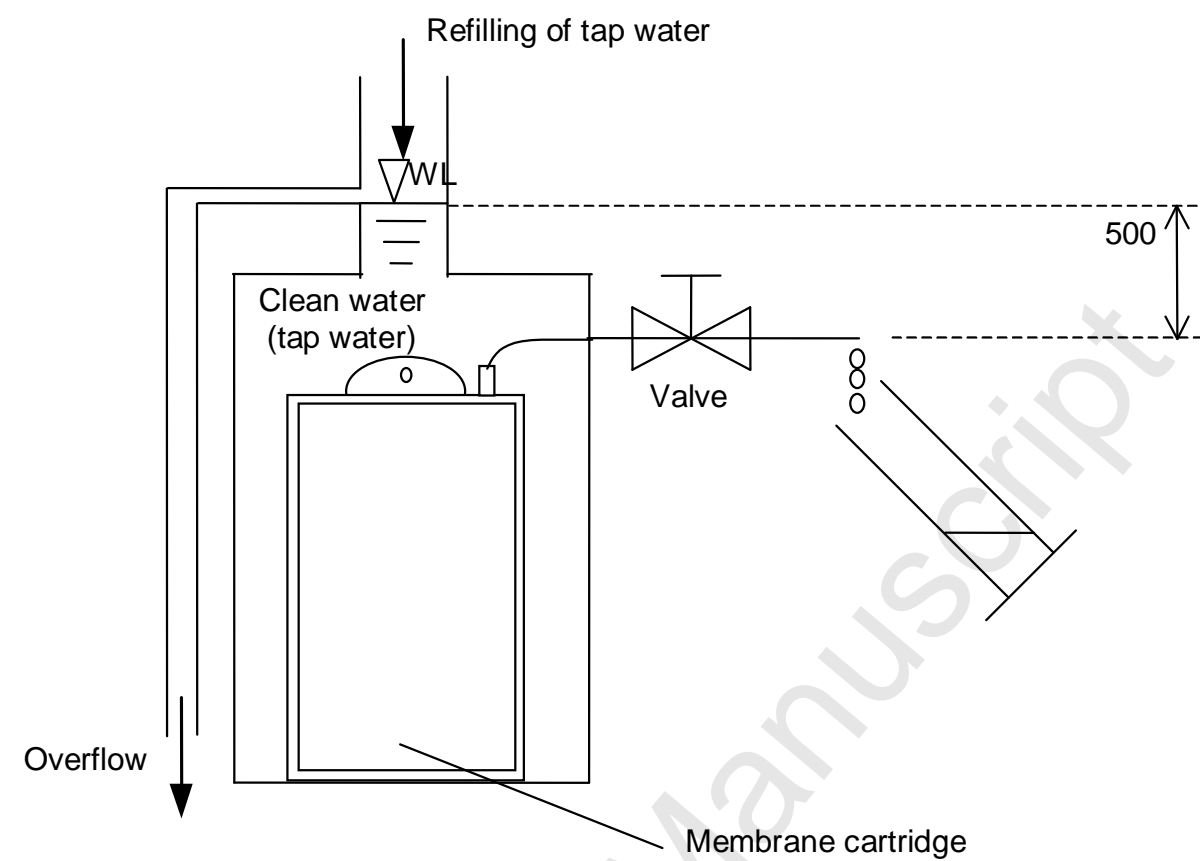
Acknowledgments

The authors gratefully acknowledge Kubota Membrane Europe for funding this research. They also express their gratitude to the engineering companies, industries and MBR operators that decided to participate in this project.

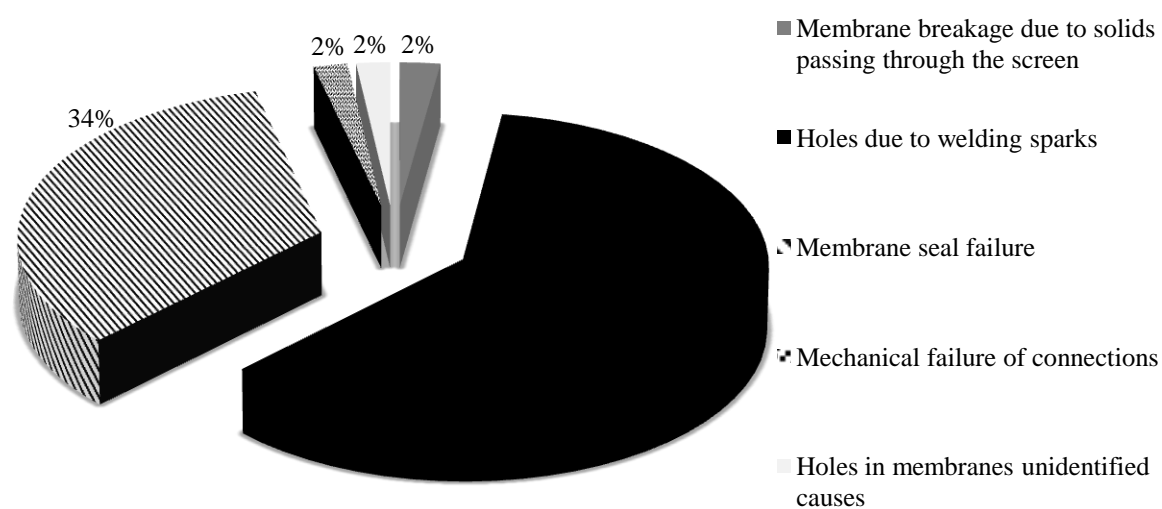
References

1. Judd, S., and Judd, C. (2010). *The MBR Book* (2nd ed.), Elsevier, Oxford.
2. Verrecht, B., Judd, S., Guglielmi, G., Mulder, J.W., and Brepols, C. (2008). An aeration energy model for an immersed membrane bioreactor, *Water Res.*, **42**, 4761-4770.
3. Kennedy, S., and Churchouse, S.J. (2005) Progress in membrane bioreactors: new advances. *Proc. of Water and Wastewater Europe Conference*, Milan, June 2005.
4. Churchouse, S, Warren, S., and Floyd, M. (2008). Feedback from the Porlock MBR plant in its 10th year of operation: An analysis of the flux, effluent quality and membrane lifetime data to date. *Aachener Tagung Wasser und Membranen*. Aachen, Germany.
5. Asano, T., Burton, F.L., Leverenz, H.L., Tsuchihashi, R., and Tchnobanoglous, G. (2006). *Water Reuse Issues, Technologies and Applications*. Edited by Metcalf & Eddy. McGraw-Hill, 2006.

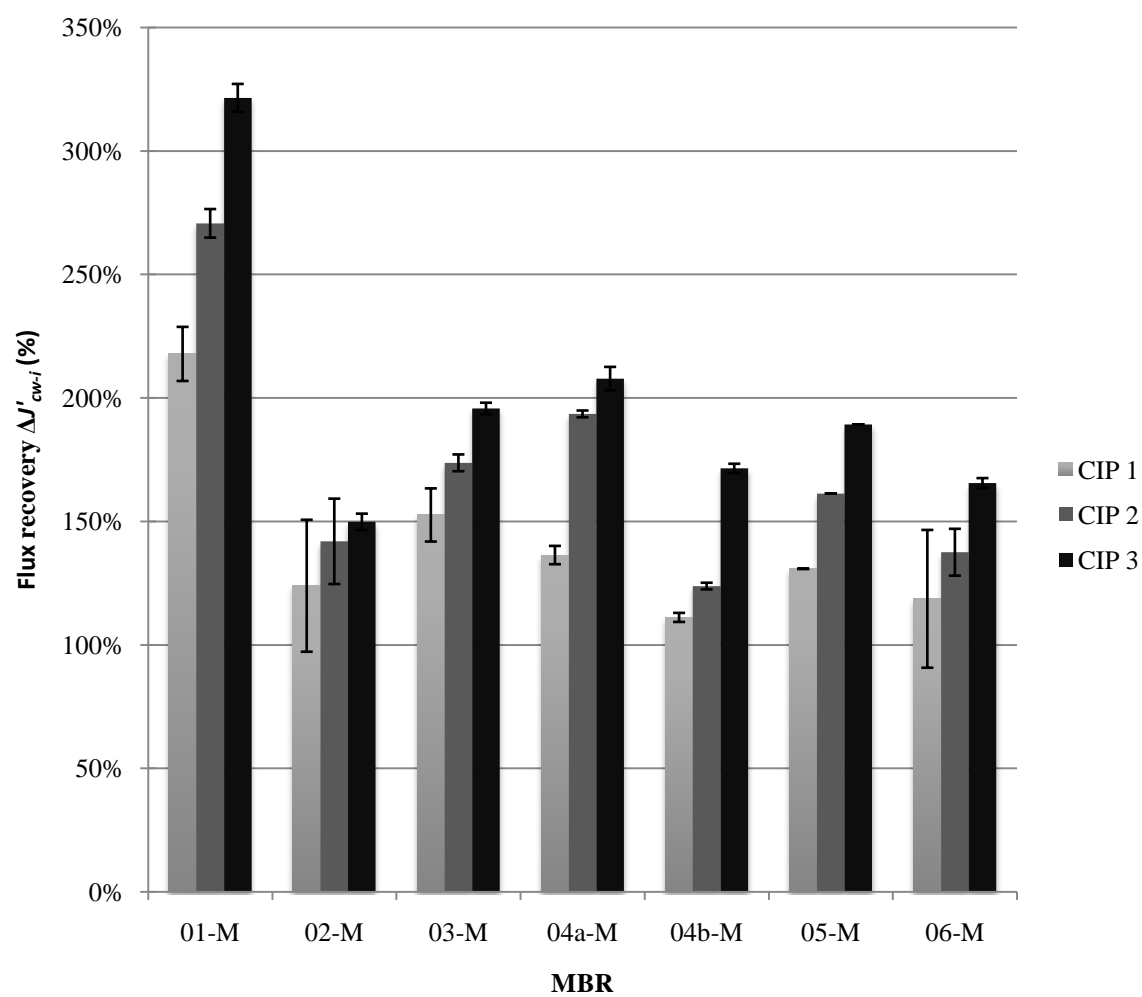
6. Judd, S. (2008). The status of membrane bioreactor technology. *Trends in Biotechnology* **26**(2), 109-116.
7. Radjenovic, J., Matosic, M., Mijatovic, I., Petrovic, M., and Barcelo, D. (2008). *Membrane Bioreactor (MBR) as an Advanced Wastewater Treatment Technology*. Vol. 5S, in *The Handbook of Environmental Chemistry: Emerging Contaminants from Industrial and Municipal Waste*, 37-101. Berlin: Springer Berlin / Heidelberg.
8. Meng, Fangang, So-Ryong Chae, Anja Drews, Matthias Kraume, Hang-Sik Shin, and Fenglin Yang. "Recent advances in membrane bioreactors (MBRs): Membrane fouling and membrane material." *Water Res.* **43** (2009): 1489-1512.
9. Le-Clech, P, A. Fane, and G. Leslie. MBR focus: the operators' perspective. *Filtration + Separation*, no. June (2005): 20-23.
10. Guglielmi, G., d Chiarani, D.P. Saroj, and Andreottola. Impact of chemical cleaning and air.sparging on the critical and sustainable flux in a flat sheet membrane bioreactor for municipal wastewater treatment. *Water Science & Techonology* **57**, no. 12 (2008): 1873-1979.



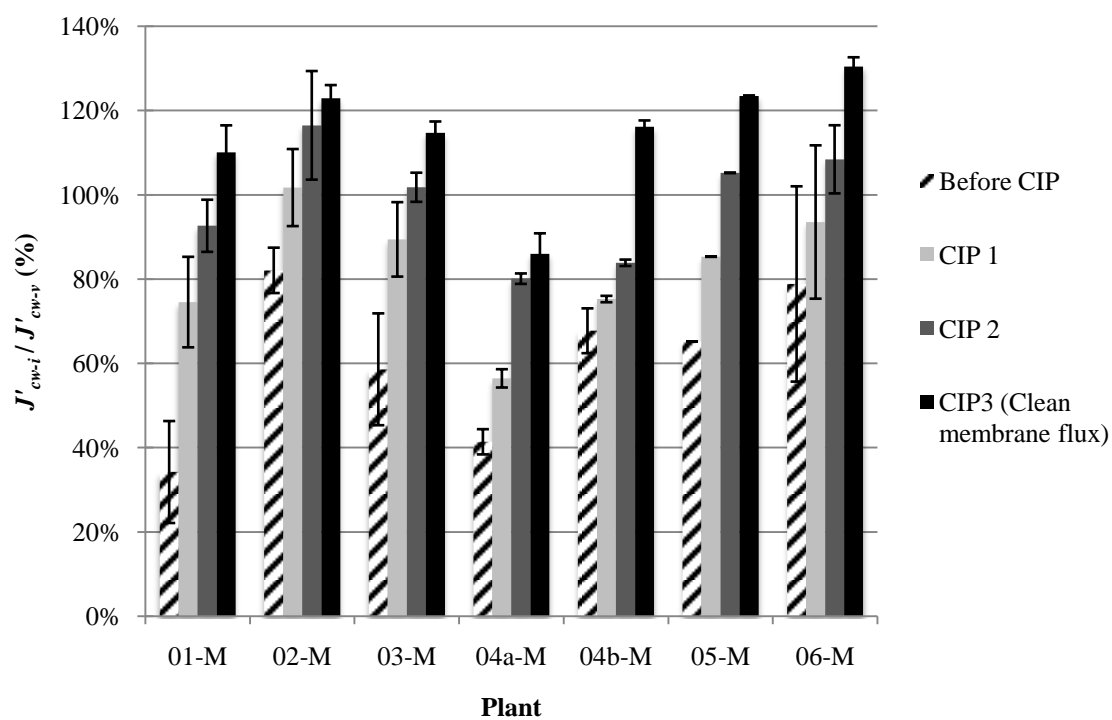
CWF rate test illustration



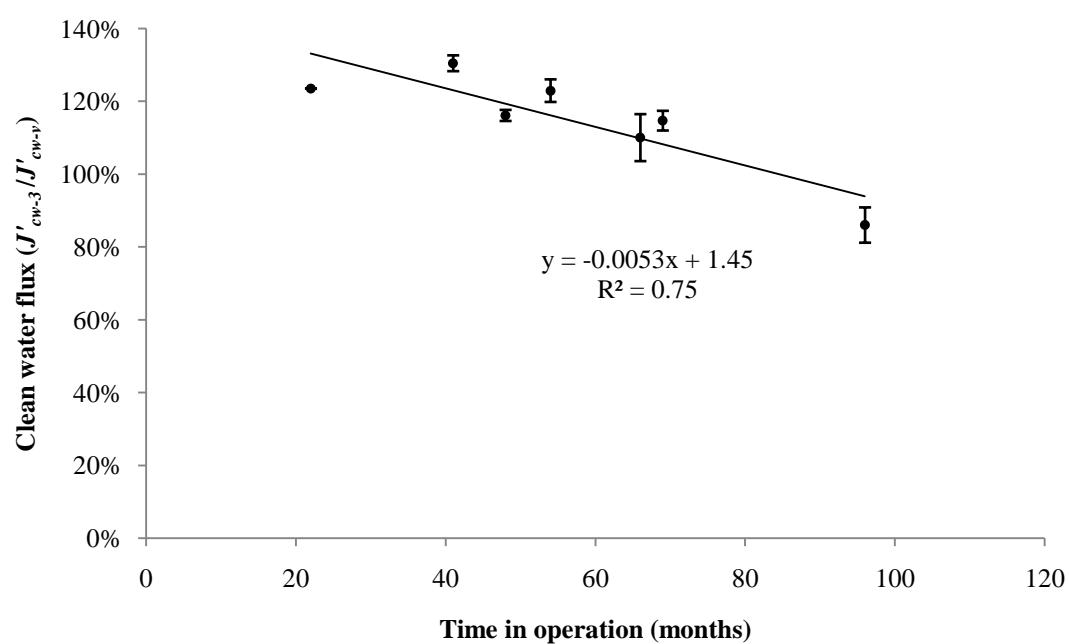
Membrane replacement causes



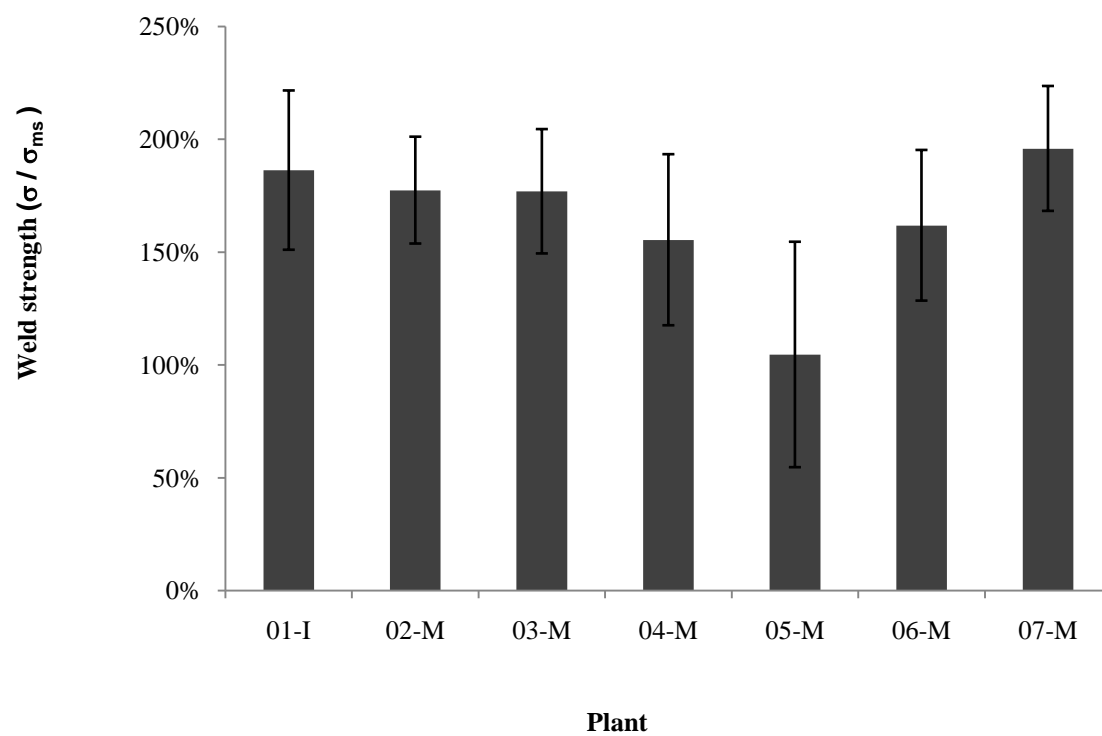
Flux recovery following consecutive chemical cleans (CWF test)



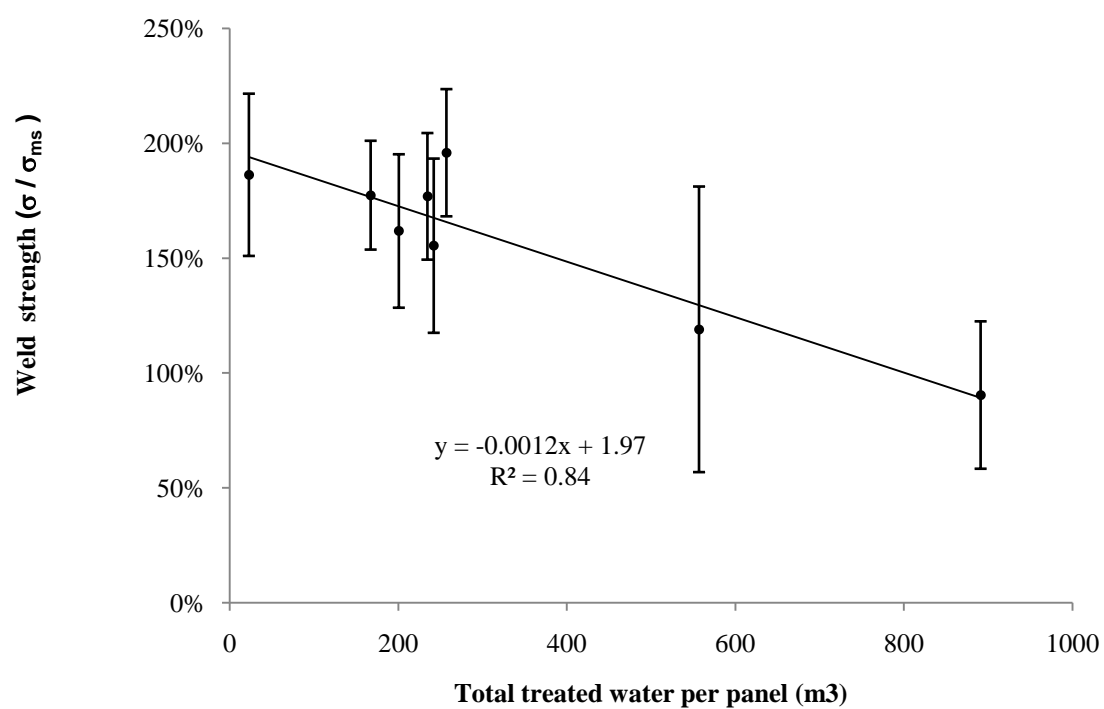
Clean water flux results (CWF test)



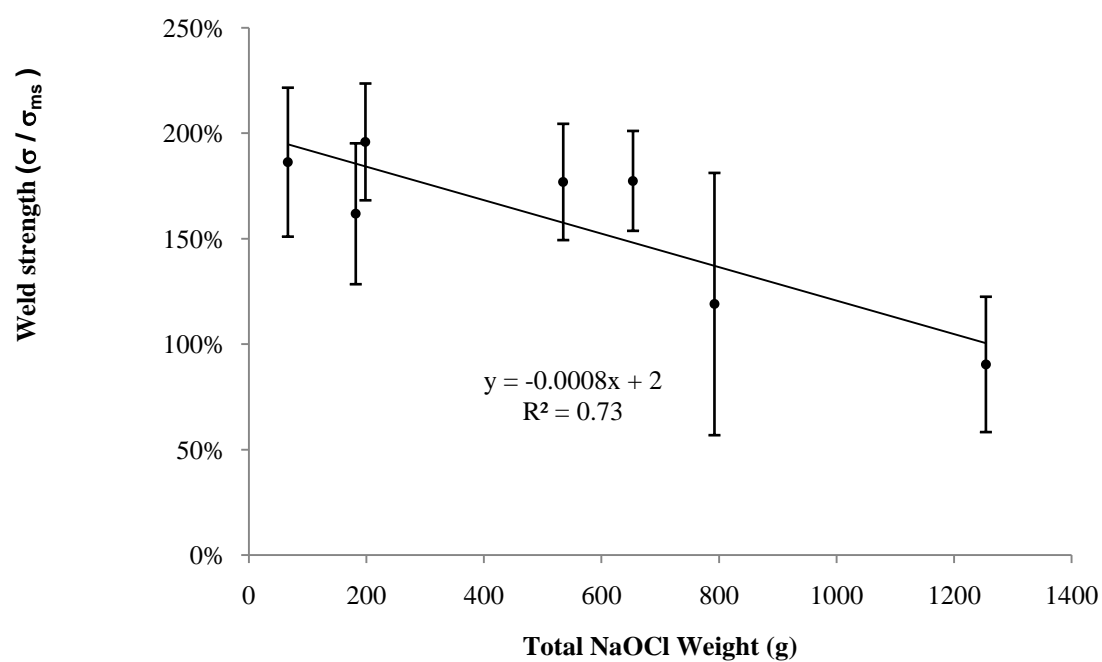
Membrane age vs. flux of the chemically cleaned membrane (CWF test)



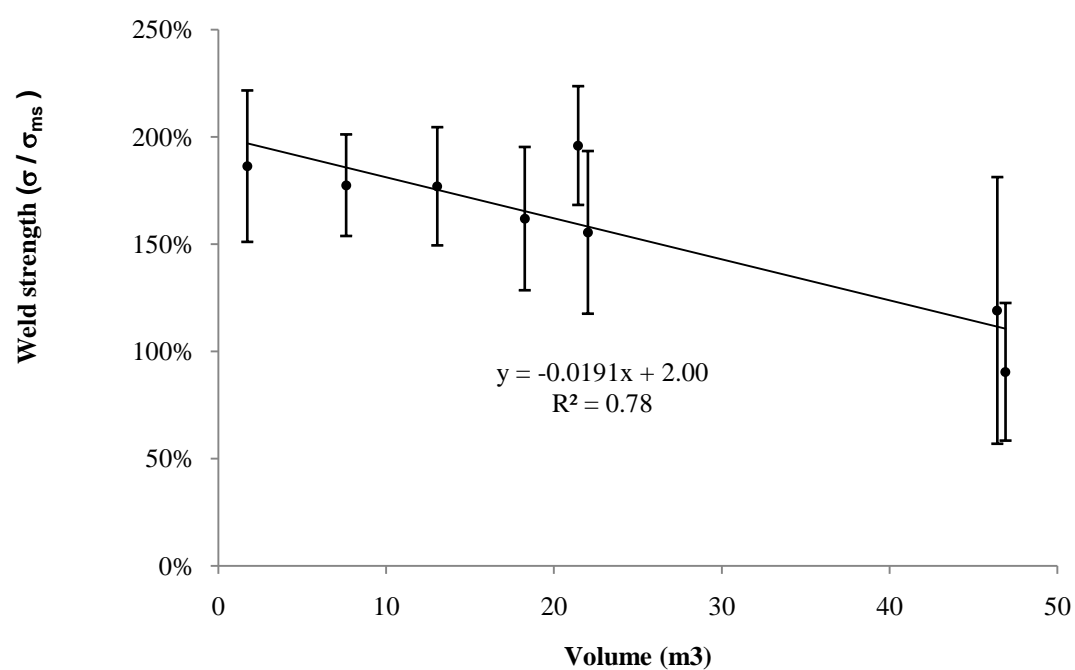
Results from the PST for the different MBRs



Total treated water per panel vs. membrane weld strength



Total weight of NaOCl dosed over the life of the plant v.s. membrane welding strength



Weld strength vs. average permeate volume per panel per CIP

Table 1: MBR plants, general information

Plant Ref.	Wastewater type	Commissioning date	ADF, m ³ /d	Aerobic tank HRT, h	SRT, d	MLSS, membr. tank, g/L	Chemical clean, NaOCl ^a	Ave. no. chemical cleans/y	Mem. Repl. (%)
1	Sewage	Dec-04	2,600	6.4	30	10-15	1.2 m ³ of 0.9%	3	0.3% ^d
2	Sewage	Dec-05	2,800	6.7	25-50	10-13	1.2 m ³ of 0.9%	3	18.2% ^e
3	Sewage	Sep-04	4,080	2.2	45-80	12	1.2 m ³ of 1%	6	0.04% ^f
4a, 4b ^b	70% Sewage, 30% Dairy ^c	Jun-02	2,930	12.2	70	10-15	0.6 m ³ of 2%	5	6.5% ^g
5	Sewage	Aug-08	1,000	4.9	20	13	1.2 m ³ of 0.5%	2	0.0%
6	Sewage	Jan-07	2,160	3.2	n.a.	17	1.2 m ³ of 0.5%	4	0.0% ^h

a. Volume and strength per module of 400 cartridges, other than Plant 4 which has modules of 200 cartridges

b. Cartridge operated since 2006 for ref 4b

c. Percentage based on hydraulic load ratio

Proportion of membrane failure type:

d. 100% replaced due to solids bypassing the screen

e. 2% replaced due to solids bypassing the screen. 98% replaced due to welding sparks damaging the membranes during installation

f. 100% replaced due to welding sparks damaging the membranes during installation

g. 90% seal failure, 5% frame or manifold failure, 5% membrane integrity failure

h. Membranes yet to be inspected or replaced; membrane integrity failure suspected

n.a. not available

Table 2: CIP procedure for CWF test

CIP	Chemical reagent	Concentration (%)	Duration (h)
1	Sodium hypochlorite	0.5	2
2	Citric acid	0.5	1
3	Hydrochloric acid	1	1