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Indirect effects of membrane configuration on MBR sludge filterability

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ABSTRACT

During a combined pilot-scale study, operating conditions were fixed for three different membrane configurations a multi tube, a hollow fibre and a flat sheet membrane type. Changes in activated sludge filterability were quantified depending on the membrane chambers, the permeate flux of extraction and the membrane chamber aeration. The aim of this study was to provide a better understanding of the interactions between MBR activated sludges and hydraulic circumstances depending on membrane configurations. Two mechanisms can be distinguished from this study: Due to practical arrangements, hydraulic conditions in each membrane unit are different, leading to different total suspended solids concentrations (TSS). These margins in TSS contents cause differences in filtration behaviour. The multi tube configuration did not affect the activated sludge filterability. The activated sludge filterability remained the same in spite of flux and air-lift velocity changes. The flat sheet and the hollow fibre configuration did affect the sludge filterability. The filterability improved significantly at high flux and at low air-lift velocity (from 20% to 90% of improvement). The TSS concentration phenomenon is likely to be responsible for the activated sludge filterability improvement. It might be due to floc structure modifications resulting in a better cake layer porosity. No correlations were found between SMP and filterability.

Keyword: Membrane configuration; Filterability; MBR; Pilot-scale

1. Introduction

Fouling remains the major research topic in the MBR field [1]. It is commonly accepted that fouling in MBR processes is mainly due to three factors: the activated sludge properties, the membrane properties and its operating conditions [2]. Activated sludge filterability is commonly linked with activated sludge properties as soluble microbial product (SMP) concentrations [3], SRT, TSS or fractionations as particulate, colloid and soluble parts [4, 5]. In these studies, SMP are still considered as a major foulant even if their contributions and the mechanisms involved are still not clearly understood [6]. Soluble and colloidal materials are assumed to be the major

contributors to the pore blocking mechanism whereas TSS content are considered to play a role within the cake layer mechanism [4].

Membrane types are mostly investigated in terms of pore size or membrane materials [7, 8]. Main observations have shown that membrane fouling is related mostly to the feedwater and activated sludge properties leading to non-consistent trends between pore sizes and fouling behaviours. In those cases, the common used comparison points are membrane fouling or permeability variations for given MBR activated sludges.

Finally operating conditions, especially coarse bubble aeration and permeate flux, are always regard as tuning parameters in order to prevent membrane fouling or membrane clogging [1810]. Even if Park *et al.*, (2005) [11] show that intense aeration may damage the floc

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structure and induce release of SMP, it is hard to find any quantifications of activated sludge filterability changes due to different operating conditions in literature. Comparisons between different membrane configurations are rarely investigated [12, 13] and hydrodynamic environments are till now never linked with the activated sludge filterability.

During this combined pilot-scale study performed by Cranfield University and Delft University of Technology, operating conditions were fixed for three different membrane configurations a multi tube, a hollow fibre and a flat sheet membrane type. Changes in activated sludge filterability were observed depending on the membrane chambers, the permeate flux of extraction and the membrane chamber aeration. The aim of this study is to provide a better understanding of the interactions between MBR activated sludges and hydraulic circumstances depending on membrane configurations. A set of analyses (composed of SMP, particle size distribution, conductivity, pH, TSS, COD) was performed for each set up of experiments.

2. Materials and methods

2.1. The DFCm

Delft University of Technology has developed a small scale Filtration Characterisation Installation combined with a measuring protocol to investigate activated sludge filterability or sludge reversible fouling potential. The DFCm (see Fig. 1) is a short- term experiment described in detail by Evenblij *et al.*, [12]. A single side stream ultrafiltration membrane tube (X-flow, diameter = 8mm, nominal pore size = 0.03μ m) forms the basic filtration system of the unit. Peristaltic pumps are used for the sludge recirculation with a cross-flow velocity of 1 m/s. The permeate is extracted at a constant flux.



Activated sludge samples collected from different MBRs can then be filtrated under identical operational circumstances. In this way differences in filterability can be related exclusively to MBR sludge properties of the sample [14].

The main step of the measuring protocol is the activated sludge filtration step. During the filtration step several parameters (TMP, resistance, flux, temperature, pH) are monitored and stored in a computer file using the software application Testpoint. The main output of an experiment is the course of the resistance during filtration. An example of the output of an experiment is represented in Fig. 2. In this graph the filtration resistance is plotted against the permeate production per membrane surface. The starting or membrane resistance is similar for all experiments and is left out of consideration when analyzing the results. As a result of fouling of the membrane during filtration, the filtration resistance will increase. The slope of the curve gives an indication of the sludge filterability; a steep curve corresponds with a poor filterability. To make a comparison between different curves each curve can be represented as the value ΔR_{20} : This value represents the increase of the resistance after a permeate production of 20 L/m^2 .

2.2. Cranfield pilot-scale plant

The pilot plant research focuses on a direct comparison of three different full-scale sized membrane configurations operated in parallel as air-lift side stream mode from a 2.2 m³ aeration tank. Those three air-lift side stream operated modules are a multi-tubular membrane (MT), a single flat sheet module (FS) and a hollow fibre (HF) module. Two special vessels were designed to facilitate the operation of FS and HF membrane in airlift side-stream mode where simulative separation distances prevalent in submerged modus were chosen for their design. For hydrodynamic comparison the filtration path length of each membrane module was fixed







to 1.45 m. The aeration tank itself consists of an internal submerged HF module ($A_m = 17.5 \text{ m}^2$) which is functioning as a HRT control module and enables the operation of the side-stream modules decoupled from the hydraulic overall performance of the pilot plant. The scheme of the pilot plant is presented in Fig. 3 and its membrane modules characteristics in Table 1.

2.3. Soluble microbial product analysis

Soluble microbial products (SMPs) were extracted by centrifugation with subsequent filtration. 200 ml of biomass of thoroughly mixed activated sludge sample were centrifuged at 10,000 g for 10 min at room temperature and the supernatant was filtrated through a 70 mm Schleicher & Schuell Grade GF 52 glass fibre filter paper (Patterson Scientific, Bedfordshire, UK). SMP were quantified as proteins and carbohydrates. Determination of proteins was done according a modified method based on (Lowry *et al.*, 1951 [15]; Frølund *et al.*, 1995 [16]). Carbohydrates were analysed according (Dubois *et al.*, 1956 [17]).

2.4. Particle size distribution analysis

Activated sludge particle sizes were measured using the Malvern Mastersizer 2000 particle analyser



Fig. 3. (a) Scheme of the pilot plant (b) Picture.

Table 1 Overview of membrane modules operated at pilot plant.

(Malvern Instruments Ltd, Worcestershire, UK). Particle size was expressed as mass median diameter (d 0.5) in μ m. The Mastersizer measurement is volumebased and according to Mie theory, assumes that the particles causing light absorption and scatter are perfect spheres. Consequently, the results are both volume based and expressed in terms of equivalent spheres. The percentage volume of particles is plotted against particle size (μ m).

The following parameter is reported:

• Mass Median Diameter (D [v, 0.5]): the particle size (μm) at which, 50% of the sample is smaller and 50% is larger.

3. Results and discussion

3.1. Preliminary experiments: daily flow variations

Figure 4 is presenting the variations in activated sludge filterability along the day. The fluctuations seems to follow the sewage loading flow pattern, namely a high loading in the early morning corresponding with a peak in term of filterability (poor activated sludge filterability quality with a ΔR_{20} equal to 4.05 10¹² m⁻¹), then a decrease during the day (improvement of the activated sludge filterability along the day till a ΔR_{20} of 3.2 10¹² m⁻¹) and finally a second peak in the evening (peak in term of filterability again ΔR_{20} equal to 3.6 10¹² m⁻¹). Due to these variations it is difficult to compare changes in activated sludge samples which are not taken at the same time. Therefore, between each experiment a measure of the activated sludge filterability of the aerobic tank was performed. Results for each membrane tank will be presented as a dynamic changes (ratio) comparing the filterability in the aerobic tank with the current ΔR_{20} in the membrane chamber ($\Delta R_{20tank}/\Delta R_{20aerobic}$). Thus, a value of 1 means that the experiment does not have any impact on the activated sludge in term of fouling potential propensity. A value of 0.5 means that the activated sludge filterability improves during the experiment and one above 1 means that the sludge quality gets worse.

ID	Туре	Operation Mode	Surface Area (m ²)	Material	Pore Size (µm)
MT	Multi tube	Air-lift side stream	3.10	PVDF	0.03
FS	Flat sheet	Air-lift side stream	1.40	PVDF	0.08
HF	Hollow fibre	Air-lift side stream	2.75	PES	0.04
AK	Hollow fibre	Submerged HRT control	17.5	PVDF	0.1



Fig. 4. Example of daily sludge quality variations.

3.2. Flux variation impacts

During this experimental period, the three air-lifted membrane chambers were operated with a fixed air lift of 35 L/min of air. The permeate flux of extraction in the pilot plant was fixed at 9 $Lm^{-2}h^{-1}$ the first and the last days of this trial. The permeate flux was fixed at 18 $Lm^{-2}h^{-1}$ on the second day. The permeate flux of extraction of the DFCm was fixed at 30 $Lm^{-2}h^{-1}$.

Results in term of activated sludge filterability are presented in Fig. 5. Different behavior depending on the membrane chamber can be observed:

- *Multi tube configuration*: There were almost no impacts on the activated sludge filterability due to the passage of the activated sludge in the membrane channel or due to the flux increase. The ration $(\Delta R_{20MT} / \Delta R_{20aerobic})$ stayed close to 1.
- *Flat sheet configuration*: A slight decrease in activated sludge filterability was noticeable at low flux (around 15% with a ration $\Delta R_{20FS} / \Delta R_{20aerobic}$ equal to 1.15). However at a high flux the activated sludge filterability between the aeration tank and the membrane chamber did not change (ratio equal to 1).



Fig. 5. activated sludge filterability evolution depending of the flux of extraction in the membrane tank of the pilot plant.

• *Hollow fibre configuration*: An activated sludge filterability decrease of 25% was observed during the low flux trial (ratio $(\Delta R_{20HF}/\Delta R_{20aerobic})$ equal to 1.25) whereas a improvement of the filterability was observed during the high flux trial (33% of improvement with a ratio of 0.67). From the three membrane configurations, the hollow fibre one seems to induce the more significant changes in activated sludge filterability under relatively high-flux conditions.

The deterioration of the filterability at low flux seems to accent the results of Park (2005) [11]. In this study high-shear circumstances (coarse bubble and high air-lift velocity) were responsible for microbial floc breakage (observed figure 6(c)) resulting in an activated sludge filterability deterioration. However a different behaviour was noticed at high flux. The improvement in filterability observed in the flat sheet and in the hollow fibre configuration under relatively high-flux circumstances seems to be linked with an increase of the TSS content, i.e. a concentration effect, compare to the aerobic tank (from 6 to 7.8 g/L). This TSS concentration effect might compensate the activated sludge filterability deterioration due to the floc size decrease (effect on the cake layer porosity). The results are presented in Figs. 6 (a), (b) and (c).

The different behaviour observed in the multi tube configuration might be resulting from a short residential time in the membrane chamber (15s, i.e. no significant activated sludge modifications had time to occur). Even if a reduction of the particle size was measured in each membrane chamber, no clear relation between particle size distribution (or SMP content-Fig. 6 (d)) and filterability was underlined.

3.3. Impact of the air-lift velocity

During this experimental period, the three membrane chambers were operated with a fixed permeate flux of 30 $\text{Lm}^{-2}\text{h}^{-1}$ in order to accent the observed phenomenon during the flux experiments. Several air-lift velocities were tested for each membrane module, namely from 11 to 25 L/min for the multi tube configuration, from 1 to 8 L/min for the flat sheet configuration and from 0.5 to 8 for the hollow fibre one. Due to a significant improvement of the activated sludge filterability in the aerobic tank the permeate flux of extraction of the DFCm was fixed at 60 Lm⁻²h⁻¹.

Results are presented in Fig. 7(a).

Multi tube configuration: The filterability did not vary in the range of tested air-lift velocities for this configuration. From 25 to 11 L/min, the ratio $\Delta R_{20MT} / \Delta R_{20aerobic}$ remained stable with a value around 1.



Fig. 6. (a) TSS depending on the day, (b) Filterability variation versus TSS, (c) Particle size distribution depending on the day, (d) Filterability variation versus protein content.

Flat sheet configuration: Activated sludge filterability seems to slightly improve when the air-lift velocity decreased from 8 to 1 L/min. the ratio $\Delta R_{20FS} / \Delta R_{20aerobic}$ during theses two states varied from 1.25 to 0.8. It seems that operating a flat sheet configuration with a relatively high air-lift velocity deteriorate the activated sludge filterability (ratio equal to 1.25) whereas an activated sludge filterability improvement (ratio equal to 0.8) is noticeable in this configuration for a low air-lift velocity (namely 1L/min).

Hollow fibre configuration: A strong correlation was observed between the activated sludge filterability and the air-lift velocity in the hollow fibre chamber (R²=0.93). The ratio $\Delta R_{20HF}/\Delta R_{20aerobic}$ dropped from 1.25 to 0.07 when the air-lift velocity decreased from 8 to 0.5 L/min. It seems that as for the flat sheet configuration, operating a hollow fibre configuration with a relatively high air-lift velocity deteriorates the activated sludge filterability (ratio equal to 1.25) whereas a strong activated sludge filterability improvement (ratio equal to 0.07, 90% of sludge filterability improvement) is noticeable in this configuration for a low air-lift velocity (namely 0.5 L/min). From the three membrane configurations, the hollow fibre one is inducing the most significant changes in activated sludge filterability under low air-lift velocity conditions.

Like during the flux experiments, filterability improvements in the flat sheet and in the hollow fibre configurations are linked with TSS content concentration (Figs. 7 (b) and (c)). The increase in TSS (concentration effect) can be correlated with the decrease of the air-lift velocity ($R^2 = 0.96$ in the hollow fibre configuration) and thus the improvement in filterability (Fig. 7(c)). Even if most of the study in the literature linked increases of TSS with membrane fouling potential increase, this kind of behavior was already observed by different research group (Le-Clech et al., 2003 [8], Defrance et al., 1999 [18]). As shown by Bae and Tak (2005) [19], the main mechanism involved in reversible fouling is the cake layer formation. They also found that the total suspended solids were the major contributor to this fouling process. As already formulated by Le-Clech et al., (2003) [8], the activated sludge filterability might improve at high TSS content (close to 12 g/L) due to the formation of a protective gel layer. This TSS concentration effect might favor the formation of a high porous media or a loosely bound cake layer. However, these hypotheses were not substantiated by the particle size distribution analyses. No trend between floc size and filterability were underlined (Fig. 7(d)). More than the TSS content itself, the TSS concentration phenomenon occurring under specific conditions is likely responsible for the activated sludge filterability improvement.

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Fig. 7. (a) Filterability variation versus air-lift velocity, (b) TSS versus air-lift velocity, (c) Filterability versus TSS, (d) Filterability versus particle size distribution.

During all these experiment periods, no correlation between SMP and filterability were emphasized. It is in accordance with the study of Yamato *et al.*, (2006) [20] and Wu *et al.*, (2008) [21] which tend to link SMP and irreversible fouling mechanisms. TSS concentration seems to be the predominant factor explaining activated sludge filterability improvement.

4. Conclusion

A combined pilot-scale study was performed by Cranfield University and Delft University of Technology. Operating conditions (namely flux and cross-flow velocity) were fixed for three different membrane configurations a multi tube, a hollow fibre and a flat sheet membrane type. Changes in activated sludge filterability were quantified depending on the membrane chambers, the permeate flux of extraction and the membrane chamber aeration. Two mechanisms can be distinguished from this study:

- 1. Due to practical arrangements (volume of the tank, sludge residential time), hydraulic conditions in each membrane unit are different, leading to different total suspended solids concentrations in each tank depending on operating conditions.
- 2. These margins in TSS contents cause differences in filtration behaviour.

These mechanisms can be explained by the theory that the MBR activated sludge is in a continuous flocculation and deflocculation status; this leads to changing levels of submicron particles causing differences in filtration characteristics.

The air-lift velocity decrease has for direct consequence an increase of the activated sludge retention time in the membrane chambers. The retention time increase leads to a total suspended solids concentration increase under constant flux operations (concentration phenomenon). Activated sludge is subjected to shear and stress conditions during its transport from the aerobic tank to the membrane chambers. If the activated sludge floc structure is disrupted during transport or pumping, a readaption time allowing a proper (re-)flocculation might be necessary to obtain a well-filterable activated sludge.

Therefore, a sufficient retention time in the membrane chambers in combination with a sufficient high suspended solids concentration seem to be essential to provide a well-filterable activated sludge.

Following conclusions can be drawn from these points:

• The multi tube configuration did not affect the sludge quality. The sludge filterability remained the same in spite of flux and air-lift velocity changes. It might be brane chamber.

- The flat sheet and the hollow fibre configuration did affect the activated sludge filterability. The activated sludge filterability improved significantly at high flux and at low air-lift velocity (from 20% to 90% of improvement).
- From the three membrane configurations, the hollow fibre one seems to induce the more significant changes in activated sludge filterability at high flux and at low air-lift velocity.
- TSS concentration increase is the major factor explaining activated sludge filterability improvement. It might be due to a floc structure modification. Foulants could have been trapped in the floc network due to the concentration phenomenon.
- No correlations were found between SMP and filterability.

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References

- Judd, S.J., (2006). The MBR book: Principles and Applications of Membrane Bioreactors in Water and Wastewater Treatment. Elsevier, Oxford.
- [2] Le-Clech, P., Chen, V., Fane, T.A.G., (2006), Journal of Membrane Science 284, 17–54.

- [3] Rosenberger, S., Kruger, U., Witzig, R., Manz, W., Szewzyk, U., and Kraume, M., (2002), Water Research 36, 413–420.
- [4] Itonaga, T., Kimura., K and Watanabe, Y., (2004), Water Science and Technology 50, 301–309.
- [5] Geilvoet, S. Moreau, A., Lousada Ferreira, M., van Nieuwenhuijzen, A., van der Graaf, J., (2007) Particle Separation Conference, IWA, Toulouse, France.
- [6] Drews, A., Vocks, M., Bracklow, U., Iversen, V., Kraume, M., (2007) Membranes for water and Wastewater Treatement Conference. IWA, Harrogate, UK.
- [7] Jae-Hoon Choi, How Yong Ng, (2008), Chemosphere 71, 853-859.
- [8] Le-Clech, P., Jefferson, B., Judd, S.J., (2003) Journal of Membrane Science 218, 117–129.
- [9] Dufresne, R., Lebrun, R.E., Lavallee. H.C., (1997) Can. J. Chem. Engng. 75, 95–103.
- [10] Metzger, U., Le-Clech, P., Stuez, R.M., Frimmel, F.H., Chen, V., (2007) J. Membr. Sci. 301 (1-2) 180–189.
- [11] Park, J.S., Yeon, K.M., Lee, C.H., (2005), Desalination 172, 181-188.
- [12] Evenblij, H., Geilvoet, S., van der Graaf, J., van der Roest, H.F., (2005), desalination 178, 115–124.
- [13] Hai, F.I., Yamamoto, K. and Fukushi, K., (2005), Desalination 180, 89–97.
- [14] Geilvoet, S.P., Moreau A.A., Lousada Ferreira, Md. C., Van Nieuwenhuijzen A.F., Van der Graaf J.H.J.M., (2007), 4th IWA International Membrane Conference, Harrogate, UK.
- [15] Lowry O, Rosebrough N, Farr A and Randall R (1951), *Journal of Biological Chemistry* 193, 365–17 [16] Frølund B, Griebe T and Nielsen P (1995)., *Applied Microbiology and Biotechnology* 43, 755–761.
- [17] Dubois M, Gilles K, Hamilton J, Rebers P and Smith F (1956)., Analytical Chemistry 28(3), 350–356.
- [18] Defrance, L., Jaffrin, M.Y., (1999), J. Membr. Sci. 157, 73-84.
- [19] Bae, T.H., Tak, T.M., (2005) Journal of Membrane Science 264, 151–160.
- [20] N. Yamato, N., Kimura, K., Miyoshi, T., Watanabe, Y., (2006) Journal of Membrane Science 280, 911–919.
- [21] Wu, J., Le-Clech, P., Stuetz, R.M., Fane, A.G., Chen, V., (2008) Water Research 2008.06.004.

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