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# IMPACTS OF THE SPECIFIC CAKE RESISTANCE ON MBR FOULING FOR WASTEWATER TREATMENT

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Abstract. Membrane bioreactor (MBR) has been increasingly used for municipal wastewater treatment and reuse due to its good effluent quality. However, membrane fouling remains the major limitation of MBR. Understanding fouling is still a key issue for a more sustainable operation of MBRs. Thus, this research presents the influence of specific cake resistance ( $\alpha$ ) on the fouling propensity in the MBR. Correlation between  $\alpha$  value with fouling resistance ( $R_f$ ), fouling rate (dTMP/dt), especially of peak height 100-1000 kDa protein-like SMPs was investigated. The result reported that the  $\alpha$  value was strongly correlated with the dTMP/dt in the MBR ( $R^2$  value of close to 1). In this study, however, there is an obvious discrepancy between the fouling resistance calculated from the resistance in the series model and the  $\alpha$  value in the supernatant filtration. These observations demonstrated that the fouling propensities of the membrane could be monitored by the transmembrane pressure and the fouling characteristics, include fouling resistance and specific cake resistance in the filtration cell.

**Keywords:** membrane bioreactor, membrane fouling propensity, fouling resistance, specific cake resistance, transmembrane pressure.

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# **1. INTRODUCTION**

The membrane bioreactor (MBR) technology has been used to treat wastewater that combines a bioreactor and membrane separation. MBR produces very high-quality treated

water containing almost no detectable suspended solids. The treated water quality is equivalent to tertiary wastewater treatment (i.e., the combination of activated sludge and depth-filtration). In addition, membrane filtration in MBR processes obviates gravity sedimentation tanks, which results in a lower bioreactor footprint, reducing waste sludge production and precise control of sludge retention time (SRT) than conventional activated sludge (CAS) processes. For all the advantages, MBR also has disadvantages mainly related to the membranes. The membrane fouling due to: (1) blockage of the smallest pores, (2) coverage of the larger pores' inner surface, (3) superimposition of particles and direct blockage of larger pores, and (4) creation of cake layer is the major problem encountered during the application of the MBR process in wastewater treatment.

Therefore, the success of MBR operation is largely dependent upon how to cope with the membrane fouling, which is affected by many factors such as the influent water quality, membrane characteristics, bioreactor operational conditions, and the membrane cleaning method. Individual fouling factors affect membrane fouling separately and/or mutually. For example, important operating conditions such as hydraulic retention time (HRT) and SRT influence membrane fouling directly. They affect the microbial characteristics simultaneously, such as extracellular polymeric substances (EPS) production or mixed liquor suspended solids (MLSS) concentration, which are important factors controlling membrane fouling. A previous study by Fu et al [1] highlighted that an increase in the proteins and carbohydrates concentrations was observed in the MBR, from  $15.00\pm3.95$  mg BSA.L<sup>-1</sup> to  $33.49\pm7.83$ mg BSA.L<sup>-1</sup> and from  $10.39 \pm 3.42$  mg glucose. L<sup>-1</sup> to  $13.61 \pm 2.72$  mg glucose. L<sup>-1</sup>, respectively when SRT decreased from 20 days to 5 days. Aida Isma et al [2] reported the biggest cake layer thickness was observed at the shortest SRT of 4 days and the longest HRT of 12h in the hollow fiber membrane bioreactor. Additionally, the effect of temperature on the total membrane resistance (Rt) was studied by Arévalo et al [3]. Their result noticed that an increase of the Rt value could relate to lower temperatures (<15°C). However, the nitrification and denitrification were not affected by various temperatures in the MBR. Besides, Berkessa et al [4] demonstrated the coupling of high mixed liquor suspended solid concentration (MLSS $\sim 22$  g.L<sup>-1</sup>) with long hydraulic retention time of 47 days influenced membrane fouling in the AnMBR. However, high performance of COD removal (>98%) was found in this study. A few previous studies by Kornboonraksa and Lee [5]; Lee and Kim [6] revealed that the membrane fouling increased with the increase of MLSS concentration. One of the key operating parameters affected the membrane fouling, especially biofouling is hydraulic retention time during MBR process. An increased HRT (from 4h to 6.67h) could decrease the total fouling resistance (from  $4.5 \times 10^{12}$  to  $2.5 \times 10^{12} \text{ m}^{-1}$ ), thus mitigating membrane fouling in a sponge-submerged MBR [7].

Fouling of membrane is generally characterized as a decrease in permeation flux or an increase in transmembrane pressure according to the operation mode, which deteriorates the MBR performance [8, 9]. Different mechanisms of membrane fouling, such as the formation of gel or cake layer, pore blocking and adsorption can appear during the MBR filtration. According to Chen et al [10], the fouling mechanism could be caused by the gel layer in the MBR operated continuously. The results reported the filtration resistance are seen to increase linearly with gel thickness, but it was independent of ionic strength and pH. A previous study, Akhondi et al [11] investigated the membrane fouling was affected by the concentration of the wastewater and the filtration flux in the submerged hollow fiber membrane system. Their results showed that a higher fouling rate was caused by an increase in feed concentration. The faster deposition rate of the particles onto the membrane surface could be due to the higher filtration flux. In addition, a strong linear correlation between feed water turbidity and specific cake resistance in the

ultrafiltration membrane process was observed by Chew et al [12]. Furthermore, the filtration performance is related to the structure or formation of the cake layer. Characteristics of cake layer by choosing appropriate coagulant and coagulation conditions are key determinants of membrane performance [13]. In addition, the reduced cake layer resistance in the MBR-G could be also ascribed to less growth of suspended biomass, lower sludge viscosity, as well as less EPS, SMP and biopolymer clusters in the cake layer [14].

During the MBR filtration, the increase continuously in filtration resistance could relate to the accumulation and compression of the cake layer. Thus, the main objective of this work is to examine the effects of the specific cake resistance in terms of fouling behavior in a lab-scale MBR for domestic wastewater treatment. The results found in this study can provide highlights for membrane fouling control and guidance for optimization in MBR applications.

# 2. MATERIAL AND METHODS

#### 2.1. MBR set-up and operation

Figure 1 presents the pilot-scale MBR. The MBR process consists of an anoxic reactor of 5.4 L volume and an aerobic reactor with a working volume of 12.6 L. One flat sheet microfiltration (MF) membrane, made of poly-sulfone (PS), with a filtration area of 0.1 m<sup>2</sup> and a pore size of 0.2  $\mu$ m, was submerged into the aerobic tank, as described in previous articles [8, 9]. The operating parameters of the MBR pilot are summarized in Table 1.

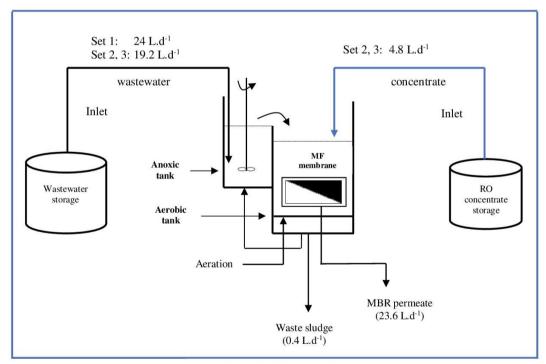


Figure 1. Schematic diagram of the MBR.

The MBR was run over 55 days, including three sets of experiment. At the end of the first set (the 40<sup>th</sup> day), the concentrate produced from reverse osmosis pilot was added directly into the aerobic tank at a flow rate of 4.8 L.d<sup>-1</sup>. The injection of RO concentrate to the MBR can be the feasibility option to reduce the serious environmental impacts occurring due to the toxic component contained in the concentrate. Additionally, no significant impacts of RO concentrate

on the MBR performances were observed in our previous study [9]. The Set 2 and Set 3 were operated during 8 days and 7 days, respectively.

1 01	5		
Operating conditions	Range		
pH	7-8.5		
Dissolved oxygen (DO)	$2 - 3 \text{ mg.L}^{-1}$		
Aeration	With big air bubbles at a flow rate of 1.5 L.min <sup>-1</sup>		
Filtration/ relaxation cycle	8 mins/ 4 mins		
MLSS	6 - 8 g.L <sup>-1</sup>		
SRT	45 days		
Net flux of MBR permeate	10 L.h <sup>-1</sup> .m <sup>-2</sup>		

Table 1. Operating parameters of the MBR system

#### **2.2. Specific cake resistance** $(\alpha)$

The stirred dead-end filtration cell (Amicon 8050, Millipore) was used to determine the  $\alpha$  value. For continuous weighting of filtrate, weight balance was connected with computer. Polysulfone (PS) membrane (Laval, France) with the same pore size of MBR membrane was used for the sludge filtration. Polyether-sulfone (PES) membrane (Orelis, France), with a pore size of 0.01  $\mu$ m was used for the filterability tests of supernatants. Constant pressure of 1 bar was applied during filtration tests.

The permeate flux, J, is proportional to the driving force for membrane filtration and inversely proportional to the sum of all the resistance:

$$J = \frac{\text{driving force}}{\Sigma \text{ resistances}}$$
(1)

The driving force for the membrane filtration is the TMP, and the resistance of permeation is the sum of resistances of the permeate viscosity:

$$J = \frac{\Delta P}{\eta \times R_t}$$
(2)

where:

J is the permeation flux (L.h<sup>-1</sup>.m<sup>-2</sup>),

 $R_t$  is the total resistance (m<sup>-1</sup>)

 $\eta$  is the viscosity of the permeate (Pa.s)

 $\Delta P$  is the transmembrane pressure (bar or Pa)

The transmembrane pressure (TMP) is calculated from pressure during membrane operation, and there is no concentrate water flow, TMP is the differences between the pressures of the outflow (permeate) and the inflow.

Total resistance  $(R_t)$  consists of intrinsic membrane resistance  $(R_m)$  and resistance arising from all kinds of fouling  $(R_f)$  as in Equation (3).

$$J = \frac{TMP}{\eta \times (R_m + R_f)}$$
(3)

where:

 $R_m$  is the membrane resistance (m<sup>-1</sup>)

R<sub>f</sub> is the fouling resistance (m<sup>-1</sup>)

The fouling resistance  $(R_f)$  is determined according to Equation (4):

$$R_{f} = \frac{\alpha \times V \times m}{A} \tag{4}$$

The specific cake resistance was calculated by Maqbool et al [15].

$$\alpha = \frac{2000 \times A^2 \times \Delta P}{\eta \times m} \times \frac{t/V}{V}$$
(5)

Where  $\alpha$  is the specific cake resistance (m.kg<sup>-1</sup>), A is the filtration membrane area (0.00134 m<sup>2</sup>),  $\Delta P$  is the applied pressure (bar), m is the mass of the biofilm (kg.m<sup>-3</sup>) and  $\frac{t/V}{v}$  (s.m<sup>-6</sup>) is the slope of the straight portion of the curve that is obtained by plotting the time of filtration to volume of filtrate (t/V) versus the filtrate volume (V). For sludge filtration, m was calculated from the MLSS value of sludge; for supernatant filtration, m was calculated by retained dissolved organic carbon (DOC)

#### 2.3. Analytical methods

The MLSS concentration was quantified by the standard method AFNOR NFT 90-105. COD was detected by a digestion reactor (HACH Co., USA) and direct reading spectrometer (DR/2000, HACH Co., USA). The determination of DOC was performed on a TOC analysis (TOC-V Series, Shimadzu, France) after samples passed through a 0.45  $\mu$ m membrane to remove bigger particles. The supernatants were prepared by centrifugation of the MBR sludge samples at 4000rpm during 10 minutes at room temperature.

HPLC-SEC-Fluorescence analysis was performed to detect the molecular weight distribution of protein-like substances. [8].

# **3. RESULTS AND DISCUSSION**

The MBR was run in a steady-state condition. The chemical oxygen demand (COD) and dissolved organic carbon (DOC) removal efficiencies were above 94% and 93%, respectively.

# **3.1.** Correlations between fouling resistance and specific cake resistance in batch filtration cell

The filtration characteristics were evaluated by the resistance. The averaged resistance values after three experimental sets are summarized in Table 2. The increase in  $R_f$  was reported during sludge and supernatant filtrations. For example, in the second set of experiments,  $R_f$  values were  $11.40 \times 10^{12}$  (m<sup>-1</sup>) and  $7.10 \times 10^{12}$  (m<sup>-1</sup>), 3.8 and 3.3 times higher than those in the Set 1, of both sludge and supernatant filtration, respectively. These values continuous increased in Set 3, resulting in overall increase in the  $R_t$ . This could cause a modification of the fouling layer structure formed during the entire experimental period.

It is well known that the specific cake resistance obtained from the membrane filtration is a quantitative measure of the fouling potential or filterability of sludge cake. As seen in Fig. 2, the R<sup>2</sup> value is closer to 1, it indicates that there is a strong linear relationship between the fouling resistance (R<sub>f</sub>) and the  $\alpha$  value in the sludge filtration. In contrast, a weak correlation was found between the specific cake resistance and the R<sub>f</sub> in the supernatant filterability test (R<sup>2</sup>=0.35). The strong correlation observed between the  $\alpha$  value and R<sub>f</sub> in the sludge filtration was due to the MLSS value, that was deposited on the membrane surface, quite stable during the experiment time. A possible explanation could be that an increase in mass deposited on the membrane surface, leading to the weak relationship between the  $\alpha$  value and the fouling resistance in the supernatant filtration. For instance, DOC mass retained by the membrane were 0.0045 kg.m<sup>-3</sup> in Set 2 and 0.011 kg.m<sup>-3</sup> in Set 3. This result indicated the constants specific cake resistance ( $\alpha$ ) and the mass of biofilm (m) are exactly corresponding to the membrane fouling rate. Consequently,  $\alpha$  value could not be a proper criterion for the estimation of membrane fouling in the supernatant filtration.

Table 2. Membrane fouling resistance in sludge and supernatant filtration.

Resistances	Sludge filterability test			Supernatant filterability test		
	SET 1 <sup>(a)</sup>	SET 2 <sup>(b)</sup>	SET 3 <sup>(c)</sup>	SET 1 <sup>(a)</sup>	SET2 <sup>(b)</sup>	SET 3 <sup>(c)</sup>
$R_m (\times 10^{12}  m^{-1})$	1.27	1.31	1.39	7.58	7.10	6.93
$R_{f}$ (× 10 <sup>12</sup> m <sup>-1</sup> )	3.01	11.40	17.50	2.14	7.10	15.50
$R_t (\times 10^{12}  m^{-1})$	4.28	12.70	18.90	9.72	14.20	22.40
$R_{f}/R_{t}(\%)$	70	90	93	22	50	69

Samples were taken from MBR on the: <sup>(a)</sup> 40<sup>th</sup> day; <sup>(b)</sup> 48<sup>th</sup> day; <sup>(c)</sup> 55<sup>th</sup> day of the filtration period.

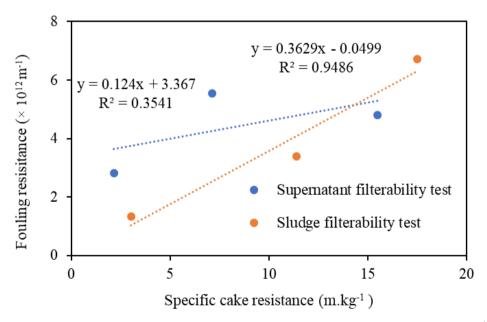


Figure 2. Correlations between fouling resistance (Rf) and specific cake resistance:  $\alpha \times 10^{-13} \text{ m.kg}^{-1}$  in sludge filtration,  $\alpha \times 10^{-16} \text{ m.kg}^{-1}$  in supernatant filtration.

# 3.2. Influence of specific cake resistance on MBR fouling

The transmembrane pressure is a key parameter to evaluate membrane performance in the MBR since it was affected directly by the membrane fouling rate. Figure 3 plotted a relationship between the fouling rate of the MBR and the  $\alpha$  value in the sludge filtration. The linear curve shows a strong relationship between the specific cake resistance and the fouling rate with an R<sup>2</sup> value of close to 1. As observed in Fig. 3, the lowest  $\alpha$  value of sludge filtration had a close similarity with the minimum fouling rate, thus, the specific cake resistance could be seen as a positive operational parameter to evaluate the membrane fouling rate (dTMP/dt) in MBR filtration process.

According to Darcy law at constant flux, Eq. (4) becomes:

$$\frac{\mathrm{dR}_{\mathrm{f}}}{\mathrm{dt}} = \frac{1}{\eta \times \mathrm{J}} \times \frac{\mathrm{d}\Delta \mathrm{P}_{\mathrm{t}}}{\mathrm{dt}} = \frac{\alpha \times \mathrm{m}}{\mathrm{A}} \times \frac{\mathrm{d}V}{\mathrm{dt}} \tag{6}$$

Eq. (6) displays the constant specific cake resistance ( $\alpha$ ) and mass of the biofilm (m) are directly corresponding to the fouling rate of MBR membrane (dR<sub>f</sub>/dt). Since the mass of the biofilm was calculated from the MLSS concentration, and it was maintained around 7.3 g.L<sup>-1</sup> in the MBR, so, the only parameter that can be considered as crucial in influencing the observed fouling rate was specific cake resistance ( $\alpha$ ).

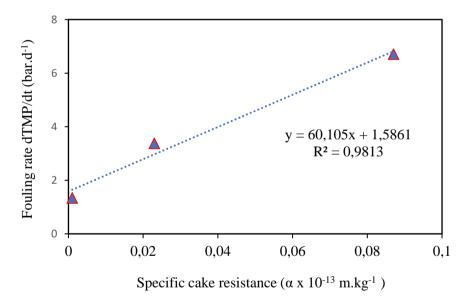


Figure 3. Correlation between fouling rate (dTMP/dt) of the MBR and  $\alpha$  value in the sludge filterability test.

Furthermore, to concern the peak height of large protein-like substances with fouling propensity of supernatant samples (set 1, set 2, set 3), the peak height of these macromolecules in MBR supernatants and  $\alpha$  values obtained from sludge filtration using 0.2 PS membranes at TMP of 1 bar are presented in Figure 4. As seen in Fig. 4, a good linear correlation between the peak height in 100-1000 kDa and  $\alpha$  values was investigated, which further demonstrated the important role of the  $\alpha$  value in the fouling propensity of the MBR.

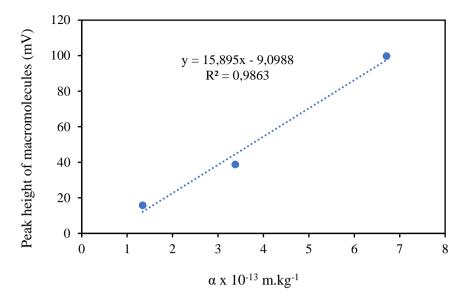


Figure 4. Peak height for MBR supernatants versus a values calculated from MBR sludge filtration.

# 4. CONCLUSION

This study conducted a sustainability evaluation of the  $\alpha$  value on membrane fouling in the MBR. For the dead-end type membrane filtration of MBR supernatant, a weak relationship between the  $\alpha$  value and the fouling resistance was observed. This suggests that  $\alpha$  cannot be used as a criterion for the estimation of membrane fouling in the MBR supernatant filterability test. However, a significant influence of specific cake resistance on fouling rate (dTMP/dt) in the MBR was found. These observations reveal the important role of both the membrane fouling resistance and specific cake resistance in the fouling propensity of the MBR.

To achieve the optimization in full- scale MBR applications, further experiments and simulations to evaluate range of acceptable operating parameters, need to be extended.

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