

Characterization of fouling-related organics in MBRs

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Abstract

Membrane bioreactors are a promising technology used for wastewater treatments. However, the presence of soluble microbial products (SMP) in activated sludge suspension and their subsequent deposition on membranes can cause a rapid decline of membrane flux, this behavior also called membrane fouling. Due to the crucial role of SMP in fouling development, SMP have attracted much attention in MBR fouling study. Despite a number of studies have been performed to study the occurrence and fouling propensity of SMP in MBRs, the formation mechanisms in particular for the MBR process, characteristics, and control strategies of SMP remain unclear. In this study, we found that the fouling-related organics in different scale MBRs differed significantly. The feedwater loading rates and MLSS concentrations in the MBRs would affect the formation of EPS and SMP substantially. The discharge of SMP-PS and SMP-PN likely depended on their levels and the MBR scale. The discharge of SMP would help to mitigate the accumulation of fouling-related organics in the bioreactor.

1 Introduction

Over last two decades, membrane bioreactors (MBRs) have been actively used for biological wastewater treatment due to the increasingly stringent effluent quality requirements¹⁻³. But, a major obstacle to the wider application of MBRs is membrane fouling⁴, which results in flux loss and requirement of chemical cleaning. Of particular importance is that MBR fouling is still unknown due to the complex nature of activated sludge to be filtered. The MBR fouling primarily results from the deposition of fouling-related organics either in bound form (bound extracellular polymeric substances, bound EPS) or in soluble form (soluble microbial products, SMP) onto membranes.

Bound EPS is mainly composed of proteins, polysaccharides, and humic acid, which are located at or outside the cell surface. SMP can be defined as the pool of organic compounds that are released into solution from substrate metabolism (usually with biomass growth) and biomass decay⁵. During the biological process, part of bound EPS can be hydrolyzed to SMP. Some SMP can be consumed by activated sludge; and some can be adsorbed by the sludge flocs and then, become bound EPS⁶. To understand on the influence of fouling-related organics on MBR fouling, considerable attempts have been made. But, these investigations were of different focuses. For example, some investigations found that bound EPS was influential to MBR fouling^{7,8}, and SMP was also paid much attention by some researchers⁹⁻¹¹.

On the other hand, the formation of fouling-related organics in MBRs highly depends

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on the operating conditions and feedwater composition. Massé et al. ¹² observed that both bound EPS and SMP decreased as the solid retention time (SRT) increased from 10 to 53 days. More recently, Liang et al. ¹³ also observed that accumulation of SMP in the MBR became more pronounced at short SRTs. In contrast, Ng et al. ¹⁴ found an increase in bound EPS when SRT increased from 0.25 to 5 days. Li et al. ¹⁵ used six lab-scale bioreactors to grow activated sludge with different carbon sources of glucose and sodium acetate, and reported that the sludge that was fed on glucose had more EPS than the sludge that was fed on acetate. Moreover, ambient conditions such as temperature, dissolved oxygen (DO) concentration and nitrate concentration were found to have influence on the formation of SMP ¹⁶. The above-mentioned results suggest that: (1) fouling-related organics are of high significance to the occurrence of MBR fouling, (2) the interrelations between bound EPS and SMP are very complex and, (3) the formation of fouling-related organics strongly relies on operating conditions and feedwater.

To date, most of the related studies were focused on the influence of operating conditions on the formation of fouling-related organics and the succedent MBR fouling. Limited information is available regarding the behavior and characteristics of fouling-related organics including the decisive factors affecting formation of fouling-related organics, the interrelation between bound EPS and SMP, the discharge or rejection of SMP, and the fouling propensity of fouling-related organics. A detailed study on the fouling-related organics will help to understand and control of MBR fouling. Therefore, the purpose of this study is to analyze the formation mechanism of fouling-related organics and to understand the fouling mechanism of fouling-related organics.

2 Materials and methods

2.1 MBR plants

The study is based on a long-term monitoring of a pilot-scale MBR plant and a full-scale MBR plant, which was performed from October 2008 to August 2009. The main operating conditions for each MBR are summarized in Table 1. The pilot-scale MBR consists of one anoxic tank and one aerobic tank, and each tank has a working volume of 0.8 m³. The pilot-scale MBR is located in a pumping station of the Berliner Wasserbetriebe, which is used to collect the wastewater and rainwater in Berlin City center. The feedwater of the pilot-scale MBR is a combined municipal wastewater, which is a mixture of domestic wastewater, industrial wastewater and rainwater. As shown in Figure 1 the full-scale MBR is composed of a series of anaerobic tanks, aerobic tanks and anoxic tanks, which is designed for enhanced biological phosphorous removal and post-denitrification and is located in a remote area on the suburb of Berlin. The feedwater consists mainly of domestic wastewater lack of industrial wastewater and rainwater. The full-scale MBR is operated with irregular sludge withdrawal. Thus, the feedwater of the full-scale MBR has high concentration in COD, TN and TP. Table 2 shows the performance of these two MBR plants on nutrient removal. The membrane modules used in both the pilot-scale MBR and the full-scale MBR were purchased from A3 Water Solutions (PVDF, 0.2 μm, Germany). The membrane module is submerged in the aerobic tank, and the coarse bubble provided by the aerator is used to reduce membrane fouling and to provide oxygen for biomass.

Table 1 Operating parameters for the pilot-scale and full-scale MBRs

	VR (m ³)	SRT (d)	MLSS (g/L)	Membrane area (m ²)	Membrane Flux
Pilot-scale MBR	1.6	13	4.6-9.1	22	10
Full-scale MBR	8.9	20-50	13-22	32	16-20

^a average value; PN and PS are protein and polysaccharide, respectively.

Table 2 Performance of the MBRs on nutrient elimination

	COD (mg/L)		TN (mg/L)		TP (mg/L)	
	Feedwater ^a	Permeate	Feedwater ^a	Permeate	Feedwater ^a	Permeate
Pilot-scale MBR						
Full-scale MBR	1206	30-45	150	4-12	21	0.1-1.0

^a average value.

2.2 Analytical methods

The extraction of bound EPS from sludge flocs was performed with a strongly acid cation exchange resin (Na-form, Dowex) according to the method of Frolund et al.¹⁷. The sample of feedwater and sludge supernatant was prepared by filtering the feedwater and activated sludge with filter paper (black ribbon, Schlericher & Schuell GmbH). Polysaccharide concentrations of feedwater, sludge supernatant and membrane permeate were analyzed according to the photometric method proposed by Dubois et al.¹⁸. D-Glucose-Monohydrate was used for calibration. Results of polysaccharide concentration are expressed in glucose equivalents. The influence of nitrite or nitrate on carbohydrate measurement was corrected according to the method proposed by Drews et al.¹⁶. Protein concentrations, expressed in equivalent of bovine serum albumin, were determined according to the method of Lowry et al.¹⁹. All the samples were analyzed as two replicates and the results were given as average value.

Mixed liquid suspended solids (MLSS) concentration was determined by Standard Methods²⁰. Capillary suction time (CST) was determined using a Triton CST apparatus (Model 200, Allied Colloids GmbH, Hamburg, Germany). Time to filter (TTF) was measured as follows: a 90 mm Buchner funnel was used to measure the time required to filter 25 mL of a 250 mL sludge sample through filter paper (black ribbon, Schlericher & Schuell GmbH). In this study, all the sludge samples were collected from the membrane chamber.

3 Results and discussion

3.1 Comparison of fouling-related organics fate in the MBRs

Table 3 Evolution of PN and PS during the MBR process

	PN (mg/L)			PS (mg/L)		
	Feedwater	Sludge	permeate	Feedwater	Sludge	Permeate
Pilot-scale MBR	23-187 (95) ^a	7.9-34 (18) ^a	0-19.2 (9.9) ^a	4.3-33 (16.3) ^a	0-18 (7.6) ^a	0-4.9 (2.2) ^a
Full-scale MBR	110-399 (206) ^a	16-99 (41) ^a	11-33 (21) ^a	11-46 (28) ^a	5.0-55 (26) ^a	0-10 (4.2) ^a

^a average value.

Basically, the behavior and fate of fouling-related organics in MBRs are determined by two processes: biological degradation and membrane rejection. During the whole study, the soluble PN and soluble PS in feedwater, sludge supernatant, and membrane permeate were regularly monitored. The data is summarized in Table 3. It is noticed that PN in both of the two MBRs were of similar fate. For example, 81% and 80% of PN in the feedwater of pilot-scale MBR and full-scale MBR could be eliminated by biological process, and 55% and 51% of PN in the sludge supernatant of these two MBRs was discharged to the permeate. This result indicates that the soluble PN in the two MBRs was of same fate.

In comparison with PN, the PS, however, was observed to have low both bio-elimination rate (e.g., 53% for pilot-scale MBR and 7.1% for full-scale MBR) and discharge rate (e.g., 29% for pilot-scale MBR and 16% for full-scale MBR), suggesting that the PS is difficult to be biodegraded by biomass and discharged by membrane filtration. Of particular notice is the PS fate in the two MBRs: the PS in the full-scale MBR had much lower bio-elimination rate and discharge rate. Actually, the PS derived from domestic wastewater should have acceptable bio-elimination potential. The high PS concentration in the sludge supernatant of the full-scale MBR might be caused by biomass metabolism, the hydrolysis or release of bound EPS, for example. Additionally, the higher SRT (20-50 days) should be another possible reason leading to the high PS concentration in sludge supernatant. In case of the pilot-scale MBR, the frequent sludge discharge would help to remove part of the fouling-related organics accumulated in the bioreactor. However, some previous attempts found that a higher SRT would be better for the control of soluble fouling-related organics or SMP^{12, 13}. Here, we must address that the two MBR plants are subject to different configuration and feedwater. Especially, the feedwater composition and loading rate are of high significance to the accumulation of fouling-related organics and the occurrence of MBR fouling²¹. Moreover, we also notice that the full-scale MBR had a lower discharge rate or higher rejection rate of PS in sludge supernatant than the pilot-scale MBR did. The membrane module itself should have no influence on the discharge of soluble PS because of the same membrane used in the two MBRs. It is assumed that the discharge of PS strongly depends on the size and chemical nature of PS in each bioreactor and the fouling layer formed on the membranes.

Figure 2 illustrates the PN/PS ratios of bound EPS and soluble fouling-related organics in feedwater, sludge supernatant and permeate. It can be seen that there was a low PN/PS in sludge supernatant, suggesting that the PS was readily accumulated in the bioreactor and PS concentration was comparable with PN concentration. However, with respect to feedwater, permeate and bound EPS, the PN was the major contributor to the total fouling-related organics. Similarly, the data in Figure 2 also reveals that the full-scale MBR and pilot-scale MBR were of different PS/PN ratios of the fouling-related organics.

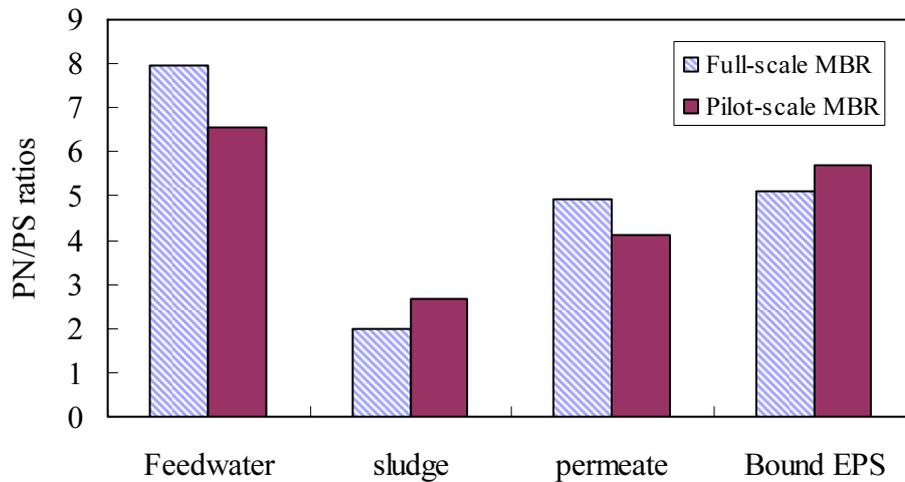


Figure 2 PN/PS ratios of soluble fouling-related organics and bound EPS

To get down to the nitty-gritty of the formation of fouling-related organics, the key factor affecting the formation and accumulation of fouling-related organics in MBRs is attempted to be analyzed on the basis of our experimental data and the unified theory proposed by Laspidou et al.⁶. The rate of soluble fouling-related organics formation is usually modeled as follows^{6, 22}:

$$r_{\text{SMP}} = (k_{\text{UAP}}q + k_{\text{BAP}})X_a \quad (1)$$

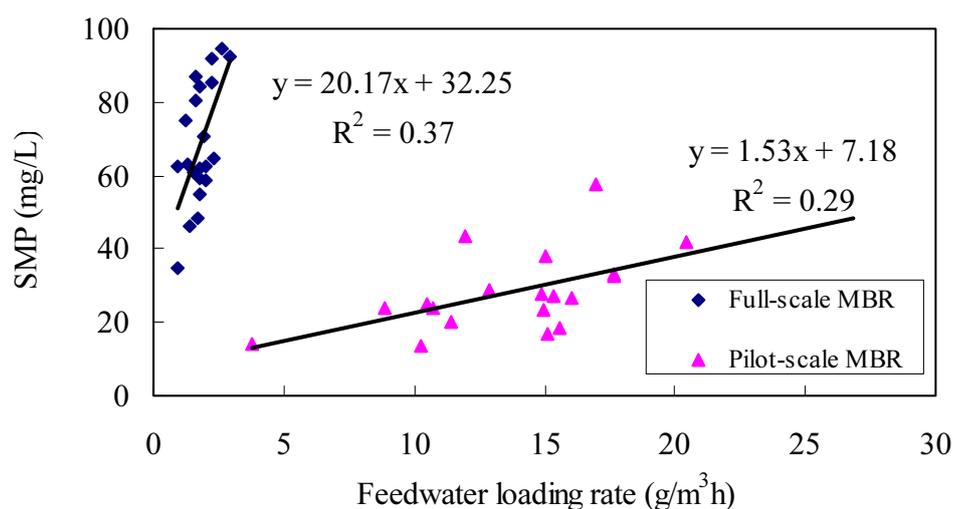
where k_{UAP} and k_{BAP} are formation rate coefficients for UAP and BAP, respectively; q is specific substrate utilization rate; and X_a is biomass concentration. Thus, it can be seen that the production of SMP is proportional to the biomass concentration. Similarly, the biodegradation rate of SMP is also directly determined by biomass concentration. In addition, the generation of bound EPS is growth-associated and is produced in direct proportion to substrate utilization, and the bound EPS hydrolyzed form BAP⁶. The feedwater and sludge concentration therefore play great role in the formation and accumulation of fouling-related organics.

3.2 Influence of feedwater loading rate on fouling-related organics

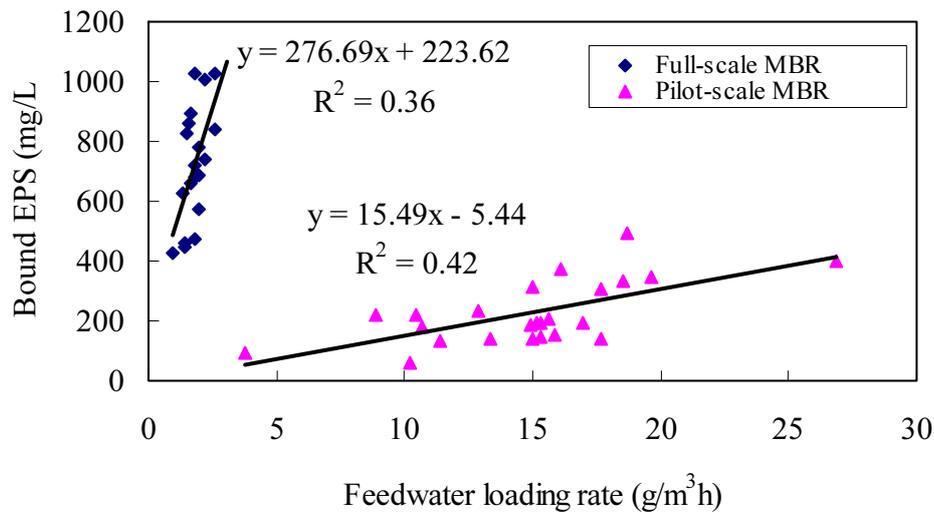
Over a long-term monitoring of the MBRs, it was found that the soluble fouling-related organics or SMP in sludge supernatant of the two MBRs had close relation with the feedwater loading rate (Figure 3a). Here, the feedwater and soluble fouling-related organics were expressed as the sum of PN and PS. The good correlation reveals that the formation of soluble fouling-related organics can be controlled or influenced by feedwater. From Figure 3a it can be seen that as the feedwater loading rate increased, the soluble fouling-related organics increased in both pilot-scale MBR and full-scale MBR. This phenomenon is in agreement with a previous investigation by Meng et al.²³ who found that the soluble fouling-related organics increased from 24.6, to 38.68, and to 51.48 mg-COD/L when the feedwater loading rate increased from 0.70-0.84, to 1.1-1.4, and to 1.7-2.1 kg-COD/(m³ day), respectively. It must be pointed out that the UAP, which is one type of SMP, is produced as a direct result of substrate utilization⁶. According to this concept, it is not surprising to find that an increase in feedwater loading rate would lead to the generation of more soluble fouling-related organics.

From Figure 3a it also can be noticed that the increase of feedwater loading rate had a much stronger influence on the formation of soluble fouling-related organics or SMP in the full-scale MBR than that in the pilot-scale MBR. It implies that the soluble fouling-related organics in the full-scale MBR was highly sensitive to the feedwater loading rate even though the full-scale MBR had a much lower feedwater loading rate (see Figure 3a). This also suggests that the production rate of UAP in full-scale MBR was 13 times than that in the pilot-scale MBR. The high production rate of SMP in full-scale MBR could be explained either by feedwater composition/concentration or by the MBR itself (i.e., the high SRT, high MLSS concentration, and MBR configuration). One of the main reasons is the feedwater used for the full-scale MBR, the PN, PS and COD concentration of which was nearly twice of that used for the pilot-scale MBR. In fact, it is hard to compare the two MBRs in detail due to the significant difference. In this study, the two MBRs are mainly used to present reproductive results or phenomena.

Figure 3b plots bound EPS as a function of feedwater loading rate for the two MBRs. Again, the bound EPS was proportional to the feedwater loading rate. As such, the influence of feedwater loading rate on bound EPS was similar to SMP. The high feedwater loading rate would accelerate the growth and metabolism of biomass and then, enhance the synthesis of bound EPS. Under high feedwater loading rate, the biomass is not able to consume the bound EPS timely, and consequently it results in the overstock of bound EPS. Meng et al.²³ reported that there were high bound EPS concentration and high sludge viscosity as feedwater loading rate increased. Nagaoka et al.²⁴ investigated the influence of feedwater loading rate on membrane fouling caused by EPS deposition in two parallel submerged membrane bioreactors, and found that the high organic loading rate (1.5g-TOC/(L day)) showed a sudden increase of the pressure and a decrease of flux after 40 days, which could not be recovered even by membrane cleanings, while the low organic loading rate (0.5g-TOC/(L day)) showed little increase of the pressure until 120 days. However, the influence of feedwater loading rate on the formation of bound EPS did not mentioned in the study.



(a)



(b)

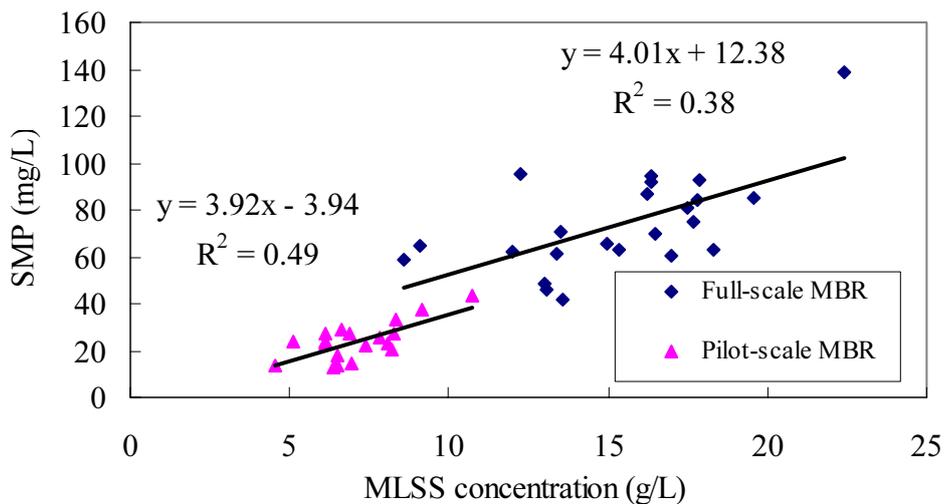
Figure 3 Concentration of (a) SMP and (b) bound EPS over feedwater loading rate

3.3 Influence of MLSS concentration on fouling-related organics

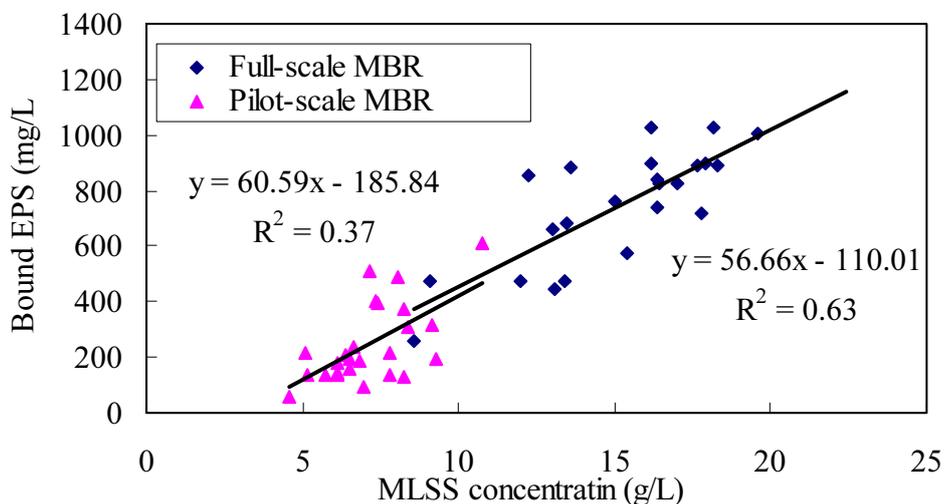
MLSS concentration has always been paid much attention during the study of MBR fouling. Previously, the role of MLSS concentration in MBR fouling was mostly attributed to the change of sludge viscosity. The increase in MLSS concentration and the corresponding rise in sludge viscosity has a negative impact on membrane permeability²⁵. Another possibility to elucidate fouling mechanism of MLSS concentration would be the influence of MLSS concentration on the production of SMP and bound EPS. As shown in Figure 4 it can be seen that both SMP and bound EPS increased with increasing MLSS concentration, indicating that MLSS concentration might be an important factor determining the formation of fouling-related organics. From Equation (1) it can also be expected that the increase of MLSS concentration would lead to the increase of SMP. As expected, the more the biomass is, the more fouling-related organics would be produced during the metabolism process. Moreover, the increase of MLSS concentration will decrease the efficiency of oxygen transfer because the increased viscosity also reduces the efficiency of mass transfer of oxygen and can therefore affect dissolved oxygen (DO) concentration^{26, 27}. Typically, the decrease of DO concentration could lead to high amount of SMP and severe membrane fouling^{16, 28}.

It is interesting to notice that the SMP in both full-scale MBR and pilot-scale MBR was equally influenced by the MLSS concentration (see the slope of the trendline in Figure 4). This phenomenon is different from the Figure 3. Of particular interest is the influence of MLSS concentration on bound EPS. It can be seen that the two trendlines in Figure 4b are close to each other. Therefore, the MLSS concentration might be a more acceptable indicator to the formation of fouling-related organics, especially when compared with feedwater loading rate. It is expected that the influence of SRT, HRT, and feedwater loading rate on MBRs converges at the change of MLSS concentration. For example, the high SRT, or low HRT, or high feedwater loading rate will result in a high MLSS concentration. To confirm whether MLSS concentration can be used as an indicator for SMP formation, a more detailed study needs to be performed by collecting more data from different MBR plants. Lastly, of high significance is the sludge growth, which

involves lag phase, exponential phase, stationary phase and death phase. It is expected that each growth phase of sludge might be of different influence on the formation of fouling-related organics. In this study, it was observed that when a great deal of sludge was discharge in the full-scale MBR, both bound EPS and SMP decreased sharply (data not shown). It is probably because the sludge growth shifted to a new growth phase, during this phase the sludge needs to consume a great deal of organic matter or fouling-related organics to generate new cells. During this phase, the sludge would produce some UAP, but produce little BAP.



(a)



(b)

Figure 4 Concentration of (a) SMP and (b) bound EPS over MLSS concentration

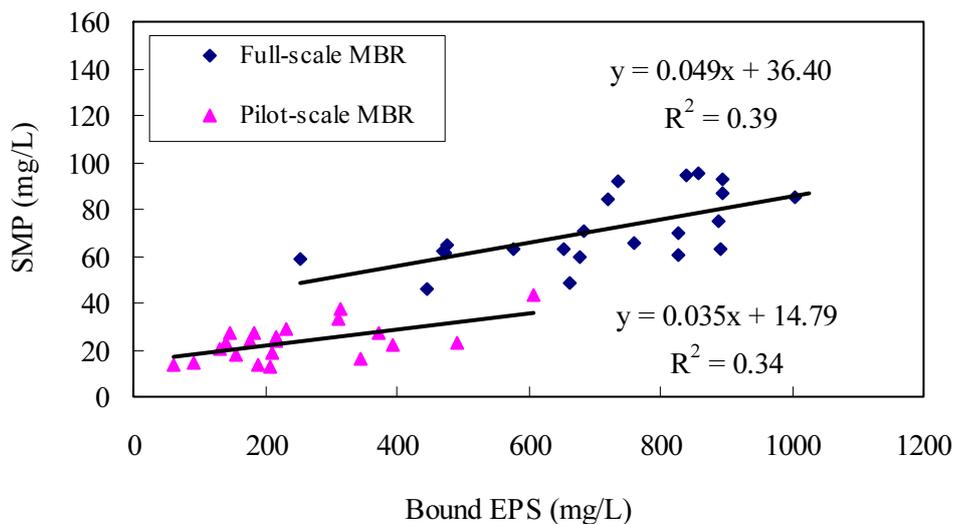


Figure 5 Relationship between bound EPS and SMP

Figure 5 shows SMP concentration as a function of bound EPS concentration. As shown in Figure 5, the relationship between SMP and bound EPS was evident for both of the two MBRs, demonstrating that there was equilibrium between SMP and bound EPS. This evidence is in agreement with one previous study by ²⁹, which reported that SMP has close relationship with bound EPS. Bound EPS could be dissolved/hydrolyzed by bacterial hydrolyze, adding to a pool of hydrolysis products that called soluble EPS ³⁰. In addition, our current study also confirms that the PN and PS derived from bound EPS are mainly composed of slowly biodegradable matter; however, the PN and PS derived from feedwater are mainly composed of readily biodegradable matter ³¹, indicating that the bound EPS is of high importance to the formation of SMP. On the other hand, the soluble fouling-related organics can also deposit or adsorb onto the sludge flocs and then, transform into bound EPS. Generally, due to the hydrophobic nature of PN, the PN has a higher affinity with sludge flocs than PS does. That is why the PN/PS ratios of bound EPS in the two MBRs reach 5. It must be addressed that besides feedwater loading rate, MLSS concentration and bound EPS, the formation and elimination of SMP is also determined by the ambient conditions, e.g. temperature and nitrate ¹⁶.

3.4 Discharge of soluble fouling-related organics

Besides the biological process, the membrane filtration acts as a selective separation for SMP. Part of SMP is likely to pass membrane if it is small enough, and some part of which would be retained by membrane. It can be concluded that the membrane filtration plays significant role in the accumulation and discharge of SMP, and thereby might cause the change of bound EPS due to the close relationship between SMP and bound EPS. In addition, the discharge of PN-rich or PS-rich water brings additional problems to the local environment (e.g., the occurrence of dissolved organic nitrogen (DON)). Therefore, it is highly desirable to go to the details of membrane separation to know the discharge behavior of SMP and in what way the SMP could be discharged.

In Figure 6 the discharged PN and PS of both MBRs is plotted as a function of the PN and PS in the activated sludge supernatant, respectively. For the PN, a good correlation between discharged PN and the PN in sludge supernatant was observed. It has to be

noticed that the soluble PN in the pilot-scale MBR relied on its concentration in sludge supernatant more dramatically. For the PS, it can be seen that there was a good correlation for full-scale MBR. But, for the pilot-scale MBR, the discharged PS was independent of the PS concentration in sludge supernatant. This evidence suggests that the PS in the permeate of the pilot-scale MBR could be maintained at a steady level, which might be due to the low PS loading in sludge and the large size of PS. Even so, as discussed in Section 3.1, the PS in the sludge supernatant of the pilot-scale MBR had a higher discharge rate. The results in Figure 6 reveal that the discharge of SMP would help to mitigate the accumulation of fouling-related organics in the bioreactor. But, the SMP larger than membrane pore is still retained in the bioreactor, and the retained SMP account for MBR fouling¹¹. Additionally, the discharged PS and PN would concern the implementation of post-treatment for water recycling, e.g. the RO fouling in MBR+RO process. Thus, the perfect approach is to do every possible to control the formation of SMP, dosing coagulants, for example.

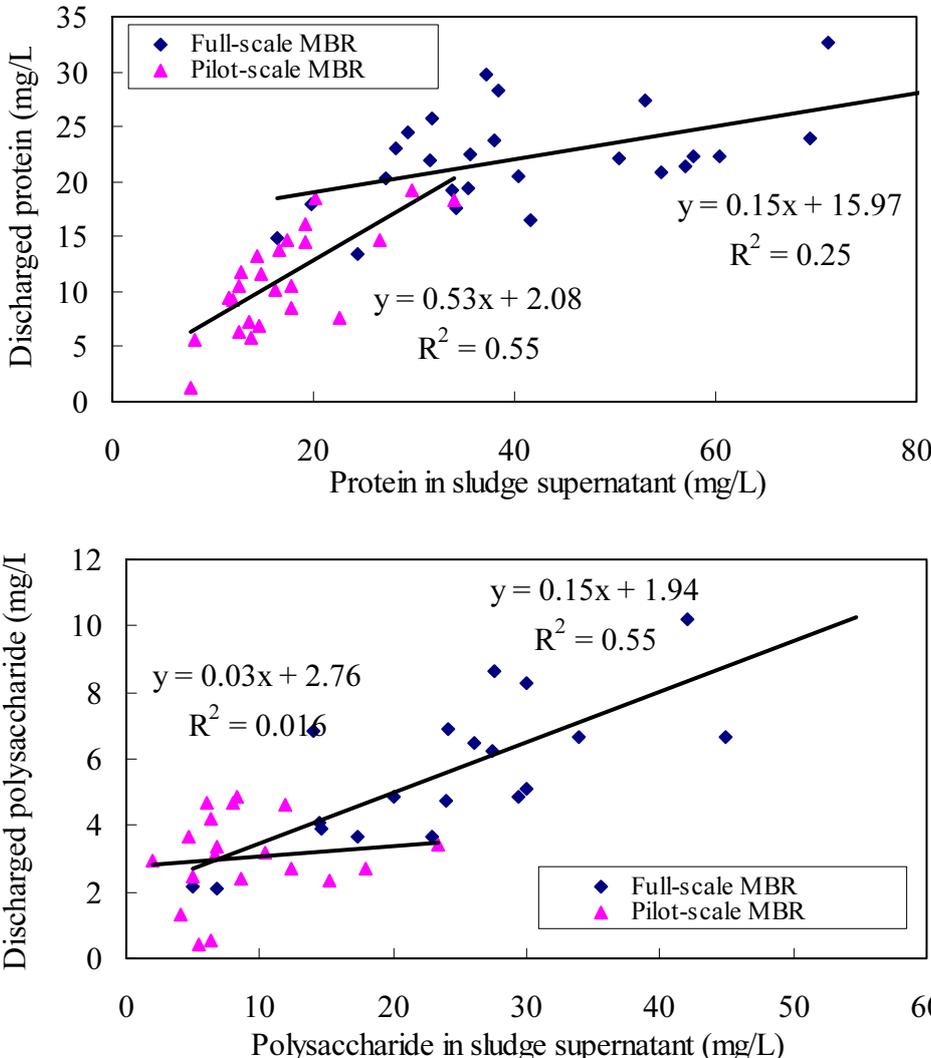


Figure 6 Discharged (a) PN and (b) PS over PN and PS in sludge supernatant

4 Conclusions

The main conclusions of this study can be drawn as following:

- (1) The fouling-related organics in different scale MBRs differed significantly.
- (2) The feedwaster loading rates and MLSS concentrations in the MBRs would affect the formation of EPS and SMP substantially.
- (3) The discharge of SMP-PS and SMP-PN likely depended on their levels and the MBR scale. The discharge of SMP would help to mitigate the accumulation of fouling-related organics in the bioreactor.

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