Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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Online publication date: 23 April 2010

To cite this Article Wang, Xiaochang C. , Liu, Qiang and Liu, Yongjun J.(2010) 'Membrane Fouling Control of Hybrid Membrane Bioreactor: Effect of Extracellular Polymeric Substances', Separation Science and Technology, 45: 7, 928 — 934
To link to this Article: DOI: 10.1080/01496391003657030
URL: http://dx.doi.org/10.1080/01496391003657030

Separation Science and Technology, 45: 928–934, 2010

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496391003657030

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Membrane Fouling Control of Hybrid Membrane Bioreactor: Effect of Extracellular Polymeric Substances

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A hybrid membrane bioreactor (HMBR) was developed by adding biofilm carriers into a conventional membrane bioreactor (CMBR), and a study was conducted on the characteristics of the activated sludge in the HMBR and CMBR with attention paid to the effect of extracellular polymeric substances (EPS) on membrane fouling. As a result, lower EPS concentrations were detected from the HMBR than the CMBR, bringing about a substantial improvement in the sludge properties. The larger particle size, more compact particle structure, and better settleability of the sludge in the HMBR resulted in lower cake layer resistance and much slower TMP increase.

Keywords activated sludge; extracellular polymeric substances; hybrid membrane bioreactor; membrane fouling

INTRODUCTION

Membrane fouling is the key problem that restricts the wide application of a membrane bioreactor (MBR) (1–3) and many studies have thus been concentrated on innovative technologies which can assist membrane fouling control. In the author's previous study (4), a hybrid membrane bioreactor (HMBR) was developed by introducing biofilm carriers into an ordinary MBR with submerged hollow fiber microfiltration (MF) membranes. Such a novel method effectively increased the total quantity of biomass in the reactor and enhanced the organic removal. Due to the simultaneous existence of suspended and attached biomasses in the HMBR, nitrification and denitrification effect was improved. Another advantage of the HMBR over the conventional membrane bioreactor (CMBR) was that it slowed down the increase of transmembrane pressure (TMP) so that a higher flux could be maintained without frequent chemical cleaning. However, a topic still remains on the mechanisms of membrane fouling control by the HMBR.

Regarding the fouling mechanisms of membrane bioreactors, attention was paid to the organic foulants generated from biological processes, and the extracellular polymeric substances (EPS) were recognized to contribute much to membrane fouling in many studies (5–7). EPS are microbial products resulting from active bacterial secretion and cell lysis and composed of a variety of organic substances such as carbohydrates, proteins, humic substances, uronic acids, and nucleic acid substances (8). The EPS located at or outside the cell surface are often called bound EPS which participate in the formation of microbial aggregates (9–10). In a bioreactor, bacteria in the activated sludge suspension and floc matrix are likely to have a dynamic double-layered EPS structure of loosely bound EPS diffused from the tightly bound EPS that surround the cells (11). There are also soluble cellular components as a product of the dissolution of the bound EPS. These are called soluble EPS or soluble microbial products (SMP) (10,12).

By definition the bound EPS (B-EPS) may have a strong influence on the characteristics of the activated sludge, for the filterability of the sludge was reported to decrease with increasing B-EPS (13). It was also shown experimentally that as the amount of the B-EPS increased, the specific cake resistance became higher (14). The increasing EPS concentration was found to be one of the factors causing flux decline in the membrane-coupled activated sludge (15).

These previous studies suggested that analyzing EPS could assist much in an investigation of membrane fouling mechanisms. In order to understand how the HMBR provided a favorable condition for membrane fouling control, the authors conducted a comparative study on the HMBR and CMBR under the same operation condition. Attention was paid to the composition of EPS in each of the reactors, and the effect of EPS on the characteristics of activated sludge particles and membrane fouling.

Received 1 November 2009; accepted 31 December 2009.
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MATERIALS AND METHODS

Raw Wastewater Characteristics

The experimental work for this study was conducted with a pilot-scale membrane bioreactor (MBR) installed at a domestic wastewater treatment plant in Xi'an, China. The raw wastewater, after flowing through a sand settler and a coarse screen, was fed to the pilot MBR as the influent. Its characteristics during the one-year experimental period are shown in Table 1.

Experimental Set-up and Operation Conditions

Figure 1 is the schematic diagram of the pilot MBR system used in this study. It consisted of a rectangular aeration tank equipped with a submerged hollow fibre micro-filtration (MF) membrane module (pore size 0.2 μm, Tianjin Motimo Membrane Technology Ltd.) and associated pumping, aerating, flow rate control, and measurement devices. The aeration tank was partitioned by a perforated wall into two rooms, one for aeration and another for accommodating the membrane module. The aeration room could perform the function of a conventional reactor without the addition of biofilm carriers, or the function of a hybrid reactor when biofilm carriers (Kaldnes K3, AnoxKaldnes Corporation, Norway) were added. The former was the operation mode of a conventional membrane bioreactor (CMBR) and the latter was the operation mode of a hybrid membrane bioreactor (HMBR). The membrane room was also equipped with air diffusers at its bottom for providing shear force on the submerged membrane for fouling control.

Table 2 shows the operation conditions of the pilot system under the CMBR and HMBR operation modes. All the operational parameters were almost identical for the CMBR and HMBR, except for the increase of the total biomass for the HMBR due to the attached biomass formation on the biofilm carriers.

EPS Extraction and Analysis

EPS extraction in this study was conducted using a series of methods as shown in Fig. 2. These methods were basically those frequently adopted in many studies (8,15–23), but were modified by the authors in order to separate EPS into different parts according to their affinity with the biomass, i.e., the activated sludge.

The sludge samples for EPS extraction were the mixed liquor collected from the aeration room of the CMBR or HMBR with MLSS concentration shown in Table 2. EPS extraction was conducted in the following ways to obtain different microbial products.

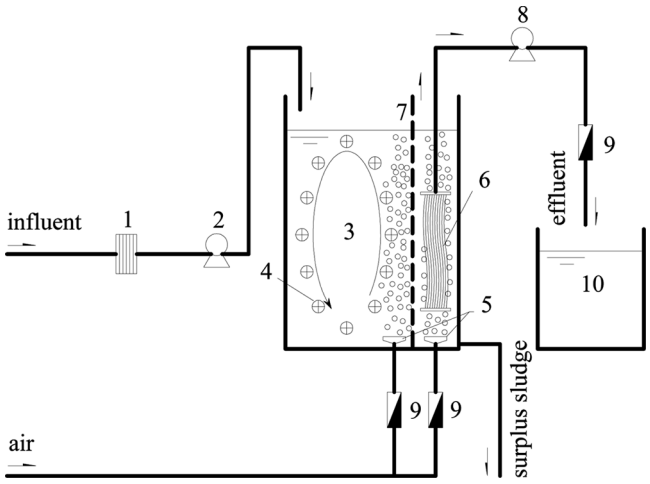

FIG. 1. Schematic diagram of the pilot system (1 – screen; 2 – feed pump; 3 – aeration room; 4 – biofilm carrier under the HMBR operation mode; 5 – air diffuser; 6 – hollow-fibre MF module; 7 – perforated wall; 8 – suction pump; 9 – flowmeter; 10 – permeate tank).

1. Simply by two membrane separations to remove microbial cells (0.2 μm membrane) and low molecular-weight metabolites (dialysis membrane of 3500 Da), the liquid obtained was considered to contain mainly the soluble microbial product which might roughly represent the soluble EPS (S-EPS).
2. By high speed centrifugation (20000 g) followed by the two-stage membrane filtration above mentioned, the microbial product loosely bound to the cells could be extracted as well as the soluble one, so the liquid obtained was considered to contain both the soluble and physically extractable microbial products which might roughly represent the S-EPS and loosely bound EPS (LB-EPS).
3. The residues from the above-mentioned centrifugation and membrane filtration were resuspended using a buffer solution (2 mM Na_3PO_4, 4 mM NaH_2PO_4, 9 mM NaCl, 1 mM KCl, pH 7). The suspension was heated at 60°C for 30 min and then centrifuged and filtered with 0.2 μm and 3500 Da membranes. The liquid finally obtained contained mainly the heat extractable microbial product which might roughly represent the tightly bound EPS (TB-EPS).

TABLE 1
Characteristics of the influent to the pilot MBR

COD (mg/L)	BOD_5 (mg/L)	NH_4^+–N (mg/L)	TN (mg/L)	TP (mg/L)	TSS (mg/L)	pH	Temperature (°C)
240–896	185–423	20.3–46.8	22.5–54.1	4.3–13.2	200–1210	7.0–7.5	13.1–25.7

TABLE 2
Operation conditions of the pilot system

Operation mode	HRT (h)	DO in the aeration room (mg/L)	Membrane flux ($L/m^2 \cdot h$)	Suspended biomass (MLSS) (mg/L)	Attached biomass (mg/L)	Total biomass (mg/L)
CMBR	10	0.5–2.1 (1.2)	10	3850–4248 (4058)		3850–4248 (4058)
HMBR	10	0.4–2.3 (1.1)	10	3810–4350 (4021)	1605–1750 (1687)	5415–6100 (5708)

Note. Value in the brackets as the average.

The concentration of each part of the EPS extracted was measured as the total organic compound (TOC) using a TOC analyzer (1030 Aurora Sin, OI Analytical).

Characterization of the Physical Properties of the Activated Sludge

In this study, the supernatant turbidity of the mixed liquor from the aeration room was used as a parameter to characterize the flocculablity of the activated sludge. The sludge volume index (SVI) was used as a parameter to characterize the settleablity of the activated sludge. The morphological characteristics of the sludge particles were observed using an optimal microscope (BX60, Olympus) equipped with a digital camera (Infinity 3, Olympus), and the size distribution of the sludge particles was analyzed using a laser granularity distribution analyzer (LS 230/SVM+, Coulter, USA).

The suspended biomass was characterized by mixed liquor suspended solids (MLSS) in the aeration room, and the attached biomass was analyzed by direct measurement of the attached solid weight following Luostarinen et al. (24).

Characterization of Membrane Resistances

In order to characterize the membrane resistance, attention was mainly paid to the specific resistance of the cake layer which was analyzed by collecting sludge from the cake layer on the outer surface of the hollow fiber, measuring its filtering property using a filtration device (Model 8200, Amicon, USA), and calculating its specific resistance to filtration (25). The total membrane resistance (R_t), pore blocking resistance (R_p), and cake layer resistance (R_c) were determined following Darcy's Law using the flux and transmembrane pressure (TMP) measured at different stages of membrane filtration (26).

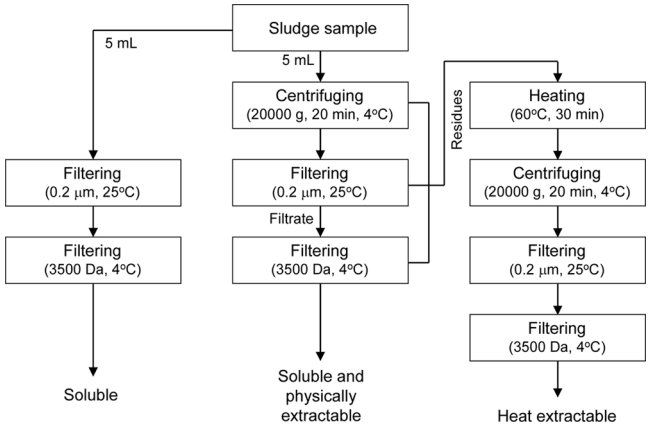

FIG. 2. Methods of EPS extraction.

RESULTS AND DISCUSSION

Characteristics of EPS Distribution

In this study, we adopted the modified physical and heating methods for EPS extraction without adding any chemical extractant. The extracted EPS included the soluble part, soluble and physically extractable part, and the heat extractable part as shown in Fig. 2. From the measured TOC of each part, we obtained gross indicators for S-EPS (the soluble), LB-EPS (the difference of the soluble and physically extractable with the soluble), and TB-EPS (the heat extractable). The sum of the latter two was the bound EPS (B-EPS). By collecting sludge samples from the CMBR and HMBR under stable operation conditions and analyzing EPS by the abovementioned methods, a result was obtained as shown in Fig. 3. The total EPS from the CMBR was measured as 86.7 mg TOC per g VSS, and that from the HMBR was 72.0 mg TOC per g VSS. There was a decrease of 17% of the total EPS by HMBR operation. Regarding the proportion of S-EPS and B-EPS, in the case of CMBR it was 16.4% vs. 83.6% while in the case

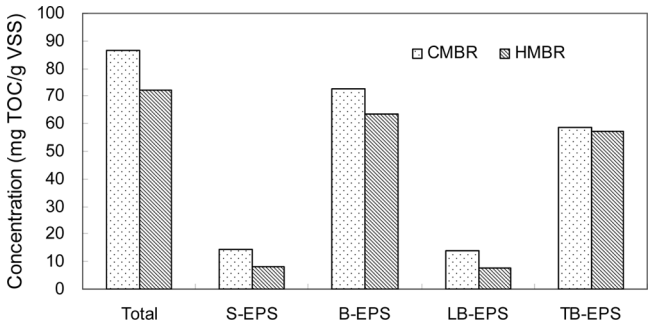

FIG. 3. EPS analysis result for CMBR and HMBR under stable operation conditions.

of HMBR it was 11.5% vs. 88.5%. It was apparent that using the extraction methods shown in Fig. 2, the B-EPS took more than 80% of the total EPS and of the B-EPS more than 80% was TB-EPS which could only be extracted by heating.

On the other hand, in the procedures of EPS extraction adopted in this study (Fig. 2), the soluble part obtained (the S-EPS in Fig. 3) was in fact the filtrate of the supernatant of the activated sludge. The measured TOC might have been influenced by the residual organic matter in the supernatant other than soluble microbial product. Comparing with this, the B-EPS obtained was considered more reliable because it contained mainly the microbial product attached to the activated sludge which could only be extracted by centrifuging and heating. The analysis result of the B-EPS was used in the following sections for a discussion of the effect of EPS on the properties of the activated sludge.

Effect of B-EPS on the Physical Properties of the Activated Sludge

During the pilot experiment of both CMBR and HMBR, sludge samples were continuously collected for EPS analysis and the supernatant turbidity and SVI of the sludge were measured for an investigation of the flocculability and settleability of the sludge particles. As a result, linear relationships could be roughly drawn between B-EPS concentration and the supernatant turbidity and SVI as shown in Figs. 4 and 5. It was apparent that as the B-EPS concentration became higher, higher turbidity was measured from the supernatant, indicating a poorer flocculability of the activated sludge with fine particles. Meanwhile, at higher B-EPS concentration, SVI also became higher, indicating a poorer settleability of the sludge floc.

Figure 6 compares the macroscopic images of the sludge floc in the CMBR and HMBR under stable operation conditions. With a higher B-EPS concentration in the CMBR (about 73 mg TOC/g VSS) the sludge floc appeared looser,

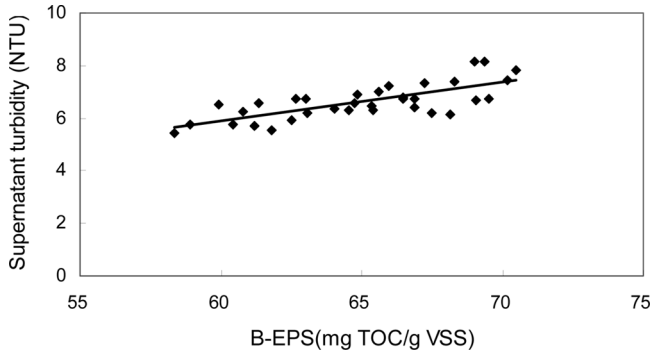

FIG. 4. Effect of B-EPS concentration on supernatant turbidity of the activated sludge.

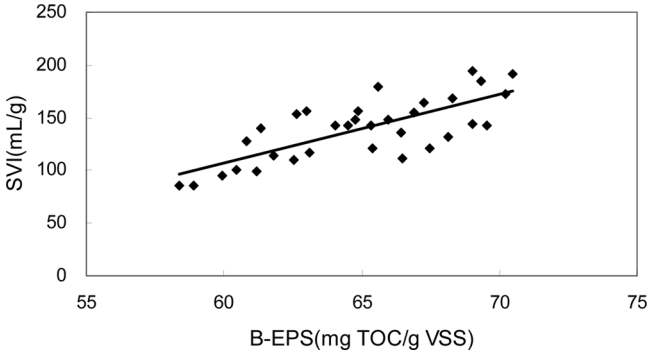

FIG. 5. Effect of B-EPS concentration on SVI of the activated sludge.

while with a lower B-EPS concentration in the HMBR (about 64 mg TOC/g VSS) the sludge floc appeared denser. By particle size distribution analysis using a laser granularity distribution analyzer, Fig. 7 was obtained for a comparison of the sludge floc in the CMBR and HMBR. The average diameter of the floc in the HMBR was measured as 56.8 μm which almost doubled the average diameter of 29.3 μm in the CMBR, indicating that much better flocculation was performed as the B-EPS concentration was lower.

Membrane Resistance and Transmembrane Pressure

Table 3 shows the calculated membrane resistance for CMBR and HMBR after 57 days from the start of operation as TMP in the CMBR increased to 20 kPa that corresponded to a prescribed maximum pressure loss when the membrane had to be chemically cleaned.

As shown in the table, after the same period of operation under a similar operation condition, R_t of the membrane in the HMBR was less than half (48%) of that in the CMBR. In both the CMBR and HMBR R_c was much higher than R_p, indicating that membrane filtration was affected dominantly by the cake layer resistance. However, in the case of CMBR the ratio of R_c/R_p was 5.49 while in the case of HMBR it was 4.22. This provided evidence that due to a substantial decrease of the B-EPS concentration in HMBR, the accumulation of the cake layer resistance was effectively slowed down. The improvement of the physical properties of the activated sludge as mentioned in the former section could be considered as the main reason for this.

The decrease of the membrane resistance, especially the cake layer resistance by HMBR operation, eventually brought about a much slower increase of TMP during filtration. As shown in Fig. 8, on day 57 when the TMP reached 20 kPa in the CMBR, the TMP value was only 9.7 kPa in the HMBR. The filtration cycle, i.e., the continuous operation time before chemical cleaning could thus be prolonged to 142 days.

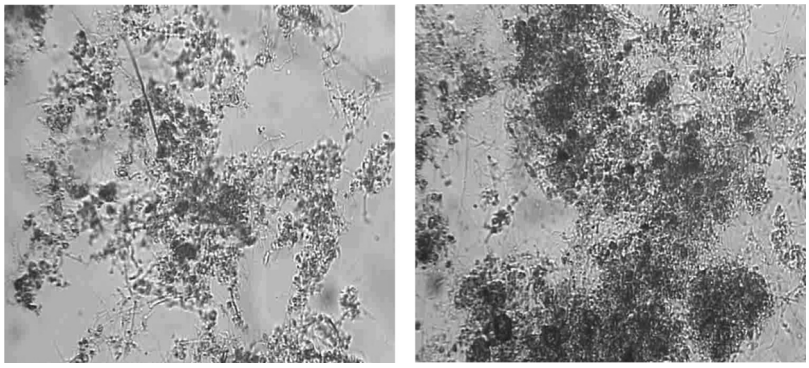

FIG. 6. Image of the sludge floc in the CMBR (left) and that in the HMBR (right).

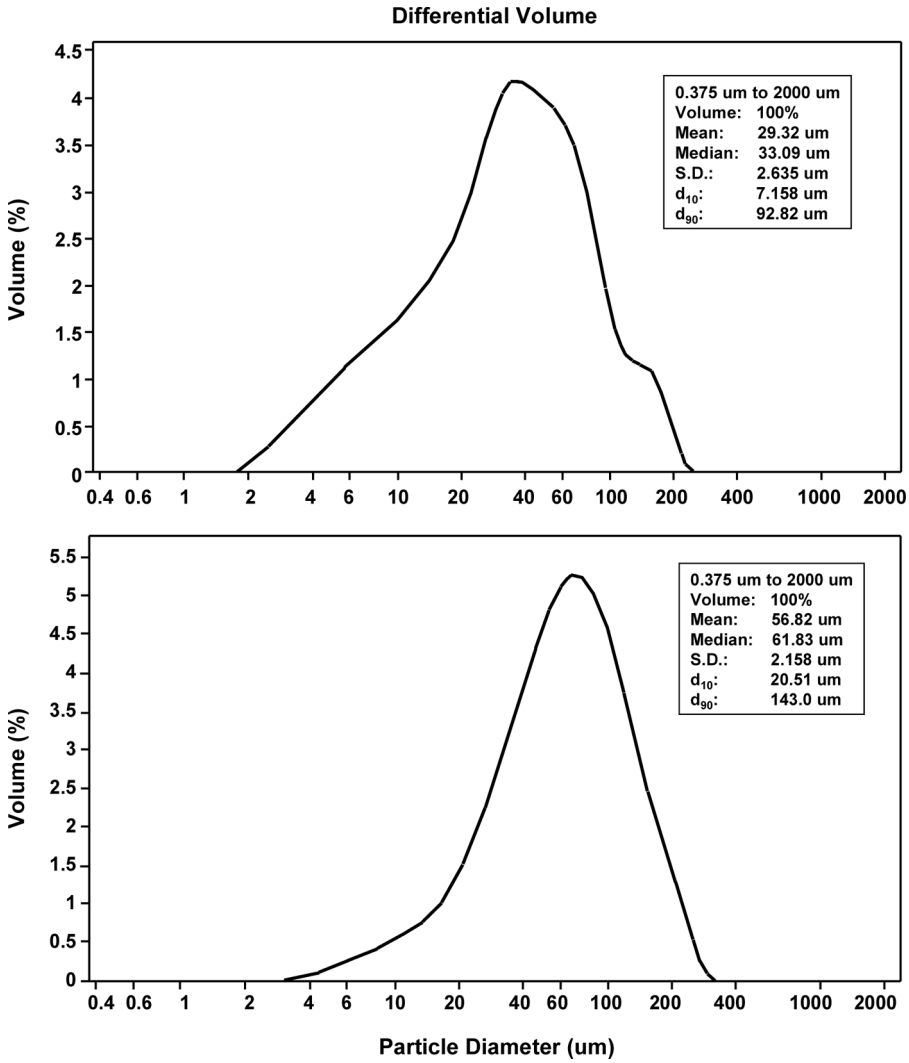

FIG. 7. Particle size distribution of sludge floc in the CMBR (top) and HMBR (bottom).

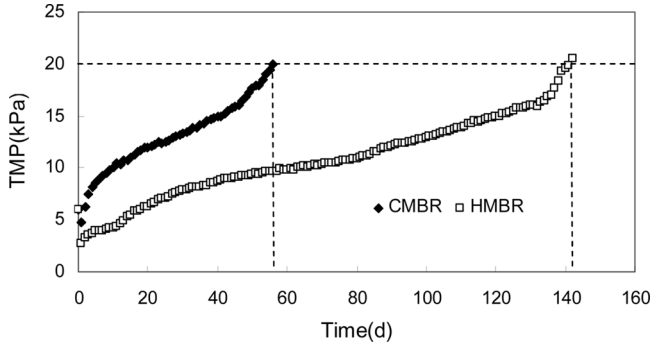

FIG. 8. Variation of TMP in the CMBR and HMBR.

Possible Mechanisms of Membrane Fouling Control by HMBR

From the discussion in the former sections we confirmed that, firstly the EPS concentration in the HMBR was lower than that in the CMBR; secondly, the physical property of the activated sludge seemed to be much improved in the HMBR as indicated by its better flocculability, settleability, size distribution, and floc structure; and thirdly, the accumulation of membrane resistance, especially the cake layer resistance, was greatly slowed down for the membrane under the HMBR operation mode. From these results, it may be reasonable to draw a linkage between EPS concentration, sludge property, and cake layer formation on the membrane. Due to the addition of the biofilm carriers, both the suspended biomass and attached biomass could coexist in the bioreactor. In addition to some apparent change in the bioreactor such as an increase of biomass quantity and prolonged sludge retention time, important changes may also have occurred in the microbial structure for the microbes were found to be more diverse in the biofilm than that in the suspended biomass (27). Because EPS are substances produced by bacteria in the activated sludge (10), the change in microbial structure would in due course affect the production of EPS in the bioreactor, though detailed information is not yet available in this regard. As B-EPS, that by definition are microbial products, located at or outside the cell surface, are found to be dominant in the extracted EPS, their effect on the formation of microbial aggregates would be important (9,10). Because these microbial aggregates are the main component of the activated sludge, their property may have a strong effect on the physical properties of the sludge. This may further affect the formation of cake layers on the membrane surface.

TABLE 3
Comparison of membrane resistance for CMBR and HMBR

Operation mode	Total resistance (R_t)	Pore blocking resistance (R_p)	Cake layer resistance (R_c)
CMBR	$5.22 \times 10^{12}\,m^{-1}$	$7.62 \times 10^{11}\,m^{-1}$	$4.18 \times 10^{12}\,m^{-1}$
HMBR	$2.51 \times 10^{12}\,m^{-1}$	$4.27 \times 10^{11}\,m^{-1}$	$1.80 \times 10^{12}\,m^{-1}$

Note. Measured on day 57 for both CMBR and HMBR.

CONCLUSION

From this study, the following conclusions could be drawn:

1. By a series of modified filtering, centrifuging, and heating methods, EPS were extracted from the sludge samples of CMBR and HMBR and their composition could be divided into soluble, physically extractable, and heat extractable parts which were taken as S-EPS, LB-EPS, and TB-EPS in this study. It was found that B-EPS (the sum of LB-EPS and TB-EPS) took more than 80% of the total EPS in both CMBR and HMBR, and the HMBR operation resulted in a 17% decrease of the total EPS concentration.
2. The physical properties of the activated sludge such as SVI, particle structure, and size distribution were found to be strongly influence by B-EPS concentration. The lower concentration of B-EPS in the HMBR brought about better flocculability, better settleability, larger floc size, and a more compact floc structure.
3. By analysing the membrane resistance, much difference was found between CMBR and HMBR regarding R_t, R_p, and R_c after the same period of operation. R_t of the membrane in the HMBR was only 48% of that in the CMBR. Although cake layer resistance was found to be dominant in both the CMBR and HMBR, the ratio of R_c/R_p was significantly lower in the HMBR than that in the CMBR. The decrease of the membrane resistance, especially the cake layer resistance by the HMBR operation, eventually brought about a much slow increase of TMP and a prolonged filtration cycle.
4. It may be reasonable to draw a linkage between EPS concentration, sludge property, and cake layer formation on the membrane. The substantial decrease of EPS, especially the B-EPS concentration, could provide an explanation of membrane fouling control by the HMBR operation.

ACKNOWLEDGEMENTS

This study is supported by the National Natural Science Foundation of China (Grant No. 50838005), the National Program of Water Pollution Control (Grant No. 2008ZX07317-004) and the Program for Changjiang Scholars and Innovative Research Team in University (Grant No. IRT0853).

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