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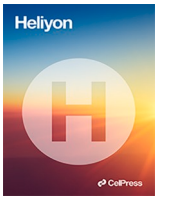

Comparative analysis of membrane fouling mechanisms induced by operation modes of membrane bioreactors with aerobic granular sludge

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ABSTRACT

This experimental work investigated fouling characteristics induced by two different configurations of membrane bioreactor (MBR), which are submerged MBR and sidestream MBR with aerobic granular sludge. Submerged membrane bioreactor with granular sludge (Sub-MGSBR) ran the longest operation time 61 days with a steady overall TMP increase rate; Sidestream membrane bioreactor with granular sludge (SS-MGSBR) performed only 39 days, which exhibited Sub-MGSBR had more efficiently retarding membrane fouling. In both membrane bioreactors with flocculent sludge (MFSBRs) as a control, membrane foulants were compact, and cake resistance was the dominant fouling factor. In MGSBRs, however, pore blocking resistance turned out the key fouling factor. Especially in Sub-MGSBR, it went beyond 75%, and there was the most conglomeration of microorganisms of foulants with the highest porosity. Extracellular polymeric substances (EPS) content of foulants proved membrane fouling was hardly just for granules accumulation into cake but microorganisms' growth in MGSBRs.

1. Introduction

Membrane bioreactor features many advantages over the conventional activated sludge systems for municipal and industrial wastewater treatment [1,2] and resource/energy recovery [3]. For successful application of membrane bioreactor (MBR), it must be maintained a steady and efficient long-term operation of the system. The membrane fouling is the major problem to reach this purpose, which can lead to a high capital expenditure and operating expenses for the long-time running of membrane module [4,5].

The common MBR system was to have the membrane module immersed in flocculent activated sludge. Recently, a new form of MBR, which couples membrane separation with not activated sludge but aerobic granular sludge, is being considered as a promising wastewater treatment technology for retarding the membrane fouling [6]. Compared with traditional MBRs, key membrane fouling factor of MGSBR was distinct. Zhou et al. once reported that severe membrane pore-blocking while the activated sludge could cause severe cake fouling [7]. It might be for that the smaller the size of aggregates, the greater the resistance of the cake formed [8]. Aerobic granular sludge is not only bigger but also more compact than activated flocculent sludge, so it is hardly to compress and form serious cake on the membrane surface eventually.

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Membrane fouling was a complicated course. So far it is generally accepted that membrane fouling results from cake layer and membrane pore clogging. In addition, microbial growth on the membrane surface weakens filtering ability [9]. There are many factors, which play important roles in affect membrane fouling. Besides sludge property and membrane characteristics, it is about operational conditions of membrane modules finally yet importantly [10]. There are commonly two different configurations of MBR, i.e. submerged MBR (Sub-MBR) and sidestream MBR (SS-MBR). Aerobic granular sludge with regular and compact structure can be regarded as a special form of biofilm, which is commonly developed by aggregation of a variety of microorganisms [11], and can separate sludge retention time (SRT) from hydraulic retention time (HRT). Therefore, there is no need of sludge circulation in SS-MGSBR, which is much more economical than traditional SS-MFSBR. Meanwhile, SS-MGSBR was very valuable for maintaining stability of aerobic granules in practical application, for stability of aerobic granules is the guarantee of membrane fouling control.

Right now, the comparison of membrane fouling between submerged and sidestream MBR with aerobic granular sludge in one integrated system has still not reported yet. Therefore, the objective of this experiment is to compare the performance and membrane fouling characteristics between submerged MBR and sidestream MBR with flocculent and aerobic granular sludge in an integrated system. A series of analyses including transmembrane pressure (TMP), membrane fouling resistance distribution, particle size distribution, three dimensional excitation–emission matrix (EEM) fluorescence spectroscopy and so on were employed in order to gain insights into the membrane fouling behaviors of MBRs. Furthermore, observation of fouling layer is to investigate the structure of foulants on the membrane surface.

2. Materials and methods

2.1. MBR and operational control

The experiment was conducted in a MBR operated as sequencing batch reactor. In order to get a completely same condition, the submerged and sidestream MBRs were integrated in one system, which was described in Fig. 1. The first Sub-MGSBR was operated in sequencing mode at cycle time of 6 h, and the synthetic wastewater was used as the feed of MBR system. Then the supernatant of reactor after settlement was partially pumped to the next reactor unit constructing a SS-MBR. MBR with flocculent sludge (MFSBR) was used as a control.

The experiment was carried out at the room temperature. The each Sub-MBR was of a working volume of 60 L, which was 0.55 m in length, 0.35 m in width and 0.4 m in height.

The seed aerobic granules were taken from the reactors in our lab, The seed flocculent sludge was from Gaobeidian municipal wastewater treatment plant located in Beijing, China. The morphology of aerobic granular sludge and activated flocculent sludge was

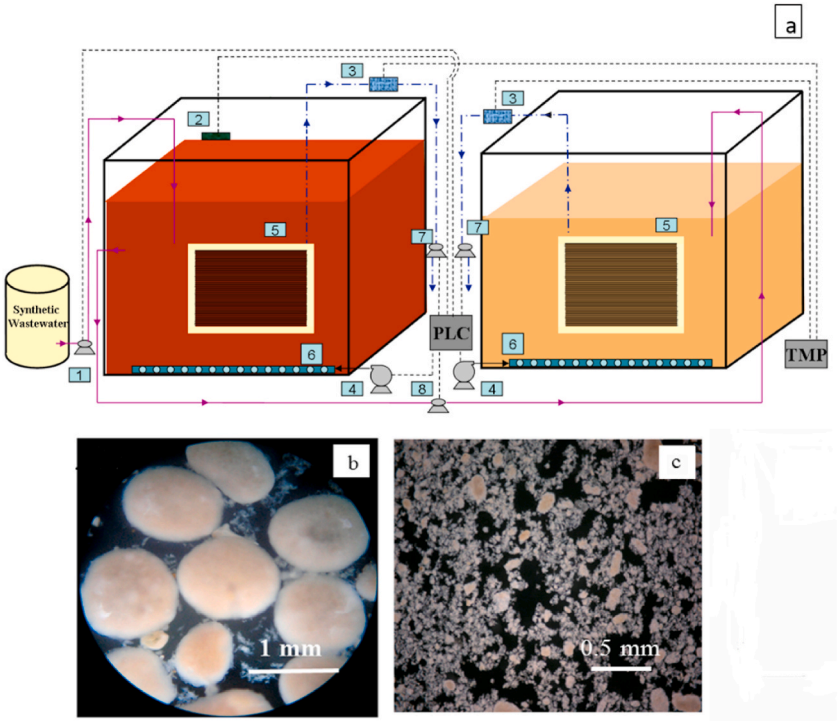

Fig. 1. Schematic diagram of the experimental reactor (a) (1: peristaltic pump; 2: water level sensor; 3: pressure transducer; 4: aeration pump (0.7 m^3/h); 5: membrane module; 6: air diffuser; 7: suction pump; 8: effluent pump (0.24 m^3/h)) and morphology of different sludges (b: aerobic granular sludge; c: flocculent activated sludge).

Table 1
Operational strategies of MBRs in the experiment.

Configuration of MBRs	Bioreactor									Filtration (min)
	MLSS (g/L)	SVI_{30} (mL/g)	Cycle (h)	Feeding (min)	Anaerobic (min)	Aeration (min)	Settling (min)	Discharging (min)	Exchange ratio (%)	
Sub-MGSBR	10.1	14	6	5	65	270	10	10	67	150
SS-MGSBR	10.1	14	6	5	65	270	10	10	67	150
Sub-MFSBR	7.0	70	6	5	65	270	10	10	50	110
SS-MFSBR	7.0	70	6	5	65	270	10	10	50	110

In every cycle, there was mixing process for 1 min by virtue of aeration just after feeding.

shown as Fig. 1. Considering the good settling performance of aerobic granular sludge, the whole experimental systems did not adopted sludge recycle process.

All membrane modules complied with an intermittent suction effluent mode of 8 min-on/2 min-off. The air flow rate 1.5 $m^3/(m^2_{membrane\ area} \bullet h)$ was not only to provide oxygen for microorganism for biodegradation of wastewater but also to produce shear stress on the membrane surface. The flux of membrane permeation was at 20 $L/(m^2\ h)$. The hollow-fiber microfiltration membranes with a pore size of 0.4 μm (Mitsubishi Rayon, Tokyo, Japan) are made of hydrophilized polyvinylidene, and each membrane module (405 × 160 mm) had an effective area of 0.5 m^2. To simulate the practical operation, the whole fouling development of each integrated MBR was monitored on-line, and membrane modules should exit and need be cleaning once TMP was beyond 50 kPa. Every integrated operational system was controlled by programmable logic controller (PLC). Table 1 exhibited the details of operational strategies in different configuration of the experiment.

The synthetic wastewater was not real one. And synthetic wastewater containing sodium acetate as the sole carbon source was fed with the following compositions: CH3COONa, 819.20 mg/L; NH4Cl, 152.85 mg/L; KH2PO4, 70.21 mg/L; NaCl, 160.00 mg/L; MgSO4·7H2O, 80.00 mg/L; CaCl2, 16.00 mg/L.

2.1.1. EPS extraction and analysis

The EPS of different membrane foulants in MBRs was extracted by the modified procedure [6,12]. Proteins were determined according to the coomassic brilliant blue method [13]. While polysaccharides were measured by phenol-sulfuric method using glucose as standard [14]. The concentration of proteins or polysaccharides was expressed as the mean.

EEM measurements of EPS extracted from membrane foulants was conducted using a PerkinElmer LS-50B luminescence spectrometer.

2.1.2. Membrane resistance analysis

Each membrane resistance (R: total hydraulic resistance; R_m: membrane resistance; R_f: membrane fouling resistance; R_P: pore blocking resistance; R_C: cake layer resistance), was prepared and estimatedin the end of experiment.

The analysis of membrane resistance was made according to Darcy law:

$$R = R_m + R_f = R_m + R_c + R_P$$

The experimental procedure on determine each resistance value was as follows. (1) R_m was measured by filtering deionized water using clean membrane module. (2) R was estimated by filtering deionized water using fouled membrane. Then $R_f = R - R_m$. (3) After flushing the surface of membrane to wipe off the cake layer, membrane used to measure the resistance of $R_p + R_m$ by filtering deionized water. And R_P was calculated then. (4) $R_C = R_f + R_P$ [6,15].

2.1.3. Other analytical methods

The particle size distribution was determined by a laser particle size analysis system (Malvern MasterSizer series 2000) within 0.02–2000 μm. The spatial structure of foulants on the membrane surface was observed by a scanning electron microscopy (SEM) system (FEIQUANTA 200).

3. Results and discussion

3.1. Operational performance and membrane fouling

Membrane fouling leads to a loss in permeability. Such a loss causes higher applied pressures. The membrane fouling profile of different MBRs are shown in Fig. 2. Obviously, MFSBRs of sidestream and submerged configuration exhibited a shorter operation time 11 and 14 days, respectively. They experienced the similar TMP variation, which quick jumped to over 50 kPa during 20–50 kPa. Similarly, Sánchez Sánchez et al. observed that the operation of MFSBR tended to be more unstable, showing a major tendency to achieve critical flux compared with MGSBR [16].

In contrast, the performance MGSBRs was much more outstanding. The SS-MGSBR run for 39 days. Significant TMP jump was not observed in its entire fouling process. Sub-MGSBR exhibited the greatest excellent operation of 61 days, and TMP rose up in a steadily

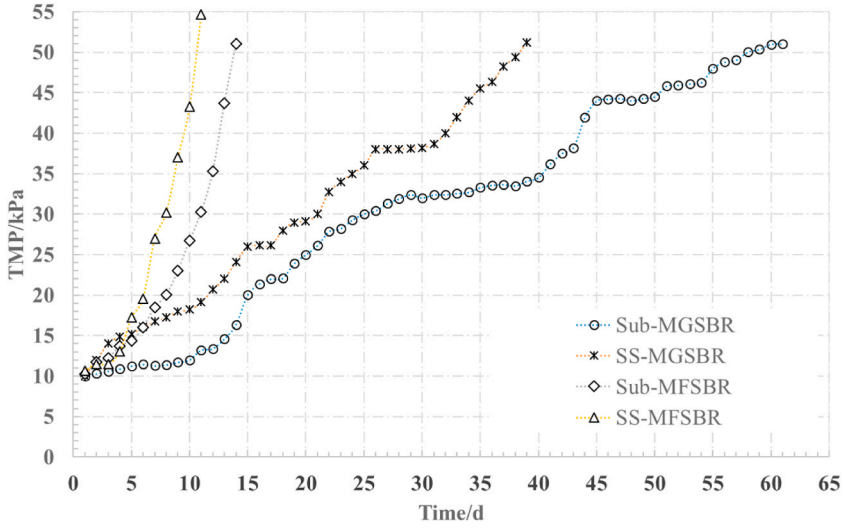

Fig. 2. Membrane fouling processes in MBRs.

overall rate during the whole operation; although slight TMP fluctuated at 15th and 45th day, and it might be concerned with the blocking of sludge, attached growth of microorganisms and cake layer development.

Obviously, these results easily indicated there was not obviously different ability of fouling control between Sub-MFSBR and SS-MFSBR; the aerobic granular sludge in MBR could effectively improve the membrane fouling development, and was really a useful way for slowing down the TMP increase rate or retarding membrane fouling. Another interesting point was that the configuration of submerged MBR had a superior performance to sidestream MBR, in either flocculent or granular sludge system. This result was also very useful to practical application.

3.1.1. Membrane fouling resistance analysis

Fig. 3 summarized the analytical measurements of different MBRs fouling resistances. Cake layer resistance R_C was the main reason for membrane fouling in flocculent sludge system. While the ratio of R_P/R_f was over 50% in aerobic granular sludge system, especially in Sub-MGSBR, which showed that the main factor of membrane fouling was pore blocking.

Because there were much less flocs in total sludge of submerged MGMBR, and the big granules were not easy to attach or clog on the surface of membrane module. Therefore, the pore blocking made a main contribution for membrane fouling in Sub-MGSBR. On the contrast, the mean size of the flocculent sludge was much smaller than that of aerobic granules; these small sludge particles were dominant in Sub-MFSBR (Fig. 4), and there was lack of abrasion of cake by big aerobic granules. Consequently, cake layer resistance was the key factor. For similar reason, the R_p of Sub-MGSBR was decreased compared with Sub-MGSBR, and was almost equal to R_c due to reduction of big sludge particles. In brief, the membrane fouling was not only relevant to the sludge morphology and structure, but also to the operational configuration.

Theoretically, macromolecules, colloids and particles onto and into the microporous membrane, the microbial attachment and biofilm formation are all relevant to the major limitation of filtration [9] In this experiment, cake layer formed fast, loose cake became dense along with TMP jumping at last [17], and TMP increased consequently in MFSBRs, cake layer resistance R_c became the key factor in MFSBRs.

However, aeration with big granules with little floc could bring strong shearing force [18,19] to slow down development of

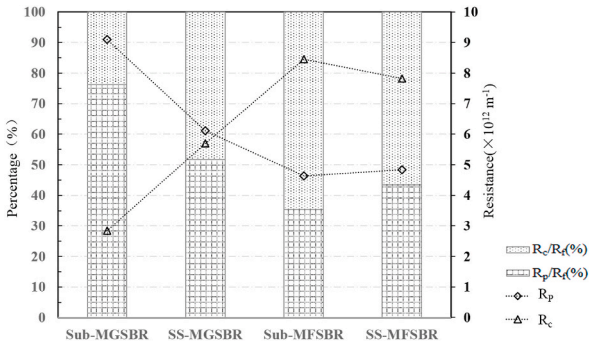

Fig. 3. Membrane fouling resistance analysis of different MBRs.

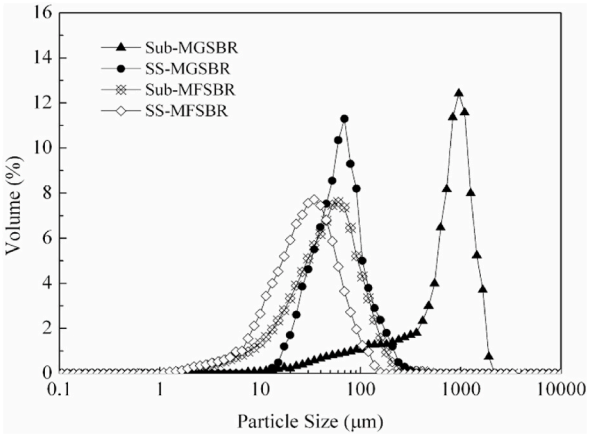

Fig. 4. Measurement of particle size distribution for sludge and supernatant to filter by different MBRs.

membrane foulants in Sub-MGSBR, and hardly induced serious cake by abrasion of mechanical cleaning with big granules [20]. Furthermore, although one-half effluent of bioreactor after settlement was through submerged membrane filtration, the other half that contained particles and flocs with small or loose structure was filtered by sidestream membrane module. In other words, the majority of sludge with better settleability still stayed in the bioreactor. Hence, the lack of hydrodynamics and consequently erosion phenomenon on the membrane fouling in SS-MGSBR led to higher R_C of contributed to 48.22% of R_f.

3.1.2. Observation of membrane foulants

Membrane fouling in its natural form is the coverage of the external or internal surface by foulants, which adsorb or simply accumulate during filtration. For the sake of understanding structure of foulants on the membrane surface, it was adopted SEM technology. SEM images of membrane samples of MBRs were showed in Fig. 5. At the end of experiments, however, rectangular pores were covered pollutants more or less. There was obvious biofouling but high porosity in membrane foulants of aerobic granular sludge system, especially in Sub-MGSBR, which was entangled with a large amount of rod-like bacteria and filamentous bacteria. The layer was with high porosity just like channels for transmission of nutrition and air in biofilm. Moreover, such structure might make a less impact on permeation than that layer in SS-MGSBR which was a little thicker and denser layer. In contrast, there was dense cake with low porosity and few microorganisms in Sub-MFSBR or SS-MFSBR.

To date, it has reported that biofilm is closely related to membrane fouling in flocculent sludge system, and membrane fouling can be controlled by employing quorum quenching which targets the growth of microorganisms [21–23]. Biofilm development starts from microbial attachment [24], which. Surface-attached cells responding to culture condition multiply in nutrition-rich medium. Once appropriate nutrients provided, biofilm grows. Nevertheless, biofilm would detach from adherent surface and then microorganisms become plankton as long as these are nutrient deprived [25]. Owing to difference in eutrophic condition of sequencing batch reactor or next tank in which membrane sidestream module immersed, there was huge difference about biofilm development between Sub-MBR and SS-MBR., biofilm on SS-MBR had difficulty in developing in thus environment deficient in nutrition. As a result, the surface of sidestream membrane modules, biofouling could not formed serious ultimately.

3.1.3. EPS characteristics of membrane foulants

The specific EPS amount in membrane foulants in MBRs showed great diversity as Fig. 6. In flocculent sludge system, the result suggested that polysaccharide was the predominant component of membrane foulants, for polysaccharide content increased up to 29.5 mg/gSS in Sub-MFSBR, 19.4 mg/gSS in SS-MFSBR, respectively. Much floc sludge deposited on the surface of membrane causing serious cake fouling, As a result, foulants in Sub-MFSBR had similar compositions and properties with the flocculent sludge. Since flocculent sludge owned high level of polysaccharide, there was high concentration of polysaccharide in these foulants, too. This current experimental data corresponded to the reported phenomenon that large-size polysaccharides was a typical kind of foulants [26].

It was protein that accumulated up to majority of EPS of aerobic granular as shown in Fig. 6b, however, there was still much polysaccharide of membrane foulants in MGSBRs. Basing on the SEM images of membrane samples (Fig. 5), the reason may be closely connected with the attached growth of microorganisms. The microbial attachment potential of aerobic granular sludge was stronger than that of flocculent activated sludge [27]. Biofilm develop more easily in MGSBRs consequently. Exopolysaccharide secreted by these microorganisms in the biofilm formed spatial and complex slime layers on membrane surfaces or pores [28]. In addition, the distribution of EPS between foulants in MGSBRs and aerobic granular sludge was so distinct that it indicated that aerobic granules did not play an important role in the amounts and properties of membrane foulants but microbial attachment and biofilm development were the key factors that affected membrane fouling.

The composition of EPS, extracted from the foulants at the end of performance of MBRs, was exhibited in Fig. 7. Three fluorescence peaks were named as peak A, B, and C, which were bound up with simple aromatic, tryptophan-like and humic-like organic

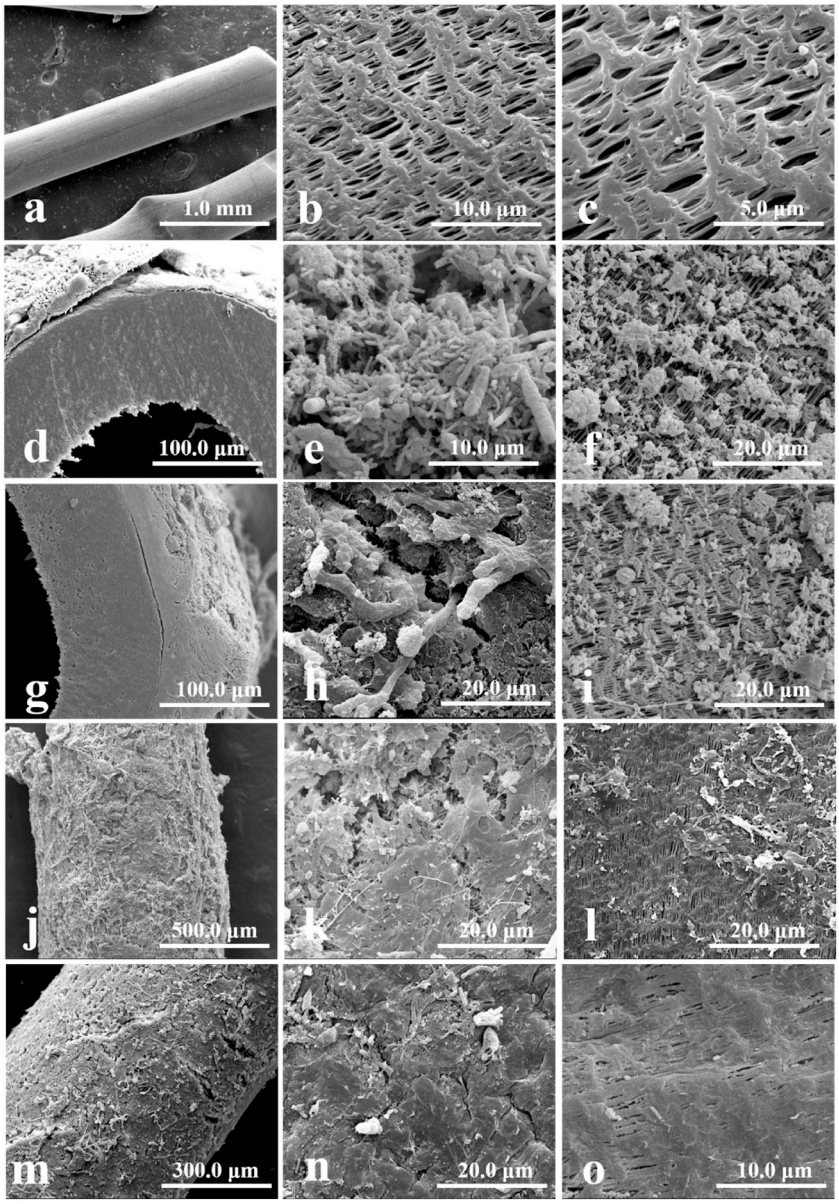

Fig. 5. SEM images of different membranes: (a, b, c): outer surface of original membrane; (d) cross section of fouled membrane in Sub-MGSBR; (e): outer surface of fouled membrane in Sub-MGSBR; (f): inner surface of fouled membrane in Sub-MGSBR; (g): cross section of fouled membrane in SS-MGSBR; (h): outer surface of fouled membrane in SS-MGSBR; (i): outer surface of fouled membrane in SS-MGSBR; (j, k, l): outer surface of fouled membrane in Sub-MFSBR; (m, n, o): outer surface of fouled membrane in SS-MFSBR.

compounds, respectively (Table 2). In aerobic granular sludge system, they were simple aromatic and tryptophan-like substance in membrane foulants. While membrane foulants included simple aromatic, tryptophan-like as well as humic-like organic substance in activated flocculent sludge system. In aerobic granular sludge, the majority of microogranisms assembled to form big and dense particles with outstanding settling property. However, much more and looser suspended solids had a tendency to deposit and turn into cake layer on the membrane surface in flocculent sludge. The different spatial structure of microorganism community resulted in the variation of EPS composition deposited on the membrane surface [29].

4. Conclusion

Compared with Sub and SS-MFSBR with similar quick increase of TMP, MGSBRs, especially Sub-MGSBR, performed much longer and more stable, which suggested that aerobic granules coupled with either membrane configuration could effectively retarding

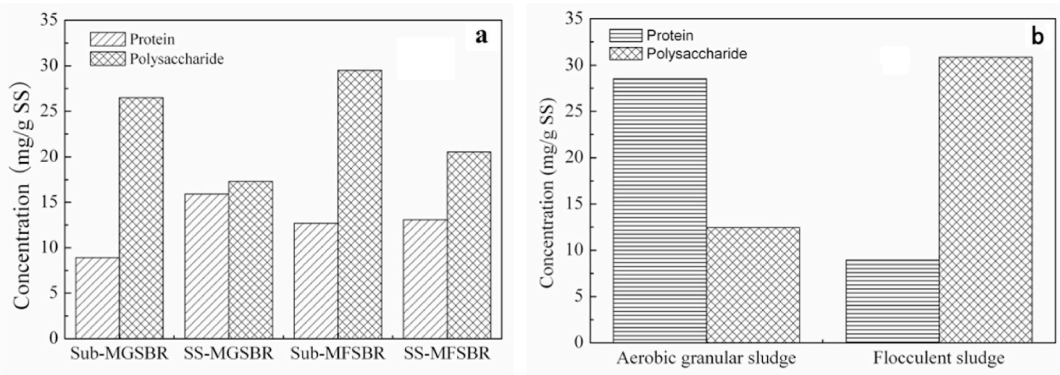

Fig. 6. Concentration of protein and polysaccharide (a: the EPS measurements of different membrane foulants; b: the EPS measurements of different sludges in submerged reactors).

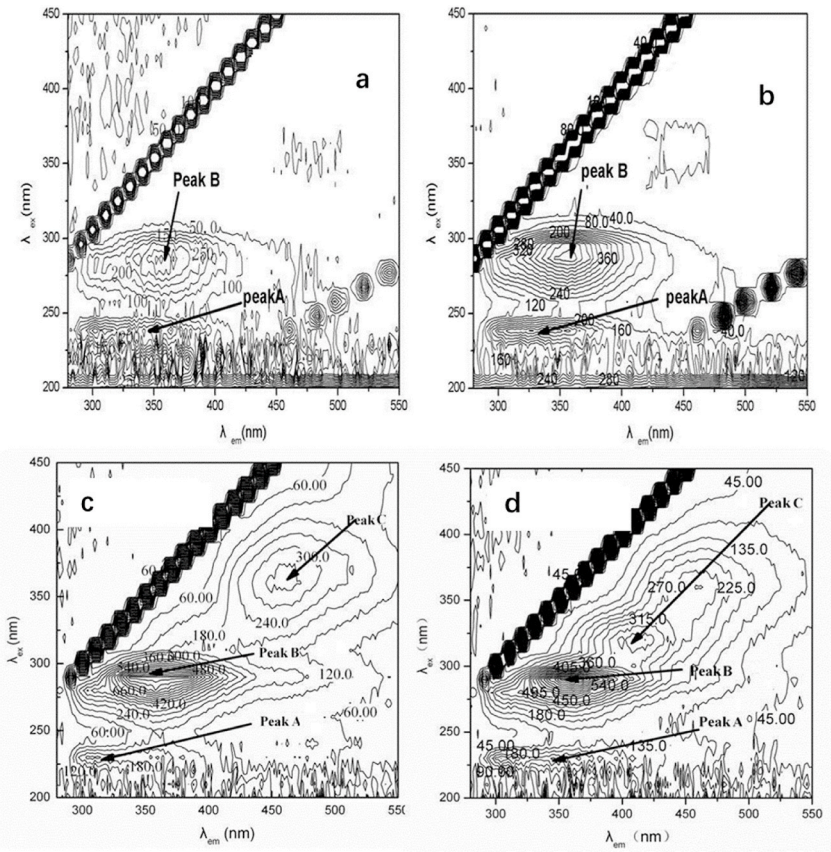

Fig. 7. EPS observations of membrane foulants in different sludge MBR systems (a: the EPS measurements of membrane foulants in Sub-MGSBR; b: the EPS measurements of membrane foulants in SS-MGSBR; c: the EPS measurements of membrane foulants in Sub-MFSBR; d: the EPS measurements of membrane foulants in SS-MFSBR).

membrane fouling. In flocculent sludge system, R_C was the key fouling factor, and membrane foulants were compact and with less biofouling. In aerobic granular sludge, however, R_P was the main fouling factor. Although there was much the most conglomeration of microorganisms of membrane surface, the fouling layer was with almost the highest porosity in Sub-MGSBR. Different membrane fouling behavior lead to different EPS composition of membrane foulants.

The coupling of aerobic granular sludge system and submerged membrane operation mode can play a very good role in reducing membrane pollution. Moreover, the biological membrane fouling in MGSBRs was serious. Therefore, it may be more effective to control membrane fouling by biological quenching, especially in Sub-MGSBR.

Table 2
Fluorescence Excitation-Emission spectrum parameters of foulants in MBRs.

MBRs	Peak A		Peak B		Peak C	
	Ex	Em	Ex	Em	Ex	Em
Sub-MGSBR	235	340	280	352		
SS-MGSBR	235	335	280	355		
Sub-MFSBR	230	330	280	355	355	450
SS-MFSBR	230	335	280	360	325	415

Author contribution statement

Yaqin Wang: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper.

Jianwei li: Performed the experiments.

Jianrong Zhu: Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data.

Data availability statement

Data included in article/supp. material/referenced in article.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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