A submerged tubular ceramic membrane bioreactor for high strength wastewater treatment

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Abstract A 4 L submerged tubular ceramic membrane bioreactor (MBR) was applied in laboratory scale to treat 2,400 mg-COD/L high strength wastewater. A prolonged sludge retention time (SRT) of 200 day, in contrast to the conventional SRT of 5 to 15 days, was explored in this study, aiming to reduce substantially the amount of disposed sludge. The MBR system was operated for a period of 142 days in four runs, differentiated by specific oxygen utilization rate (SOUR) and hydraulic retention time (HRT). It was found that the MBR system produced more than 99% of suspended solid reduction. Mixed liquor suspended solids (MLSS) was found to be adversely proportional to HRT, and in general higher than the value from a conventional wastewater treatment plant. A chemical oxygen demand (COD) removal efficiency was achieved as high as 98% in Run 1, when SOUR was in the range of 100–200 mg-O/g-MLVSS/hr. Unexpectedly, the COD removal efficiency in Run 2 to 4 was higher than 92%, on average, where higher HRT and abnormally low SOUR of 20–30 mg-O/g-MLVSS/hr prevailed. It was noted that the ceramic membrane presented a significant soluble nutrient rejection when the microbial metabolism of biological treatment broke down.

Keywords Membrane bioreactor; membrane rejection; SOUR; specific flux rate; submerged ceramic membrane

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Introduction

The activated sludge process is one of the most prevalent solutions for biological wastewater treatment (Behac *et al*., 1984). However, the efficiency of this conventional treatment is restricted by the difficulty of separating suspended solids from the effluent by gravitational settling (Benefield and Randall, 1980), which limits the biomass concentration to about 5 g/L, and consequently the required reactor volumes are large (Bailey *et al*., 1994). In addition, the process generates large quantities of excess sludge and its treatment and disposal represent 50% of total treatment cost (Defrance and Jaffrin, 1999).

The Membrane Bioreactor (MBR) is devised to overcome these constraints, in which the large-size settling tank is replaced by a membrane filtration unit permitting the extraction of a high quality effluent (Defrance *et al*., 2000). The advantage of MBR is having a higher biomass concentration, mixed liquor suspended solids (MLSS) between 5–20 g/l, which results in a higher rate of removal of chemical oxygen demand (COD) and a smaller excess sludge production. Moreover, the space occupied by the sewage plant is substantially reduced due to the absence of settling tanks and the reduction in bioreactor size made possible by the higher biomass concentration (Cote *et al*., 1998).

MBR was initiated with organic membranes, which are widely used compared to their ceramic counterparts. However, the severe fouling problem caused frequent replacement of organic membrane units due to their poor ability to regenerate. Thus, the application of a tubular ceramic membrane in a submerged MBR system is under investigation here, for its possession of robust resistance against extreme pH, pressure, and corrosive cleaning reagent (Cicek *et al*., 1999). The submerged MBR system, instead of the well established cross-flow MBR system, is considered in the present work, due to its superiority in terms of space reduction, energy conserving, and less initial capital and maintenance costs (Rui *et al.*, 2000; Stephenson *et al.*, 2000).

The present work focused on rheological and biological performance of this MBR system. The separation task is performed by a membrane unit, which broke the inherent relationship between sludge retention time (SRT) and hydraulic retention time (HRT) in conventional activated sludge processes, and it enabled the feasibility of longer SRT, which eventually leads to the minimum sludge discharge. This concept of prolonged SRT has been studied and proven within the operation period. The MBR system, as its name implies, might be sensitive to the biological treatment process. Hence, this issue was explored to study the effects of the potential debilitation in microbial metabolism.

Materials and methods

The submerged MBR system, as shown in Figure 1, consisted of a bioreactor and a tubular ceramic membrane unit. The ceramic membranes (USF Filtration, France) had an external Al_2O_3 layer at a pore size of 0.20 μm, with external diameter of 1 cm. An activated sludge reactor with working volume of 4.0 L was fed with the synthetic wastewater. Monitor sensors, which controlled the feeding speed via solenoid valve, maintained the constant water surface level in MBR and therefore HRT was solely dependent on effluent flowrate.

The feed used in this study simulated high strength wastewater containing high amounts of glucose and protein. It has been designed to provide all the inorganic and micronutrients, as well as nitrogen, phosphorus and carbon sources for the development of the biomass. The detailed composition of the synthetic wastewater is depicted in Table 1.

Sludge was taken from the Jurong wastewater treatment plant, Singapore, and assimilated for four weeks before the MBR operation. The experiment was performed in four runs according to HRT of MBR. In Run 1, HRT was kept at 1 day, and specific oxygen utilization rate (SOUR) was maintained at 100–200 mg-O/g-MLVSS/hr. From Run 2 to 4, HRT varied from 6 days to 3 days, and SOUR was designed to be abnormally low, at 20–30 mg-O/g-MLVSS/hr, which meant microbial metabolism activity was constrained to simulate a potential breakdown in biological treatment process. The SRT of MBR remained at 200 days throughout 142 days experiment time span. To supply sufficient oxygen for bacteria activities, dissolved oxygen (DO) was held at greater than 4 mg-O/L.

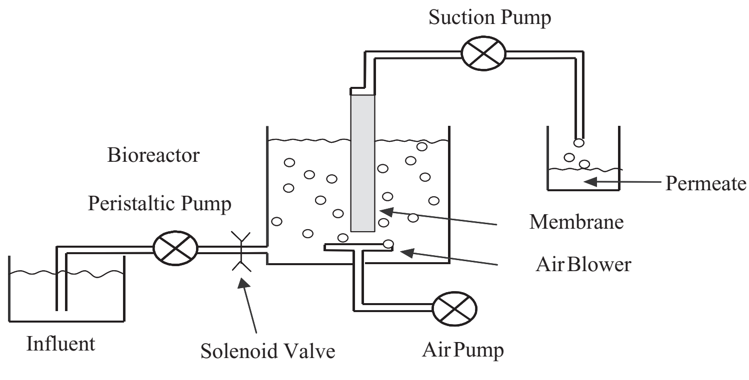

Figure 1 Schematic diagram of the submerged tubular ceramic membrane bioreactor

Table 1 Synthetic high strength feed composition

Components	COD	Glucose	Peptone	Beef extract	NH_4Cl	K_2HPO_4	$CaCl_2$-$2H_2O$	$MgSO_4$-$7H_2O$	$FeSO_4$-$7H_2O$
Concentration (mg/L)	2,400	1,400	800	250	200	45	30	25	20

When the suction pressure became higher than 30 kPa, the tubular ceramic membrane was backwashed. The temperature of the mixed liquor was maintained at 25°C within a temperature-controlled room. Operating conditions of the four runs are summarized in Table 2. The only difference in operation between Run 3 and Run 4 was that the former was the continuation from Run 2, while the latter started from newly assimilated activated sludge.

The organic compounds and particulate matter in the mixed liquor of the submerged MBR system were monitored on a daily basis during the start-up stage, and twice per week for the rest of experiment duration. However the flux rate was measured every day. The standard methods mentioned below refer to the *Standard Methods for Examination of Water and Wastewater* (1998): COD according to method 5220C; MLSS and mix liquor volatile suspend solids (MLVSS) following method 2540D and 2540E, respectively; SOUR according to method 2710B, sludge volume index (SVI) taken by method 2710D. While total organic carbon (TOC) was analyzed by a Shimadzu TOC analyzer, Model 5000, and size distribution was done by a Malvern Mastersizer, Model Microplus.

Results and discussion

Rheological performance

Figure 2 shows the size distributions of activated sludge from the aeration tank of the conventional plant with that of sludge from a submerged MBT after 80 days of operation. It is clear that the general size of sludge flocs from MBR decreased noticeably, compared to its counterpart from a conventional plant. The most frequent size dropped from 158 μm in the aeration tank to 48 μm after 80 days of operation in the submerged MBR. This was attributed to the fact that the aeration in the MBR system, which is meant to prevent fouling on the membrane surface, broke down the mean size of sludge flocs. Moreover, the smaller sludge particles are desirable, as they enhance mass transfer process, so that a higher organic removal rate and better oxygen utilization are achieved.

Figure 3 shows the filtration performance of submerged MBR for 142 days. As the average HRT remained constant during the duration of each run, the notorious fouling problem has caused the undesirable increase in trans-membrane pressure to keep a fixed effluent

Table 2 Average operating conditions of the membrane bioreactor

	Run 1	Run 2	Run 3	Run 4
Duration (day)	30	32	48	33
SRT (day)	200	200	200	200
HRT (day)	1	6	3	3
PH	7.4–7.7	7.4–8.2	.4–8.2	7.4–8.2
DO (mg-O/L)	> 4	> 4	> 4	> 4
Temperature (°C)	25	25	25	25
SOUR (mg-O/g-MLSS/hr)	100–200	20–30	20–30	20–30

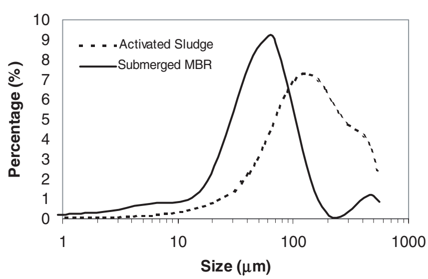

Figure 2 Size distribution of the sludge from a conventional plant and submerged MBR

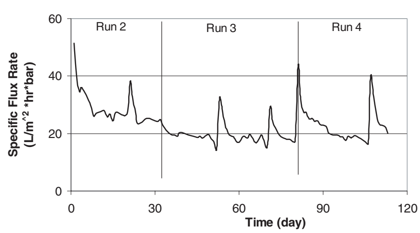

Figure 3 Filtration performance of submerged MBR

flowrate. This resulted in the continuous deterioration in filtration performance, in terms of specific flux rate (SFR). The peaks appearing during the experiment duration represented the events of backwash, which restored partially the SFR value, as illustrated in Figure 3. The SFR in Run 1 is generally greater than that of other runs, due to its shorter HRT and more frequent backwash. Run 3 had the lowest SFR, because it continued from Run 2 and therefore the accumulated fouling effect was more severe than that of other Runs.

It is observed that the SFR of this submerged MBR with a prolonged SRT of 200 days is about 15–40 L/m^2·hr·bar, which is lower than values reported by other researchers for ceramic membranes who used a much shorter SRT (Trouve *et al.*, 1994; Ahn *et al.*, 1998). Although SRT has no direct impact on SFR, a prolonged SRT might have side effects on the other characteristics of the MBR system, which eventually induced a lower SFR. Further studies should be conducted to investigate this issue.

The backwash techniques were applied whenever the suction pressure exceeded 30 kPa. A close look was taken at the filtration deterioration and the restoration effect of backwash. Shown in Figure 4, the SFR of a new membrane unit dropped tremendously upon placing in a bioreactor, which corresponded to the forming of inter-pore clogging and a biological gel layer adhering to the new membrane surface. After filtering for one day, the SFR became very stable, or in other words, decreasing at an unnoticeable rate, though SFR was far below the values obtained immediately after system start-up. This phenomenon might indicate that the biological gel layer has been completely formed and the forming of the second fouling layer – dynamic membrane is taking place. The existence of the dynamic membrane can be verified by the third curve in Figure 5, which was measured right after backwash. The air-jet backwash technique employed here removed the loose second fouling layer, thus restoring the SFR to the full extent on the second day. However, the third curve had a steeper slope than the second one, which was attributed to higher MLSS. Comparing to Figure 3, it can be noted that various backwash techniques would induce a different extent of regeneration of SFR. Therefore, a relevant experiment needs to be carried out in order to find the most suitable backwash technique that reflects the unique features of ceramic membranes.

Biological performance

The suspended solids were retained in the bioreactor by membrane unit, as the total suspended solid removal efficiency was as high as 99% in this study. Hence, the MLSS of MBR system, as shown in Figure 5, increased over the time, and was stabilized at the value, which depends on HRT. Theoretically, the bacteria community tended to accommodate the changing environment to maintain a constant F/M ratio (Stephenson *et al.*, 2000), and it was proven by Table 3. It is noted that both Run 3 and 4 were carried out under the same conditions and reached the same stabilized MLSS, but Run 3 took a longer time to stabilization. This is due to the fact that it continued from Run 2, and the bacteria had to assimilate the sudden drop in HRT from 6 days to 3 days. In general, shorter HRT resulted in higher

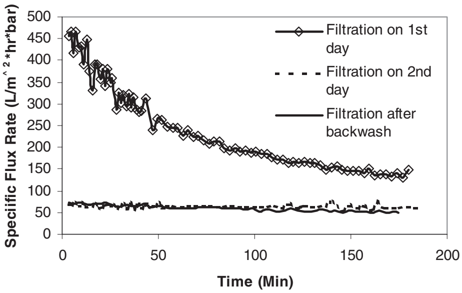

Figure 4 The deterioration of filtration performance

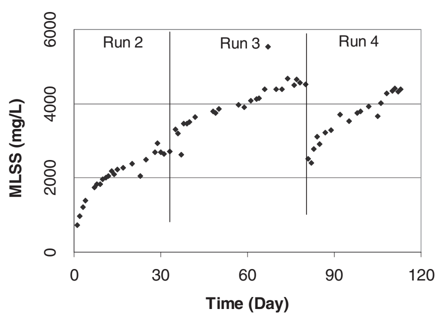

Figure 5 The MLSS of submerged MBR system

MLSS. A typical activated sludge plant treating municipal wastewater where SRT are in the range of 5 to 15 days, had an F/M ratio of 0.2 to 0.4 per day (Stephenson *et al.*, 2000). While Table 3 revealed a lower F/M ratio for the MBR system with an SRT of 200 days, which means less sludge yield. Therefore, the MBR system with a prolonged SRT has the greater market potential, since it discharged much less sludge.

The COD removal performance of the submerged MBR was listed in Table 4. Under the normal situation, such as Run 1, the COD removal efficiency was consistently higher than 98%, even for the high strength wastewater influent with COD = 2,400 mg/L. The COD of the effluent was always lower than 50 mg/L, the severe discharge standard in Europe. The SOUR plays a very important role in nutrient removal. The excellent performance in Run 1 was mainly attributed to its high biodegradation. Table 5 reveals in Run 1, 95.3% of the soluble organic carbonaceous nutrient, indicated by TOC, was removed by biodegradation, and only 2.6% of TOC removal was due to the presence of the membrane unit. However, the high biodegradation stemmed from the high MLSS, which was a result of membrane separation, an inherent advantage of the MBR system.

Lower nutrient removal efficiency was expected for Run 2 to Run 4, because of the lower MLSS and SOUR (Xia *et al.*, 2001). However, despite its abnormally low SOUR, the overall COD removal efficiency was still greater than 90%, for the synthesized high strength influent, although the effluent possessed a higher COD, compared to that of Run 1. Table 5 reveals the rationale for this high de-nutrient capability. The ill-performed microbial metabolism could only consume 35.7% of TOC, which resulted in an extremely high TOC supernatant in the bioreactor. This debilitation of bacteria biodegradation means a disaster for the conventional treatment plant, since the supernatant with high TOC of 607.6 mg/L will be discharged to the settling tank, where little TOC will be removed, and then to the environment, which imposes eutrophication on the receiving body. While this hazardous catastrophe did not happen for the submerged MBR system under study, this was due to the high membrane rejection of the ceramic membrane. After filtrating through the fouling layer, consisting of a biological gel layer and dynamic membrane, and the membrane itself, the TOC of the effluent was at 67.8 mg/L, about an 89% drop in TOC. In other words, 57.1% of the TOC of the influent was removed by membrane separation. Combining the percentage removals of TOC, the bioreactor coupled with the membrane unit could eventually remove more than 90% of TOC after the wastewater went through the submerged MBR system, even at the abnormally low SOUR of 20 to 30 mg-O/g-MLVSS/hr.

Analysis of biological performance under both the normal situation and abnormal conditions, low SOUR, has provided a good insight into the nutrient removal ability of the

Table 3 Biological condition of the submerged MBR

	Run 1	Run 2	Run 3	Run 4
Stabilized MLSS (mg/L)		2,400	4,600	4,400
HRT (day)	1	6	3	3
F:M ratio (/day)		0.17	–0.17	0.18

Table 4 Average COD removal performance of the submerged MBR

	Run 1	Run 2	Run 3	Run 4
Influent COD (mg/L)	2,400	2,400	2,400	2,400
Effluent COD (mg/L)	40	150	175	175
Removal efficiency %	98.33	93.75	92.71	92.71

Table 5 TOC removal performance in various stages

	Influent TOC (mg/L)	Reactor TOC (mg/L)	Percentage removal %	Effluent TOC (mg/L)	Percentage removal %	Membrane rejection %
Run 1	935.0	43.8	95.3	19.6	2.6	55.3
Run 4	944.9	607.6	35.7	67.8	57.1	88.8

submerged MBR system. Under the normal biological operating conditions, the microbial metabolism consumes most of soluble nutrient, and the membrane rejection slightly further polishes the quality of the effluent. The insoluble nutrients, together with other suspended solids, are retained in the bioreactor due to the membrane separation, and eventually are utilized to maintain a sustainable bacteria community. When there is a breakdown in biological treatment, symbolized by low SOUR in this case, the supernatant of the bioreactor contains an extremely high nutrient level, and the membrane, coupled with the fouling layer on its surface, possesses a crucially high membrane rejection rate, as high as 88.8%, which greatly alleviates the nutrient level of effluent. The membrane unit provides the second defense frontier for the environment, and plays a significant role in nutrient removal when the biological treatment process fails. This new credit brings another inherent advantage to the MBR system over the conventional activated sludge treatment plant.

Conclusion

On the laboratory scale, the tubular ceramic membrane unit, submerged in an aerated bioreactor, produced more than 92% COD and 99% suspended solids reduction throughout a time span of 142 days. Thus, this study proved the feasibility of the long-term application of a MBR system with a prolonged SRT of 200 days in high strength wastewater treatment. The introduction of a submerged membrane unit reduces considerably the space requirement for wastewater treatment, as it phases out the settling tank in the conventional activated sludge plant. The prolonged SRT provides a great promise for commercialization of this new MBR system, as it substantially reduces the amount of disposed sludge, and then the cost of sludge treatment. The ceramic membrane used in this study exhibited outstanding regeneration ability, and various backwash techniques have been employed successfully to restore the SFR.

The high MLSS was achieved in this study due to the membrane retention of suspended solids in the bioreactor. The high MLSS yielded excellent nutrient removal efficiency under the normal conditions. The stabilized MLSS was adversely proportional to HRT, as the bacteria community tried to regain the prevailing F/M ratio when subjected to changing rheological conditions. The SOUR had an influential effect on nutrient removal efficiency. Lower SOUR, corresponding to a failure in microbial metabolism, rendered lower COD reduction. However the ceramic membrane unit showed impressive nutrient rejection ability, up to 88.8% in terms of TOC removal, which, combined with the ailing biodegradation, resulted in a reasonably high COD removal efficiency of more than 90%. This new phenomenon, explored under abnormally low SOUR conditions, discovered another inherent advantage of MBR system over the conventional activated sludge plant: the membrane unit functions as the second protection line, when there is a breakdown in the biological treatment process.

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