

Dual Functions of an MF Membrane for Aeration and Filtration in a Sequencing Batch Membrane Reactor

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Introduction

Membrane bioreactors (MBRs), consisting of an aerobic reactor with suspended biomass and microfiltration membranes for liquid-solids separation, are finding many applications in water and wastewater treatment. MBRs have been used for treatment of municipal and industrial wastewaters and for reclamation of municipal wastewater for potential reuse in public water supplies. A MBR with powdered activated carbon (PAC) addition was applied for drinking water treatment to remove nitrate, natural organic matter and pesticides, and to disinfect the water. With respect to treatment efficiency and system stability MBRs have several advantages over conventional processes. However, the application of membranes to biological wastewater treatment is still limited due to issues of membrane fouling and high energy consumption. Backflushing with permeate or air, the addition of PAC to the reactor, and formation of a dynamic membrane, precoat or hydrophobic skin layers atop the membrane have been introduced to reduce fouling in crossflow MBRs, but these are still in an early stage of evaluation. Another alternative to a crossflow membrane operation was to use a submerged membrane with permeate removal by vacuum suction. The power consumption per unit volume of treated water was greatly reduced by eliminating the circulation pump, but the permeate flux was reduced to an impractical low level of below 2 L/m²-h. While the use of submerged membranes appears promising there is a need to develop methods that minimize flux decline from fouling.

The purpose of this study was to investigate the fouling control method and to observe the effect of the reactor operating dissolved oxygen (DO) concentration on nitrogen removal regarding potential reuse. Treatment performance, solids production, and permeate flux in a continuous long-term operation were determined for the sequencing batch membrane bioreactor (SBMBR) for different feeding conditions, aeration modes, and operating solids retention times (SRTs). Biological nitrogen removal was evaluated with nitrogen mass balances and during transient tests with different DO concentrations.

Materials and Methods

Seed Sludge and Synthetic Wastewater. Return activated sludge (RAS) taken from the Wastewater Treatment Plant was used as seed sludge for this study. The average composition of the RAS was suspended solids, 3,140 mg/L; volatile suspended solids, 2,560 mg/L; soluble COD, 693 mg/L; DOC, 214 mg/L; soluble NH₄-N, 214 mg/L; and soluble PO₄-P, 155 mg/L.

Sequencing Batch Membrane Bioreactor. The SBMBR studied consisted of a 4-L batch fed reactor with a dead-end microfilter submerged in the reactor bottom. Aeration was provided through either the membrane or the diffuser. Air pressure for the membrane aeration was 2.04 bar (30 psi). The reactor was pressurized at 0.68 bar (10 psi) to withdraw the treated water (permeate) through the membrane during the filtration step. The SBMBR system was operated at room temperature with a 4-h (6 cycles a day) cycle duration in sequence of 0.5-h Fill, 2.0-h Aeration, and 1.5-h Filtration/Idle times.

Microfiltration Membrane. A Polycap AS filtration capsule cartridge (Whatman, Arbor

Technologies, Inc., USA) consisting of a microfiber glass depth media (prefilter), a nylon final microfilter and polypropylene support was used for this study after modification. The polypropylene capsule housing was removed at one end to expose the membrane surfaces directly to the mixed liquor. The rating of the microfilter is $0.45\ \mu\text{m}$ with an effective filtration area of $800\ \text{cm}^2$ and a typical bubble point of 0.68 bar (10 psi).

Analytical Methods. Feed wastewater and treated effluent (permeate) samples normally obtained twice a week were analyzed for TOC, COD, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{PO}_4^{3-}\text{-P}$, and TKN. TOC was determined using an OI carbon analyzer (Model 700, OI Corp., USA). COD, $\text{NH}_4^+\text{-N}$, $\text{NO}_3^-\text{-N}$, $\text{PO}_4^{3-}\text{-P}$ were determined using the reactor COD digestion (with low range reagents), Nessler, cadmium reduction, and ascorbic acid methods from Hach, respectively. TKN concentrations were determined spectrophotometrically using the Hach Nessler method after digesting the sample with a mixture of sulfuric acid and hydrogen peroxide at 440°C . Dissolved oxygen (DO) concentrations were measured with a YSI DO meter (Model 58, USA) by continuously circulating the mixed liquor through an external tubing line connected to the reactor.

Results and Discussion

Treatment Performance. Table 1 summarizes the overall performance of the membrane bioreactor at different operating conditions. The MLVSS concentration increased steadily from 1,660 mg/L to 5,710 mg/L from period A through G, but in period H it dropped from 5,710 mg/L to 2,000 mg/L. A further decrease to 1,060 mg/L was observed when the system was operated with an 8-day SRT (period I). Depending on the feed change and biomass concentration, the F/M ratio ranged from $0.08 - 0.43\ \text{day}^{-1}$. The biomass change and solids yield will be discussed in detail in a subsequent section.

The lowest treatment efficiency for all the parameters was observed during period A, probably due to start-up acclimation of the microbial population to the synthetic wastewater. During periods B through I, the effluent concentrations of COD and TOC varied from 3 – 9 mg/L and 1.7 – 2.8 mg/L (corresponding to > 98% removal), respectively. The effluent $\text{NH}_4\text{-N}$ concentrations were always less than 0.1 mg/L, oxidized nitrogen ($\text{NO}_x\text{-N}$) was in the range of 3.2 – 5.6 mg/L, and organic nitrogen in the effluent was negligible in all cases, resulting in 87 – 93% nitrogen removal. The effluent turbidity was 0.17 ± 0.05 NTU. Changes in the feed composition and aeration mode had little effect on the effluent quality. However, the combined membrane/diffuser aeration would be a more economical application compared to the membrane-only aeration, because the diffuser can supply air at a lower pressure and thus lower energy requirement.

During period G, the effect of different air rates and DO concentrations on the system performance was evaluated. In Figure 1, the L1 aeration step DO concentration profile represents the normal system operating condition. A test for the other DO conditions was done no more than once per day. Conditions L5 and L6 represent aeration periods at low DO concentrations with limited aeration. Figure 1 shows two distinctive aeration regions in the DO profile, indicating the effect of switching from membrane aeration to diffuser aeration. The DO concentration was zero during the Fill period.

The effluent $\text{NO}_x\text{-N}$ ($\text{NO}_2\text{-N} + \text{NO}_3\text{-N}$) was similar for the higher DO concentration conditions L1, L2, and L3 (3.5-7.2 mg/L DO). With decreasing DO concentrations in conditions L4, L5, and L6 the amount of $\text{NO}_x\text{-N}$ removal increased by 1.0 mg/L in L4 and 3.0 mg/L in the extreme low DO condition L6. The total inorganic nitrogen (TIN, $\text{NH}_4\text{-N} + \text{NO}_x\text{-N}$) concentration similarly decreased. The decreasing $\text{NO}_x\text{-N}$ concentrations suggest that simultaneous nitrification and denitrification (SNdN) was occurring during the low DO aeration periods. Material balances on nitrogen were done to further evaluate the possibility

of SNdN in the SBMBR system during the normal reactor operation with the generally higher DO concentration (condition L1).

Simultaneous Nitrification and Denitrification. Nitrate removal during the mixed unaerated anoxic Fill period was measured, but a nitrogen mass balance was necessary to determine if nitrate reduction was occurring during the aeration period of the reactor cycle. SNdN would be indicated by a difference between the predicted $\text{NO}_x\text{-N}$ concentrations with time, based on $\text{NH}_4\text{-N}$ oxidation and nitrate reduction only during the unaerated fill period, and the measured $\text{NO}_x\text{-N}$ concentrations with cycle time. The mass balance assumed that the amount of nitrogen used for cell synthesis was negligible due to the immeasurable solids production during this period as shown later in this paper.

The depth of dissolved oxygen penetration within the biomass floc was estimated to investigate the potential for SNdN at different DO concentrations and SOURs in the reactor (Figure 2). The floc DO penetration depth depends on the bulk liquid DO concentration and SOUR. An observed SOUR was determined at the end of the React step from the slope of the DO profile, i.e., at the start of filtration. This provides information at the higher DO concentration near the end of the aeration period. To figure out the floc DO penetration at the lower DO concentration period when SNdN was expected, approximate React SOURs at the initial stage of the React step were estimated based on actual DO measurements during the 30-40 min membrane aeration period. The non-steady state SOUR is calculated by subtracting the rate of change of mixed liquor DO concentration from the oxygen transfer rate (OTR), as shown in equation (1) and (2). The rate of mixed liquor DO change can be obtained from the slope of the DO profile during this period.

$$SOUR_{react} = \frac{1}{MLVSS} \left(OTR - \frac{dC_L}{dt} \right) \quad (1)$$

$$OTR = K_L a (C_e - C_L) \quad (2)$$

Where

$\frac{dC_L}{dt}$ = rate of change of mixed liquor DO, mg/L-h;

$K_L a$ = overall liquid film coefficient, 1/h;

C_e = equilibrium mixed liquid DO concentration, mg/L;

C_L = mixed liquid DO concentration at time, mg/L;

Figure 2 shows that during the high DO aeration period (DO approximately 7.2 mg/L) the floc was completely aerobic and no denitrification or SNdN should have occurred. However, the floc aerobic depth estimate for the 30 – 40 min membrane aeration period was less than 53 μm or about 75% of the floc radius, which supports the assumption that a SNdN was occurring to account for nitrogen losses during aeration. Also, further SNdN could be expected to take place even over a longer period after 40 min when the aeration regime was switched from membrane aeration to diffuser aeration, since the DO was sharply decreasing for another 7 min of aeration.

Single cycle mass balances on nitrogen at different DO levels are summarized in Figure 3. The percent of total nitrogen lost in the React step (SNdN) increased from 27% to 47% with lowering DO levels from condition L1 to L6, while the percent of total nitrogen that remained in the reactor correspondingly decreased from 42% to 20%. The percent NO_x lost in the Fill step was $29 \pm 5.5\%$ and the estimated percent N in cell synthesis was about 1.9%. The percent $\text{NH}_4\text{-N}$ remaining in the reactor was very low although the fraction increased slightly to about

2.6% at the DO level L6. Overall, substantial denitrification of 56 – 75% was achieved in the batch membrane reactor. Therefore, operation of the SBMBR at lower DO concentrations helps to improve the nitrogen removal efficiency as long as nitrification is completed during aeration.

Biomass Accumulation and Yield. From 50 to 250 days with no significant intentional solids wasting, the MLVSS concentration steadily increased. During period F, the MLVSS concentration sharply increased due to the feeding of Renton RAS. For the operation with no intentional solids wasting (calculated 1,400-day SRT, period A through H), the biomass yield, accounting for sampling and inventory changes averaged only 0.03 mg VSS/mg COD. But at the 8-day SRT (period I) the yield increased to a more typical value of 0.3 mgVSS/mg COD. Thus the operation of the membrane reactor at the long SRT made it possible to substantially reduce the sludge production because of the long time available for cell decay. Membrane filtration played a critical role in achieving the long SRT, since the membrane could separate the effluent liquid (treated effluent to be discharged) from the mixed liquor completely regardless of the settling characteristics of the biosolids.

Membrane Permeability. When the membrane was operated with a new one, the flux was about 200 L/m²-h-bar in the first cycle and then declined continuously for the next 30 days. During the rest of the operational period, however, the flux was relatively stable at 34 ± 4.4 L/m²-h-bar, regardless of operating conditions. The stabilized specific flux of about 34 L/m²-h-bar (actual flux, 24 L/m²-h) in this continuous operation was much higher than the flux of 2 - 18 L/m²-h achieved by other researchers choosing the submerged-type filtration and comparable to the normal flux ranging from 5 – 40 L/m²-h-bar in crossflow filtration. The improved flux in this system probably resulted from the use of the membrane for aeration generating an air backwashing effect that periodically released the biomass cake layer that accumulated at the membrane surface. Compared to crossflow filtration, this batch filtration with combined membrane/diffuser aeration also provides the benefit of eliminating the need for a circulation pump to maintain a high tangential velocity.

The flux is plotted against the biomass concentration in the reactor in Figure 4 for the period after the initial stage of continuous flux decline, i.e., after 100 days of operation. The flux remained relatively stable although the solids concentration in the reactor varied from 710 mg/L to 10,000 mg/L as MLSS. The absence of any correlation between flux and biomass concentration is contradictory to the findings of other investigators for crossflow filtration. The results of the current tests suggest that the (reversible) filtration resistance contributed by the biomass cakes represent a relatively small fraction of the total resistance. Presumably, the presence of the bulky filamentous organisms in the reactor caused the biomass cake layer to be more loosely packed during filtration.

Conclusions

A sequencing batch reactor using a microfiltration membrane for effluent filtration with air sparging through the membrane for fouling control was operated for over 1 year to investigate treatment efficiency and long-term membrane permeability at different operating conditions. The following conclusions could be drawn.

During the whole period of operation, effluent turbidity was below 0.2 NTU and substantial removal of COD and nitrogen was achieved. A significant amount of nitrogen removal (47%) occurred by simultaneous nitrification – denitrification (SNdN) when the DO concentration was maintained below 0.6 mg/L, but over a longer react time.

At a long operating SRT with no significant intentional biomass wasting the biomass yield was very low at 0.03 mg MLVSS/mg COD. A bulky biomass developed in the reactor but it had no consequence on treatment performance since membrane filtration was used for liquid-

solids separation.

After an initial irreversible flux decline, a stable flux was maintained at about 34 L/m²-h-bar irrespective of the biomass concentration. The periodic air backwashing method used to control membrane fouling prevented continuous progression of biofouling.

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Table 1. Summary of operational conditions and effluent concentrations.
(No intentional solids wasting for period A - H, 8-d SRT for period I)

Time Period, days	Feed Change	Aeration Mode ^{a)}	F/M Ratio, day ⁻¹	COD, mg/L	TOC, mg/L	NH ₄ -N, mg/L	NO _x -N, mg/L	MLVSS, mg/L
0 - 63 (A)	No	M	0.27	15.1 (16.4)	4.7 (3.4)	2.95 (7.76)	9.4 (6.5)	1660 (1160)
63 - 102 (B)	+ Glucose ^{b)}	M	0.29	2.6 (3.0)	2.3 (0.54)	0.09 (0.05)	4.4 (1.1)	2310 (981)
102 - 144 (C)	+ Glucose	M/D	0.16	8.5 (2.6)	2.3 (0.36)	0.04 (0.02)	3.4 (0.4)	4130 (131)
144 - 165 (D)	+ Glucose	M	0.13	5.0 (1.8)	2.8 (0.22)	0.05 (0.02)	4.8 (0.9)	4910 (149)
165 - 176 (E)	+ Glucose	M/D	0.13	4.2 (0.6)	2.6 (0.18)	0.06 (0.02)	4.8 (0.0)	4920 (190)
176 - 204 (F)	+ RAS ^{c)}	M/D	0.08	5.8 (3.2)	2.7 (0.30)	0.03 (0.01)	5.5 (1.1)	5680 (959)
204 - 295 (G)	No	M/D	0.08	5.2 (3.5)	2.1 (0.19)	0.03 (0.02)	5.6 (1.1)	5710 (982)
295 - 309 (H)	- Peptone +Glucose ^{d)}	M/D	0.23	5.8 (0.3)	1.7 (0.24)	0.02 (0.02)	3.2 (0.8)	2000 (1390)
309 - 348 (I) ^{e)}	No	M/D	0.43	3.9 (1.9)	2.0 (0.16)	0.03 (0.02)	4.5 (0.4)	1060 (412)

^{a)} M, membrane only aeration for 120 min; M/D, combined aeration provided by the membrane for 10 min followed by the diffuser for 110 min.

^{b)} 131.3 mg/L of glucose was added to the basic feed

^{c)} Return activated sludge was injected to the reactor at a rate of 60 mL/day.

^{d)} Bacto peptone was subtracted from the basic feed but glucose was added to keep the same COD level.

^{e)} The only period of time when biomass wastage occurred at 8-d SRT.

Values in parentheses indicate standard deviation.

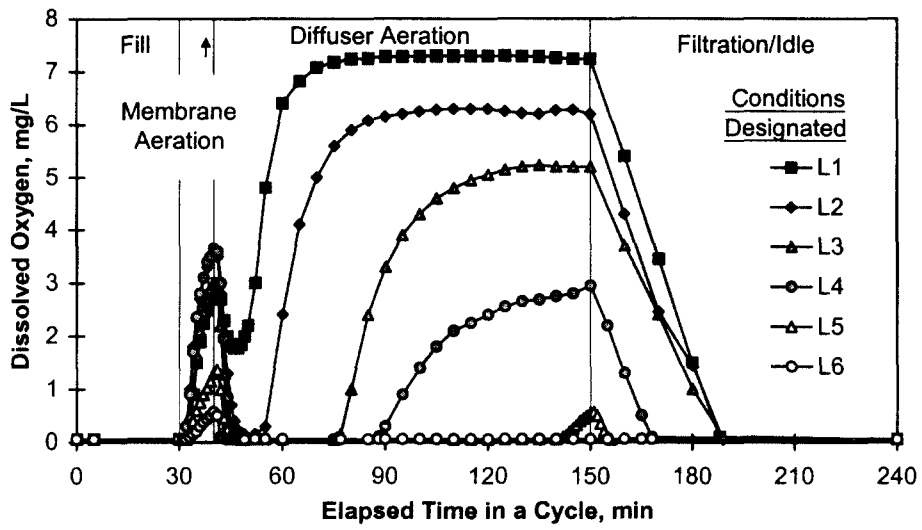


Figure 1. DO concentration variations with different air supply rates during a cycle.

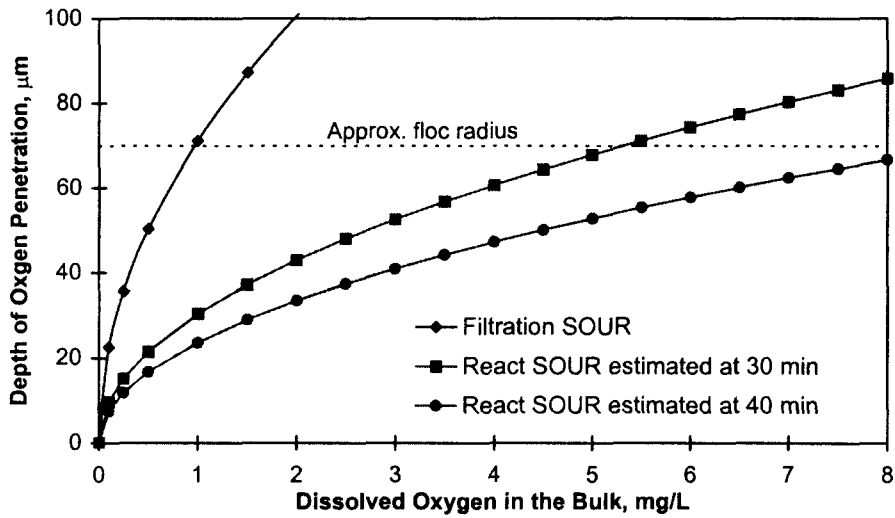


Figure 2. Calculated oxygen penetration depth versus bulk liquid DO concentration at the beginning and end aeration period.

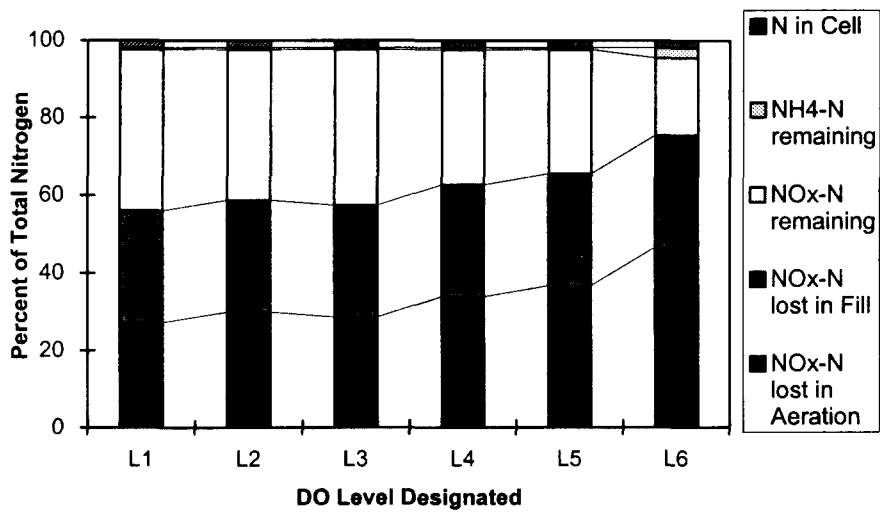


Figure 3. Single cycle nitrogen mass balance at different operating DO concentrations (designated in Figure 1).

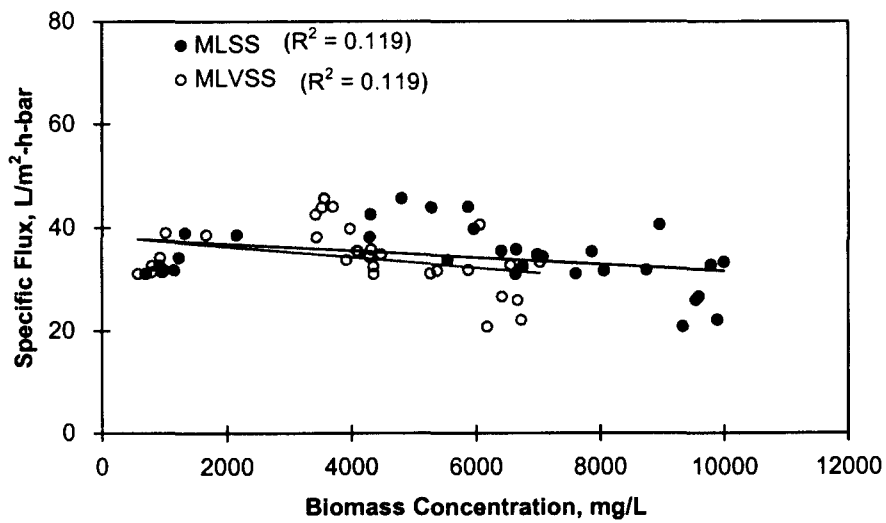


Figure 4. Specific flux versus biomass concentration.