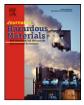


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Optimizing aeration rates for minimizing membrane fouling and its effect on sludge characteristics in a moving bed membrane bioreactor

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1. Introduction

Membrane bioreactors (MBRs) have been used as an innovative and promising option for wastewater treatment and reuse. Membrane bioreactor technology encourages wastewater reuse and improves water sustainability. This technology is simple to operate, needs modest technical support, takes up little space and can remove many contaminants from wastewater in one step [1].

Biofilm reactors have been successfully used in water and wastewater treatment for over a century. Systems using biofilm processes have the following advantages: (i) less complex operation compared to that of activated sludge systems, (ii) the ability to increase biological reaction rates through the accumulation of high concentrations of active biomass and (iii) the high resistance of this attached biomass to overloading and toxic compounds. Moreover,

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ABSTRACT

In MBR processes, sufficient aeration is necessary to maintain sustainable flux and to retard membrane fouling. Membrane permeability, sludge characteristics, nutrient removal and biomass growth at various air flow rates in the membrane and moving bed biofilm reactor (MBBR) compartments were studied in a pilot plant. The highest nitrogen and phosphorous removal rates were found at MBBR aeration rates of 151 and 85 L h⁻¹ and a specific aeration demand per membrane area (SAD_m) of 1.2 and 0.4 $m_{3.1}^2$ m⁻² h⁻¹, respectively. A linear correlation was found between the amount of attached biofilm and the nutrient removal rate. The aeration rate in the MBBR compartment and SAD_m significantly influenced the sludge characteristics and membrane permeability. The optimum combination of the aeration rate in the MBBR compartment and SAD_m were $151 L h^{-1}$ and $0.8-1.2 m_{air}^3 m_{membrane}^{-2} h^{-1}$, respectively.

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a biofilm makes the maintenance of high biomass age possible, which favors the selective development of specific slowly growing bacteria, such as nitrifiers, as it reduces their washout from the system [2]. One of the most prevalent biofilm-based processes involves the moving bed bioreactor (MBBR). It generally requires an upgrade of industrial and municipal wastewater treatment facilities to increase organic loading and simultaneous nutrient removal [3]. Even still, it has been reported that settling characteristics of MBBR sludge were less efficient than that of conventional activated sludge [2].

Although MBRs offer effective separation of pollutants and tolerance to high or shock loadings, MBR technology is currently facing some research and developmental challenges such as membrane fouling, high membrane cost, and the need for pretreatment. Membrane fouling, which increases operational cost and shortens the life of the membrane, is the most difficult challenge. To overcome the membrane-fouling problem, various studies have been conducted to understand and minimize membrane clogging. These efforts include the use of intermittent instead of continuous suction, the addition of alum and natural zeolite, association with SMBR and powdered activated carbon (PAC), and the use of modified cationic polymers [1].

Mixed liquor suspended solid (MLSS) concentration is one of the most important factors that affect membrane fouling. An alternative to the MBR is the use of a combination of a biofilm reactor with membrane separation of the suspended solids (BF-MBR), which

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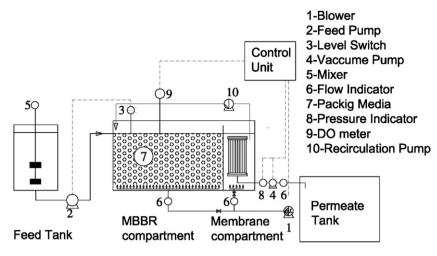


Fig. 1. Flow diagram of experimental setup.

may reduce the effect of membrane fouling by high biomass concentrations [4].

Aeration is another important parameter in the design and operation of an MBR. Aeration is required for biotreatment, floc agitation and membrane scouring. It is the most costly factor in terms of MBR energy consumption. Air scouring is necessary in submerged membrane systems to generate localized cross-flow conditions along the membrane surface, which reduces cake deposit on the membrane. Nevertheless, the relationship between aeration and flux or trans-membrane pressure (TMP) decline is still not fully understood. Aeration rates in MBR systems are based on previous data and normally recommended by the membrane supplier [5,6]. Aeration comprises almost 50% of the total energy requirements of MBR operation [7]; therefore, it is very important to optimize aeration rate in MBR processes.

Studies investigating membrane fouling in MBR processes have reported the significance of aeration rate in the reduction and promotion of membrane fouling. Ivanovic and Leiknes evaluated the impact of aeration rates on particle colloidal fractions in the biofilm MBR. In this study, a desirable range for SAD_m was estimated, for the given membrane reactor design and operating conditions, to be higher than 1.68 but lower than 3.37 m³ m⁻² h⁻¹[5]. Research has also shown that the floc size decreases with an increase in the velocity gradient (G), and the extracellular polymeric substance (EPS) concentration increases at high shear tension in submerged MBRs [8,9]. High amounts of floc-bound EPS (bEPS) and soluble EPS (SMP) have a negative impact on sludge properties such as the filamentous index (FI), settling behavior and the ability to dewater [10]. The bulking sludge has a higher bEPS concentration, whereas the deflocculated sludge has a higher free EPS concentration; an increase in the free EPS concentration can do great harm to MBRs [11].

Results of a study on three parallel MBRs operated under different aeration intensities (150, 400 and $800 L h^{-1}$) showed that either a small or a large aeration intensity had a negative influence on membrane permeability. The large aeration intensity resulted in a severe breakup of sludge flocs and promoted the release of colloids and solutes from the microbial flocs to the bulk solution. The sludge supernatant would become heterogeneous as the aeration intensity increased [12].

All of the aforementioned studies insist on the importance of membrane aeration rates on foulant concentrations and membrane fouling in MBR processes. To date, there are no reports found on the effects of aeration rate in the MBBR compartment on membrane fouling.

The advantages of moving bed membrane bioreactor (MB–MBR) process in comparison with conventional MBR include: (a) less

sludge production rate due to high biomass retention time, (b) simultaneous nitrification-denitrification and phosphorous removal due to oxygen gradient in biomass layer on packing media, (c) more durable to toxic and organic shocks, (d) higher volumetric loading rate, and (e) less suspended solids concentration that results less membrane fouling [4,13–15]. In this study, the aeration rate in MBBR and membrane compartments in the moving bed membrane bioreactor (MB–MBR) was studied, and the optimum aeration rate in both compartments was determined separately. The objective of this study was to investigate the aeration rate on nutrient removal, FI, SMP and EPS concentration and finally membrane permeability in a pilot plant operating with a MB–MBR process.

2. Materials and methods

2.1. Experimental equipment

As shown in Fig. 1, the experimental equipment was made of Plexiglas with a length, width and depth of 50, 23, and 40 cm, respectively. The total volume of the system was 46 L, of which the effective volume was 35 L; the effective volume consisted of the biofilm and membrane zones, the volumes of which were 25 and 10L, respectively. A hollow fiber membrane module (polyvinylidene fluoride, hydrophilic, pore size 0.1 µm, effective surface area 0.2 m², Nanofilm, Australia) was installed in the membrane compartment that was separated from the MBBR compartment by a mesh Plexiglas sheet. In the MBBR, the biomass grew on carriers that moved freely in the water volume by aeration. The biofilm carriers were made of poly propylene (PP) and shaped as small cylinders, which had two crosses on the inside of the cylinder and "fins" on the outside. The size of each piece of the packing media was 10mm in diameter and 7mm in height. The packing media (supplied by JESCO Co., Iran) was used in the pilot plant reactors at 70% filling-fraction, giving an effective specific surface area of 350 m² m⁻³. Synthetic wastewater was fed into the reactor with a pump, and its flow was controlled with an electrode level switch. The permeate suction was done using a peristaltic pump (Prominent Dose, Germany) to remove permeate continuously from the MBR. The air diffusers, controlled by 2 air flow meters (AGA, Hitchin Herts, UK), were installed directly at the bottom of the membrane module to reduce membrane fouling and to supply oxygen to the microorganisms. Air scouring of the membrane was applied continuously but with varying flow rates. For minimizing concentration polarization in the membrane compartment, a recirculation pump was installed between the MBBR and membrane compartments.

Table 1

Constituents of the synthetic wastewater used in this study.

Compounds	Concentration range (mg L ⁻¹)			
Organics and nutrients				
Sodium acetate (NaCOOH)	180-200			
Glucose ($C_6H_{12}O_6$)	180-200			
Sucrose	180-200			
Starch	180-200			
Milk powder	180-200			
Ammonium sulfate ((NH ₄) ₃ SO ₄)	400-450			
Potassium phosphate (KH ₂ PO ₄)	140-150			
Trace nutrients				
Calcium chloride (CaCl ₂ ·2H ₂ O)	0.37			
Magnesium sulfate (MgSO ₄ ·7H ₂ O)	5			
Manganese chloride (MnCl ₂ ·4H ₂ O)	0.28			
Zinc sulfate (ZnSO ₄ ·7H ₂ O)	0.45			
Ferric chloride anhydrous (FeCl ₃)	1.45			
Cupric sulfate (CuSO ₄ ·5H ₂ O)	0.4			
Cobalt chloride (CoCl ₂ ·6H ₂ O)	0.4			
Sodium molybdate dihydrate (Na2MoO4·2H2O)	1.25			

For starting up the process, the reactor was seeded with activated sludge from a local wastewater treatment plant (West Township in Tehran-Iran) and acclimatized to synthetic wastewater. The filtration was made with a 9.5 min permeation and 0.5 min relaxation cycle [16].

The experiments were conducted using a synthetic wastewater to avoid any fluctuation in the feed concentration and provide a continuous source of biodegradable organic pollutants. The constituents of the synthetic wastewater are given in Table 1. The pH value of the MBR was controlled at 7 ± 0.5 by adding soda ash to the reactor's influent; NaHCO₃ was used as a pH buffer and carbon source for nitrifying bacteria [17]. The reactors were operated at 20 ± 2 °C using two thermostatic heaters.

During the entire experimental period, the reactor was operated at a steady-state operational condition with an SRT of about 35 days, wasting 1 L mixed liquor daily, and with an organic loading rate of 1.12-1.26 kg COD m⁻³ d⁻¹.

The process performance and extent of sludge filtration of the MB–MBR process was investigated with 20 different combinations of aeration rates in the MBBR and membrane compartment including the following: five different aeration rates of 42, 85, 151, 296 and 380 Lh^{-1} in the MBBR compartment and four different aeration rates of 0.4, 0.8, 1.2 and 1.6 m³ m⁻² h⁻¹ expressed as SAD_m in the membrane compartment [6].

The specific aeration demand per membrane area (SAD_m) is calculated using the following equation:

$$SAD_{\rm m} = \frac{Q_{\rm air}}{A_{\rm m}} \tag{1}$$

where Q_{air} is the air flow rate (m³ h⁻¹) and A_m is the membrane surface area (m²).

2.2. Analysis

The presented results were all obtained from the MB–MBR system at steady state. For evaluating the process performance, the samples were collected from one entire cycle, and the mixed liquor was used to assay the sludge characteristics and its filterability. The samples taken from bioreactors were filtered using fine pore filter paper with a pore size of 125 mm (CHMLAB GROUP, Spain). The dissolved oxygen concentration (DO) was measured using a DO meter (MI-65 Martini Instruments), and the pH value was measured using a pH meter (HACH-Germany). The measurement of COD, MLSS, total nitrogen (TN), oxidized nitrogen (NO₃⁻–N and NO₂⁻–N), ammonium nitrogen (NH₄⁺–N), orthophosphate concentration (ortho-P) and total phosphorous (TP) was carried out using a

Table 2	
Measured quality of the feed wastewater and permeate.	

Parameter	Mean	Maximum	Minimum	Standard deviation
Influent				
COD	943.57	980.00	880.00	34.71
TN	75.71	86.00	68.00	6.07
TP	24.21	31.00	19.00	4.51
$OLR (g COD L^{-1} d^{-1})$	1.21	1.26	1.13	
Permeate				
COD	19.07	32.00	10.00	4.86
TSS	-	-	-	-
TN-N	6.07	14.00	4.00	2.84
TP	7.36	18.00	3.00	4.38
NH ₄ -N	0.62	0.90	0.40	0.13
NO ₃ -N	3.06	9.20	1.20	2.25

spectrophotometer (Dr-5000, HACH-Germany), and the MLSS content was analyzed according to standard methods [18].

The extraction of bound EPS was based on the following heat extraction method [19,20]: 1 ml sludge suspension was taken and centrifuged at 3500 rpm for 10 min. Free EPS (or SMP) was obtained in the supernatant through a membrane filter. The sludge pellets were resuspended to their original volume using deionized water. The sludge was then heated at 90 °C for 10 min, and the suspension was centrifuged again at 10,000 RPM for 10 min. The supernatant was centrifuged twice for 15 min at $12,000 \times g$ to remove the remaining floc components. Bound EPS was obtained by filtering the supernatant through a membrane filter. The bEPS and SMP were normalized as the sum of hydrocarbon and protein, which were analyzed using the phenol/sulfuric acid method and Bradford method, respectively [11,21].

The amounts of biomass fixed on the carriers were determined as follows: 50 ml of packing media (about 26 pieces) was taken out of the bioreactor and put in a beaker with 500 ml deionized water in it. Next, the carriers were stirred with a magnetic stirrer for 60 min, and the fixed biomass was washed out from carriers. The suspension was then dried and weighed to calculate the concentration of the biofilm in the MBBR [13].

The filamentous bacteria density was quantified as a filament index using a previously documented method [22]. According to this method, the number of filamentous organisms was rated on a scale of 0–6, where 0–6 corresponded to none, few, some, common, very common, abundant and excessive presence of filamentous organisms, respectively [23].

3. Results and discussion

3.1. Effects of aeration rate on process performance

Different aeration rates, in the presence of sufficient dissolved oxygen, did not affect the organic removal efficiency due to the fact that a stable permeate quality was observed during the entire experimental period. Nevertheless, because the aeration rate affects biofilm thickness and stability on packing media, the aeration intensity has an effect on biological nutrient removal in biofilm-based processes. The measured quality parameters for the wastewater feed and permeate from the membrane reactor are summarized in Table 2.

Fig. 2a and b shows the nutrients in the MBBR permeate versus aeration rates. It shows that the aeration rate in the MBBR compartment is more effective than SAD_m on nitrogen and phosphorous removal. The highest nitrogen removal occurred at an MBBR aeration rate of $151-296 L h^{-1}$ and a SAD_m of $1.2 m^3 m^{-2} h^{-1}$. The values for phosphorous removal were $85-151 L h^{-1}$ and $0.4-1.2 m^3 m^{-2} h^{-1}$, respectively.

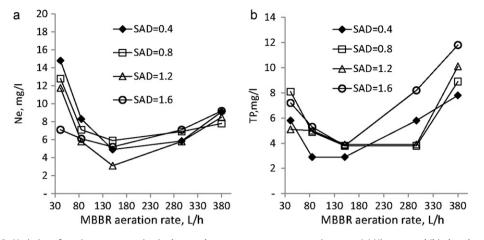


Fig. 2. Variation of nutrient concentration in the membrane permeate versus aeration rates. (a) Nitrogen and (b) phosphorous.

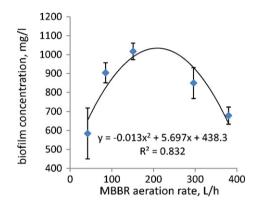


Fig. 3. Attached biofilm versus aeration rate.

As shown in Fig. 3, there is a parabolic correlation between the amount of biofilm on the packing media and aeration rate ($R^2 = 0.83$). Since the reactor was operated without any anoxic and anaerobic zones, it is suggested that the main effective parameter on N and P removal is dissolved oxygen gradient in the biofilm layer. There is a linear correlation between the removal of phosphorous and nitrogen and the amount of attached biomass on biomass carriers ($R^2 = 0.76$ and $\alpha < 0.01$ for effluent phosphorous and $R^2 = 0.93$ and $\alpha < 0.01$ for effluent nitrogen). This can be related to the anaerobic conditions that occur in the inner biomass layers and luxury uptake in aerobic conditions; in addition, nitrogen was removed via SND in the mixed liquor and attached biofilm of the reactor (Table 3).

In very low aeration rates ($<85 L h^{-1}$), DO in reactor decreases that leads to increase in ratio of sloughing on biofilm growth and the mass of biofilm (thickness of biofilm layer) decreases that leads to decrease the removal efficiency. Also in very low DO ($<2 m L^{-1}$)

Table 3

The correlations among sludge	and permeate characteristics
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nitrification does not occur completely and concentration of ammonia in permeate increases. In higher aeration rate (specially more than 295 L h⁻¹) the turbulent increases and more collisions occurs between moving bed packing media that results the sloughing of biofilm layer specially in outer side of the media parts. As shown in Figs. 2 and 3 decrease in amount of biofilm results increase the concentration of N and P in membrane permeate and vice versa. High turbulence not only removes the biofilm layer from the packing media, but also leads to shredding the suspended biofilm slots and flocs to very smaller parts. Because of this fact that rate of simultaneous nitrification-denitrification in smaller flocs is lower than big flocs, the high turbulent condition decreases rate of simultaneous nitrification-denitrification in the reactor. A higher aeration rate leads to a higher shearing force, which can shred sludge flocs [8] and accelerate biomass sloughing, which reduces the rate of phosphorous uptake and simultaneous nitrification denitrification (SND).

3.2. Effects of aeration rate on sludge characteristics

The variations of sludge characteristics with aeration rates are shown in Fig. 4. It can be seen that the aeration rate plays an important role in the evolution of sludge properties. The correlations among sludge characteristics are summarized in Table 3. It can be seen from Table 3 that sludge volume index (SVI) and SMP have a significant influence on membrane permeability ($\alpha < 0.01$), but a weak correlation was found between membrane permeability and bEPS and FI ($\alpha > 0.05$).

EPSs in the form of either bEPS or SMP are currently considered as the predominant cause of membrane fouling in MBRs [24]. Bound EPSs have been reported as key membrane foulants in MBR systems [25–28]. In contrast, several studies have also reported that bound EPSs had little correlation with membrane fouling, and instead, found SMPs to have a greater impact on sludge filtration

Parameter	Permeability	SMP	EPS	SVI	FI	Attached biofilm	Ne	TPe
Permeability	1.000	505ª	250	411	407	.731 ^b	734 ^b	400
SMP	505 ^a	1.000	.775 ^b	.150	.007	659 ^b	.594 ^b	.569 ^b
EPS	250	.775 ^b	1.000	111	127	516 ^a	.448ª	.555ª
SVI	411	.150	111	1.000	.351	446ª	.472 ^a	009
FI	407	.007	127	.351	1.000	.004	.102	426
Biofilm	.731 ^b	659 ^b	516 ^a	446 ^a	.004	1.000	934 ^b	761 ^b
Ne	734 ^b	.594 ^b	.448ª	.472ª	.102	934 ^b	1.000	.599 ^b
TPe	400	.569 ^b	.555ª	009	426	761 ^b	.599 ^b	1.000

^a Correlation is significant at the 0.05 level (2-tailed).

^b Correlation is significant at the 0.01 level (2-tailed).

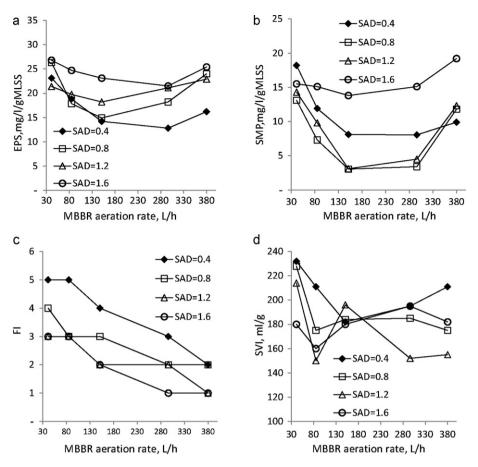


Fig. 4. Effects of aeration rates on sludge characteristics, (a) bound EPS, (b) SMP, (c) SVI, and (d) filamentous index.

[28–30]. The changes in the bEPS and SMP concentrations during the experiments are shown in Fig. 4a and b. Although the amount of bEPS fluctuated, it had a weak correlation with the aeration rate. which is consistent with other findings [8,12]. The SMP had a strong correlation with aeration rate, but it was not linear. There was also a strong correlation found between aeration rate and filamentous bacteria in the reactor. At lower aeration rates, filamentous organisms in the flocs increased, as others have previously documented [31]; however, at higher aeration rates (> $151 L h^{-1}$), we observed a reduction in filamentous organisms. As shown in Table 3, excess filamentous bacteria led to a decline in permeability, but due to a decrease in filamentous organisms at the higher aeration rates, we found a small correlation between FI and membrane permeability. Bulking sludge can cause severe cake fouling, which is induced by the fixing action of filamentous bacteria. The boundary fractal dimension of bulking sludge flocs is much larger than that of normal sludge. The flocs of bulking sludge have more irregular shape than the flocs of normal sludge. The irregular shape of bulking sludge can greatly impair the membrane filtration process [32]. In Fig. 4c and d, the FI and SVI variation with aeration rate has been plotted. At higher aeration rates, we observed little to no excess filamentous bacteria, but due to floc shredding, the settling velocity decreased, and higher SVI values were measured.

3.3. Effects of aeration rate on membrane fouling

The membrane permeability at the various combinations of aeration rates in MBBR and membrane compartments during the experiment are shown in Fig. 5. As shown in Fig. 5, the membrane permeability was decreased in the extreme low and high aeration rates, and there is a parabolic correlation between the biofilm amount on packing media and the aeration rate ($R^2 = 0.83$). The highest amount of attached biofilm on the packing media occurred at 151 L_{air} h⁻¹. At both low and high aeration rates, the foulant concentration increased (Fig. 4), which led to a loss of membrane permeability.

The correlation coefficients between membrane permeability and the concentration of soluble EPS were also presented in Table 3. We have observed that the membrane permeability decreases with increasing SMP concentration, which is in agreement with previous literature [2,33–35], yet, there was a weak correlation between the EPS content in the sludge and the membrane permeability, which has also been documented [31,36–38].

3.4. Optimum aeration rate

As shown in Figs. 2–5, the effects of aeration rate on TN, TP, LMH, EPS, FI, SVI and SMP are different. For example, in the range

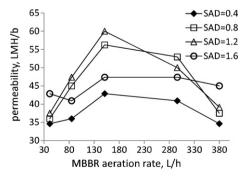


Fig. 5. Membrane permeability in different aeration rates.

of 151 L_{air} h⁻¹, amount of biofilm and membrane permeability are maximum and the concentration of total nitrogen and total phosphorous are minimum, while the minimum concentration of EPS were resulted in the rate of 185 Lh⁻¹ and the minimum concentration of SMP were resulted in the rates of 152–185 Lh⁻¹. On the other hand there is not significant correlation between aeration rate and SVI, on the other hand the minimum number of filamentous organisms was observed at highest aeration rate.

It is clear that we are not able to find an optimum aeration rate for all of these parameters. Therefore we should select the most important factor to find an optimum range. The most important factors in MB–MBR process are membrane permeability and nutrient removal rate. The correlations among these parameters were given in Table 2. The most important parameters on membrane permeability and nutrients removal are SMP concentration and amount of biofilm on packing media, respectively. Based on these, fortunately there is an optimum range of aeration rate ($151-184Lh^{-1}$ in MBBR compartment and $0.8-1.2 \text{ m}^3/\text{m}^2$ h in membrane compartment).

4. Conclusion

This investigation demonstrated that the aeration rate in the MMBR plays a significant role not only in membrane fouling control but also in the removal of nutrients from wastewater. The relationship between the aeration rate in MBBR compartment and membrane permeability was not linear, but was maximized at each combination of SAD_m and MBBR aeration rate. The optimum combination occurred at a SAD_m of 0.8 and 1.2 m³ m⁻² h⁻¹ and a MBBR aeration rate of 151 L_{air} h⁻¹. With this combination, we achieved the greatest membrane permeability, the least foulant concentration and the highest SND.

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