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Effects of the hydraulic retention time on the fouling characteristics of an anaerobic membrane bioreactor for treating acidified wastewater

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ABSTRACT

This study focused on the effect of hydraulic retention time (HRT) effect on microbial activity and fouling potential in submerged anaerobic membrane bioreactors (AMBRs).Three submerged AMBRs were operated at HRT of 14, 16, and 20 days with polypropylene U-shaped hollow fiber microfiltration membranes (nominal pore size = 0.45 µm; effective filtration area = 0.003 m2). Extracellular polymeric substances (EPS) and soluble microbial production (SMP) production increased as HRT was decreased from 20 to 14 days: from 24.2 to 29.4 mg/g volatile suspended solids (VSS), from 105.2 to 357.1 mg dissolved organic carbon (DOC)/L, respectively. Although high production of EPS and SMP at short HRT caused high cake resistance, it improved soluble COD removal efficiency by preventing small particles or macromolecules from passing through the membrane. Microbial floc in sludge showed similar resistance values regardless of HRT, while colloids and solutes had higher fouling potential than microbial floc, and this trend was severer at short SRT. In conclusion, in the operation of submerged AMBRs, HRT can significantly affect the cake formation on the membrane surface, which causes severe fouling. However, this cake layer plays a major role in additional organic removal, as it acts as a dynamic membrane.

Keywords Anaerobic membrane bioreactor (AMBR); Hydraulic retention time (HRT); Membrane fouling; Sludge characteristics; Dynamic membrane

1. Introduction

The success of anaerobic digestion is mainly attributed to efficient uncoupling of solid retention time (SRT) and hydraulic retention time (HRT) due to slow growth rates of anaerobic microorganisms. An anaerobic membrane bioreactor (AMBR), combining anaerobic digestion and membrane technology, is a promising alternative to conventional anaerobic digestion. With application of microfiltration or ultrafiltration, AMBR can offer a solid free final effluent and ultimately decrease the burden of post-treatment, which is one of the major disadvantages of the conventional anaerobic biological process.

A major obstacle to wide application of AMBRs, however, is membrane fouling. Studies have found that a number of factors affect membrane fouling: hydrodynamic conditions, membrane materials, sludge properties, substrate compositions, and so on [1]. HRT is a very

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important operating parameter in biological processes, being correlated not only to treatment efficiency but also to the biomass characteristics [2].

Generally, sludge consists of two different fractions: microbial floc and supernatant containing colloids and solutes. In an aerobic membrane bioreactor (MBR), it has been reported that each constituent affects membrane fouling differently due to having different physicochemical and biological properties [3]. However, few studies on sludge constituents and their contribution to membrane fouling have been carried out in AMBRs.

In this study, therefore, we investigate the HRT effect on overall performance and sludge properties in submerged AMBR operation for treating acidified wastewater. Also, the contribution of each sludge constituent to membrane fouling was evaluated at various HRTs.

2. Methodology

2.1. Operating conditions of AMBRs

Three submerged AMBRs having a working volume of 0.6 L were semi-continuously operated at different HRTs (14, 16, and 20 days) under mesophilic (35°C) conditions. Polypropylene U-shaped hollow fiber microfiltration membrane modules (Sumitomo Electric Fine Polymer Inc., Japan) were placed in the middle of the reactors. The fiber had a nominal pore size of 0.45 μm and an effective filtration area of 0.003 m². In order to avoid fluctuation in the feed and provide a continuous source of completely degradable organic pollutants, synthetic wastewater was used as a substrate. The concentration of the synthetic substrate, which replicated the effluent from the optimized acidogenic reactor, was 25 g/L of chemical oxygen demand (COD) and 13.5 g/L of volatile fatty acids (VFAs), as indicated in Table 1. The feed and effluent streams were controlled by two individual peristaltic pumps (Cole-Parmer, USA), which were set to the required HRT. Inoculums of 19.5 g/L of volatile suspended solids (VSS) were taken from an anaerobic digester in a local wastewater treatment plant.

Table 1
Physical and chemical characteristics of the synthetic substrate.

Compound	Concentration (mg/L)	Compound	Concentration (mg/L)
Organic and nutrients		Trace nutrients	
Maltose	10,007.9	KH_2PO_4	1,350
Glucose	1,121.0	K_2HPO_4	1,750
NH4Cl	1,406.5	$CaCl_2{\cdot}2H_2O$	380
$NaHCO_3$	1,606.8	$MgCl_2{\cdot}6H_2O$	500
Acetate	2,608.1	$FeCl_2{\cdot}4H_2O$	20
Propionate	1,588.0	$MnCl_2{\cdot}4H_2O$	0.5
iso-Butyrate	679.0	H_3BO_3	0.25
n-Butyrate	5,413.0	$ZnCl_2$	0.25
iso-Valerate	1,610.0	$CuCl_2$	0.15
n-Valerate	2,061.0	$Na_2MoO_4{\cdot}2H_2O$	0.05
		$CoCl_2{\cdot}6H_2O$	2.50
		$NiCl_2{\cdot}6H_2O$	0.25
		Na_2SeO_3	0.25
		Na_2SO_4	150

2.2. Resistance analysis

The resistance-in-series model, which describes the permeate flux-transmembrane pressure (TMP) relationship over the entire domain of pressure, was used to analyze membrane fouling resistances. Based on this model, the permeate flux on the applied TMP can be described by Darcy's law as follows:

$$J_v(t) = \frac{1dV}{Adt} = \frac{\Delta P}{\mu(R_t)} = \frac{\Delta P}{\mu(R_m + R_c + R_f)}\,, \qquad (1)$$

where J_v is the permeate flux, m³/m²/h; V is the total volume of permeate, m³; A is the membrane area, m²; ΔP TMP, Pa; μ is the permeate viscosity, Pa·s; R_t is the total resistance, m⁻¹; R_m is the intrinsic membrane resistance, m⁻¹; R_c is the cake layer resistance, m⁻¹; and R_f is the fouling resistance, m⁻¹, which is often related to adsorption of solutes and pore blocking.

The experimental procedure to obtain each resistance value was as follows: (i) R_m was obtained by measuring the water flux of ultra-pure water having a resistivity of approximately 18 MΩcm; (ii) R_t was calculated from the final flux data of biosolids filtration and TMP; and (iii) the membrane surface was then flushed with deionized water and cleaned with a sponge to remove the cake layer. After that, the pure water flux was measured again to obtain the resistance of $R_m + R_i$: The fouling resistance (R_i) was calculated from processes (i) and (iii) and the cake resistance (R_c) from processes (ii) and (iii).

Membrane resistance was analyzed using an Amicon model 8200 ultrafiltration stirred cell (200 mL process volume). An Amicon cellulose acetate membrane YM 30 (MWCO: 30,000 Dalton, diameter: 63.5 mm, Amicon Inc., USA) was used for the filtration of sludge following procedures provided in the literature [4]. Prior to the filtration test, the concentration of the sludge sample was adjusted to 3,000 ± 100 mg/L of suspended solids (SS) to avoid any concentration effect on the membrane fouling. The filtration test was performed at a constant suction pressure of 10 psi. R_t was obtained by filtration of the sludge sample from each AMBR, and R_m, R_f, and R_c were estimated by the same method as described above. A supernatant containing colloids and solutes was produced by centrifugation (3500 rpm for 5 min) from each sludge sample.

2.3. Analytical methods

For extraction of extracellular polymeric substances (EPS), the sludge was concentrated by centrifugation (3200 rpm, 10 min) and the pellet that remained after decanting the supernatant was resuspended with saline water (0.9% NaCl solution). This process was carried out twice more at the same speed and time. The EPS extraction was performed by heat treatment of the resuspended solution at 90°C for 1 h. After cooling to room temperature, the solution was centrifuged at 3200 rpm for 30 min.

The extracted solution was analyzed for total carbohydrates and proteins. Carbohydrates in the EPS were determined according to the phenol–sulfuric method with glucose as a standard [5]. Absorbance was measured against a blank at 480, 484, and 490 nm with a Beckman UV-Visible Spectrophotometer. Proteins were determined by the Folin method with bovine serum albumin as a standard [6]. After incubation at 35°C for 30 min, absorbance was measured at 562 nm versus a reagent blank.

The CH_4 gas content was analyzed using a gas chromatograph (GC, Gow Mac series 580) equipped with a thermal conductivity detector (TCD) and a 2 m × 2 mm stainless-steel column packed with a Porapak Q mesh (80/100).

3. Results and discussion

3.1. Performance of AMBRs

The average CH_4 production rate (MPR) was increased as the HRT decreased: 0.10, 0.15, and 0.28 $m^3/m^3/d$ at HRT 20, 16, and 14 days, respectively. During operation of the reactor, MPR in all AMBRs showed a wide fluctuation, as reported in a previous study [7]. This may be partially explained by the loss of CH_4 with the permeate due to the membrane suction and irregular rise of gas bubbles confined in membrane modules. Also, it has been reported that CH_4 solubility in water was 15 mL/1,000 mL at 1 atm and 35°C [8], a level which could lead to a low MPR.

The performance of AMBRs can be affected by different operating conditions such as membrane morphology, reactor configuration, substrate characteristics, and microorganisms. In this study, AMBRs showed performance comparable to that documented in a previous study conducted under similar operating conditions [9], while the performance was lower than that of studies applying real wastewater and a UF membrane [10,11].

Table 2
COD removal efficiency in AMBRs at different HRTs

HRT (days)	Influent	Reactor	Effluent
14	25,000[a]	1909.8 ± 90.9	95.1 ± 8.6
	(100)[b]	(92.4 ± 0.4)	(99.6 ± 0.0)
16	25,000	1388.3 ± 80.7	152.5 ± 22.1
	(100)	(94.5 ± 0.4)	(99.4 ± 0.1)
20	25,000	642.8 ± 26.9	162.1 ± 12.9
	(100)	(97.5 ± 0.1)	(99.4 ± 0.1)

[a]Results are expressed as the average ± one standard deviation.
[b]Data in parentheses are removal efficiency percentage; average ± one standard deviation.

Table 2 shows the COD removal efficiency for all of the AMBRs at a steady state. The removal efficiency of the reactors was calculated with the soluble COD concentrations of the supernatant. Soluble COD concentration of the effluent, meanwhile, provided an overall indicator of the removal efficiency of overall the AMBRs. As shown in Table 2, COD concentrations in the reactor increase slightly with decreasing HRT, and this is a result of an increased organic load on the biomass and the production of soluble microbial products (SMP) due to stress at such low HRT [12]. Even though the COD concentration in the reactor was highest at a HRT of 14 days, the overall COD removal efficiency of the AMBRs was greater than 99% regardless of the HRT. This can be explained by the high fraction of soluble COD rejected by the cake layer on the membrane, which is also known as a dynamic membrane [13].

The observed difference of 2–7% in the COD removal between the bioreactor and the overall process is attributed to the soluble COD removed by membrane rejection or degraded by a biofilm when passing through the membrane. Previous studies found that the soluble COD in the reactor is consistently two to three times higher than that in the effluent due to the rejection of some organics by the membrane [8,13]. In this study, however, the soluble COD in the reactor was four to twenty times higher than that in the effluent. This is attributed to the much higher organic loading rate, i.e., 2.5g COD/L/d, than that employed in previous studies.

3.2. EPS production

EPS is known to be very heterogeneous, comprising a variety of polymeric materials: carbohydrates, proteins, lipids, and nucleic acids. In this work, however, the sum of the amounts of total carbohydrates and proteins was considered to represent the total EPS, because these are

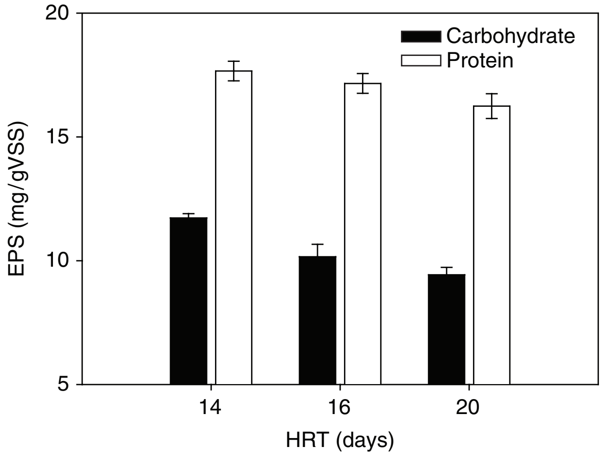

Fig. 1. The concentration of EPS components at different HRTs.

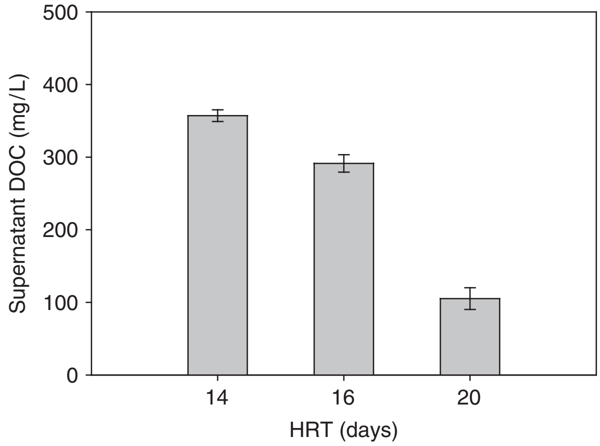

Fig. 2. The concentration of supernatant DOC at various HRTs.

the dominant components typically found in extracted EPS [14]. The amount and composition of EPS depend on the growth conditions of the biofilm or sludge flocs [15].

Figure 1 shows the concentration of EPS components (carbohydrates and proteins) in microbial floc at each HRT. The concentration of carbohydrates and proteins increased as HRT decreased: 24.2, 27.3, and 29.4 mg/g VSS at HRT of 20, 16, and 14 days, respectively. Carbohydrates are synthesized extracellularly for a specific function, while proteins can exist in the extracellular polymer network due to the excretion of intracellular polymers or cell lysis [16]. The change of protein concentration resulted from the change of microbial activity according to the HRT variation, while variation of the carbohydrates is related to the food to microorganism (F/M) ratio. At longer HRT and lower F/M ratios, carbohydrates in the microbial floc decline as an available carbon source. In contrast, at shorter HRT and higher F/M ratio, excess carbon substrates are likely to be converted to polymers accumulated as EPS [15].

3.3. SMP production

Supernatant dissolved organic carbon (DOC) can be considered as SMP if all soluble organics from the feed are easily degraded by microorganisms and there is no significant contribution of feed to the residual soluble COD. In this study, it was found that the concentration of supernatant DOC, indicating SMP concentration increased as HRT decreased: 105.2, 291.4, and 357.1 mg DOC/L at HRT 20, 16, and 14 days, respectively (Fig. 2).This confirms that microorganisms produce much more SMP due to stress under high organic loading rate (OLR) conditions [8]. Another contributor to increased SMP at shorter HRT is the release of EPS in bulk solution. SMP from cell lysis and released EPS adsorbs on the membrane surface and forms a gel structure, which acts as a dynamic membrane [14]. Therefore, it can be concluded that SMP production at shorter HRT forms a dynamic membrane, which prevents solutes and colloids from adsorbing onto the membrane surface and narrowing pores.

3.4. Filtration resistances

In order to evaluate the membrane fouling potential of sludge at different HRTs, membrane resistance was analyzed. As shown in Table 3, total resistance was increased as HRT decreased, i.e., 2.56, 2.96, and 3.39×10^{13} m^{-1} at HRT of 20, 16, and 14 days, respectively, and this increase was mainly caused by cake formation

Table 3
Comparison of each resistance in AMBRs at different HRTs (unit: m^{-1}).

Unit: m^{-1}	HRT 14 days		HRT 16 days		HRT 20 days	
R_t	3.39×10^{13}	(100%)	2.96×10^{13}	(100%)	2.56×10^{13}	(100%)
R_c	2.96×10^{13}	(87.3%)	2.54×10^{13}	(85.8%)	2.15×10^{13}	(83.7%)
R_f	0.24×10^{13}	(7.0%)	0.21×10^{13}	(7.1%)	0.22×10^{13}	(8.5%)
R_m	0.19×10^{13}	(5.7%)	0.21×10^{13}	(7.1%)	0.20×10^{13}	(7.8%)

on the membrane surface. A slight rise in R_c, 2.15, 2.54, and 2.96 × 10^{13} m^{-1}, was observed with HRT decrease from 20 to 14 days. Although there was not a large difference in SS concentration with HRT variation, both total and cake resistance increased as HRT decreased. The cake formation derives from EPS production from anaerobic microorganisms. It was previously reported that specific resistance increased linearly with rise in bound EPS from 20 to 130 mg g^{-1} SS [14].

Higher EPS production at shorter HRT led to the cake layer formation and ultimately improvement of treatment efficiency. The membrane surface formed a thin and porous layer, which acted as a dynamic membrane, preventing the passage of small particles or macromolecules. The relationship between cake resistance and formation of a fouling layer that can act as a dynamic membrane was shown in a previous study [17]. Although this cake formation increased the total resistance of the membrane, it facilitated improved treatment efficiency and the production of a high quality effluent, as described in Section 3.1.

3.5. Contribution of sludge constituents to fouling

In order to investigate the contribution of each constituent (microbial floc and supernatant) of sludge to fouling at different HRTs, further batch filtration tests were performed. Resistances in microbial floc were quantified by the differences between resistances in the sludge and supernatant. The summation of R_c and R_f of each fraction in sludge samples from each AMBR is presented in Fig 3. It was found that the value in microbial floc had similar values regardless of HRT in the AMBRs: 1.26, 1.22, and 1.24 × 10^{13} m^{-1} at HRT of 14, 16, and 20 days. On the contrary, the values of the supernatant enlarged as HRT decreased, and the relative contribution to the total resistance increased from 52% to 63% as HRT decreased from 20 to 14 days. From this, it is determined that solutes and colloids, mainly resulting from cell lysis, are likely to have higher fouling potential than microbial flocs, and this trend becomes severer at short HRT.

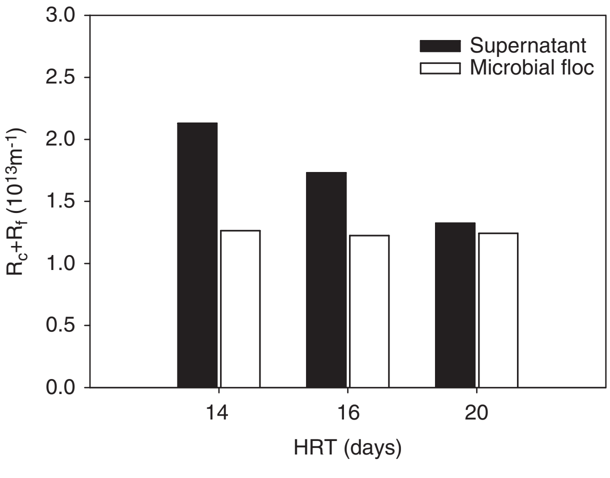

Fig. 3. Resistance of various fractions in sludge samples at different HRTs.

Several researchers have quantified the fouling potential in aerobic MBRs caused by each fraction of sludge. In Table 4, the results of the present study are compared with those of recent research, which quantified the contribution of each sludge fraction in an aerobic MBR (corresponding results for AMBRs have not been reported yet). The differences were caused by the membrane properties, the sludge characteristics, and the operational type. Despite these differences, it is clear that supernatant and microbial floc respectively contribute to increased resistance in AMBR operation.

3.6. Fouling characteristics of AMBR operation at various HRTs

In this study, EPS and SMP were found to be key contributors to membrane fouling, especially at shorter HRT. As HRT decreased, total resistance increased. This increase was mainly caused by the cake resistance, which showed a strong positive correlation with total EPS and SMP concentration. In short, with a decrease of HRT, EPS and SMP production increased, and ultimately cake formation and total resistance increased. This confirms that HRT affects microbial activity and eventually alters the production of metabolic substances (EPS and SMP). The increase of cake formation, however, provides high COD removal, even at high OLR. Consequently, even though AMBR operation at short HRT causes severe cake formation and fouling, it provides better performance due to a dynamic membrane effect.

Fouling resistance by supernatant ($R_{supernatant}$) increased as HRT decreased, while $R_{microbial\ floc}$ did not show a remarkable change with HRT variation. Hence, it is concluded that HRT has a stronger influence on $R_{supernatant}$ than $R_{microbial\ floc}$, and solutes and colloids in the supernatant have higher fouling potential, especially at shorter HRT.

Table 4
Relative contribution (%) of various fractions in sludge to membrane fouling.

References	This study			[3]			[18]
Fractions	HRT 14 d	HRT 16 d	HRT 20 d	SRT 20 d	SRT 40 d	SRT 60 d	SRT 20 d
Supernatant (%)	63	59	52	37	28	29	76
Microbial floc (%)	37	41	48	63	72	71	24

4. Conclusion

This study investigated the effect of HRT on the overall performance and microbial activity in the operation of submerged AMBRs for treating acidified wastewater. Key parameters with respect to membrane fouling were also evaluated under different HRT conditions. The following conclusions were obtained.

1. The AMBR with a HRT of 14 days showed the highest MPR (0.28 $m^3/m^3/d$) and COD removal (99.6%). A large amount of soluble COD (7.2%) was removed by the cake layer on the membrane surface. This cake layer played a critical role in additional organic removal as it acted as a dynamic membrane.
2. Total EPS and SMP concentration increased as HRT decreased, and these metabolic substances increased filtration resistance by forming a cake layer on the membrane surface. Short HRT affected the anaerobic microbial activity mainly due to stress at high OLR, and led to severe membrane fouling.
3. The overall fouling resistance increased as HRT decreased. Two different fractions of sludge in AMBR showed different fouling potential. Microbial floc showed similar resistance values regardless of HRT, whereas colloids and solutes appeared to have greater fouling potential than microbial floc, and this trend was severer at short SRT.

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