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Influence of Mechanical Mixing Rates on Sludge Characteristics and Membrane Fouling in MBRs

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Abstract: The influence of shear intensity (G) induced by mechanical mixing on activated sludge characteristics as well as membrane fouling propensity in membrane bioreactors (MBRs) was investigated. Four MBRs were operated at different mechanical mixing conditions. The control reactor (MBR_0) was operated with aeration only supplemented by mechanical stirring at 150, 300, and 450 rpm in MBR_{150} , MBR_{300} , and MBR_{450} , respectively. It was found that the MBR_{300} demonstrated minimum rate of membrane fouling. The fouling potential of the MBR_{300} mixed liquor was lowest characterized by the specific cake resistance and the normalized capillary suction time (CST_N). Moreover, it was found that the mean particle size reduced with an increase in the shear intensity. These results reveal that membrane fouling can be significantly mitigated by appropriate shear stress on membrane fibers induced by mechanical mixing condition.

Keywords: Membrane fouling, shear intensity, particle size distribution, extracellular polymeric substances (EPS), specific cake resistance

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INTRODUCTION

Membrane bioreactor (MBR) offers several advantages compared with conventional wastewater treatment processes including high biodegradation efficiency, excellent quality of effluent, smaller sludge production and compactness (1). However, the widespread application of the MBR process is constrained by membrane fouling and it is considered as the most serious problem affecting system performance. Fouling results in permeate flux decline due to the interaction of membrane and activated sludge leading to frequent membrane cleaning and necessary membrane replacement. Fouling of membranes in MBRs is determined by three factors, namely: the nature of the feed to the membrane, the membrane properties, and the hydrodynamic condition experienced by the membrane (2).

So far, several techniques for fouling control have been investigated including sub-critical flux operation, intermittent suction and backwashing (3). Membrane scouring with bubbling has been an effective hydrodynamic technique for fouling reduction in submerged MBRs. In recent researches, MBRs were operated with diffuser at the base to maintain aerobic condition and additional diffuser for air scouring of the membrane module (4–5). Lee et al. (6) found that membrane fouling in terms of rate of trans-membrane pressure (TMP) rise was dependent on air flow rate and TMP rose up more slowly with the increase in air flow rate. However, the high aeration intensity necessary to provide effective bubbling also leads to changes in the growth rate, the F/M ratio and the microbial community of sludge (7). Moreover, hollow fiber (HF) modules possessing high membrane surface area to footprint ratio are prone to excessive fouling due to the poor hydrodynamic conditions within the fiber bundles. It was found that the axial velocities induced by bubbling within HF module could be up to 10 times lower than the one outside the fiber bundle and the surrounded (center) fibers performed poorly compared to the outer fibers (8). High bundle packing density causes heterogeneous scouring with bubbling and low shear intensity on surrounded fibers leads to solids accumulation between the fibers in the bundle. The present study was aimed at investigating the influence of mechanical mixing on HF membrane fouling propensity and on physical properties of the activated sludge. The membrane fouling behaviors and the sludge suspension characteristics from the MBRs at different mixing conditions were compared to determine the optimum mixing rate.

MATERIALS AND METHODS

Experimental Setup and Operation

HF membrane modules (Mitsubishi Rayon) were submerged in bioreactors with 10 L working volume. The HF membranes were made of polyethylene

having a pore size of 0.1 μm and an effective membrane filtration area of 0.42 m^2 . Synthetic wastewater simulating municipal wastewater was used as a substrate in the biological process with COD:N:P ratio of 100:10:2 and an organic loading rate (OLR) of 2.4 $\text{kg}/\text{m}^3 \cdot \text{d}$. The composition of synthetic wastewater included dextrose (516 mg/L), soya protein (250 mg/L), NH_4Cl (229 mg/L), KH_2PO_4 (70 mg/L), CaCl_2 (10 mg/L), MgSO_4 (10 mg/L), and FeCl_3 (3 mg/L). pH in the bioreactors was maintained between 7.0 and 7.5 using NaHCO_3 (750 mg/L). Domestic activated sludge was acclimatized over a period of two months before seeding of the MBRs.

A constant air supply by filtered compressed air through air diffusers was maintained at a flow rate of 5 L/min in all the MBRs. Based on cross-sectional area of bioreactor, the air flow rate was equivalent to an aeration intensity of 10.6 $\text{m}^3/\text{m}^2 \cdot \text{h}$. Dissolved oxygen (DO) in the MBRs was maintained between 2 and 5 mg/L. The varying condition among the four MBRs was the mechanical mixing with no stirring in control reactor (MBR_0) followed by stirring at 150, 300, and 450 rpm in MBR_{150} , MBR_{300} , and MBR_{450} , respectively. The membrane filtration was operated in a cyclic mode (10 min on, 2 min off) at a constant flux to maintain a hydraulic retention time (HRT) of 8 h. For submerged MBRs, intermittent suction is an effective approach for suppression of fouling (9). The permeate suction pressure was recorded using digital manometers connected to the suction line of the membrane modules.

Analytical Methods

Mixed liquor suspended solids (MLSS), volatile mixed liquor suspended solids (MLVSS), chemical oxygen demand (COD), and capillary suction time (CST) were determined according to APHA (10). Particle size distribution (PSD) of sludge samples was determined by light scattering technique using Mastersizer S (Malvern, UK). The particle size range was measured between 0.05 and 750 μm with an instrument accuracy of $\pm 1\%$.

EPS Analysis

Mixed liquor samples of 50 mL from the four MBRs were taken and cooled immediately at 4°C to minimize microbial activity. Soluble EPS was obtained by centrifugation of the mixed liquor at 4000 g for 20 min followed by high speed centrifugation at 20,000 g for 20 min and separation of the supernatant (2). Bound EPS was extracted from the mixed liquor using cation exchange resin (CER) extraction method (11). The CER (DOWEX HCR-S/S, Dow Chemical Company, USA) used was in Na^+ form with bead size distribution range between 16–50 mesh. The centrifuged sludge was re-suspended in phosphate buffer solution and the CER (70 g CER/g VSS) was added and mixed at 600 rpm for 1 h. Then the mixture

was centrifuged twice at 4000 g for 10 and 20 min, respectively, to obtain the supernatant as bound EPS. Carbohydrate and protein fractions of the soluble and bound EPS were measured by the colorimetric methods of Dubois et al. (12) and Lowry et al. (13), respectively. D-Glucose and Bovine serum albumin (BSA) were used as carbohydrate and protein standards, respectively.

Determination of Membrane Fouling

The extent of membrane fouling in the MBRs was monitored in terms of rise in TMP with operational time. In this regard, flux and TMP were recorded on regular basis. The membrane fouling rates ($d\text{TMP}/dt$) were determined from the TMP profiles. The operation was stopped when TMP reached 30 kPa and chemical cleaning procedure was carried out. Prior to the chemical cleaning, the membrane unit was physically washed with tap water to remove visible cake layer from the membrane fibers. Then the membrane unit was immersed for 8 h in a solution constituting NaOCl (effective chlorine concentration of 3,000 mg/L) and 4% aqueous NaOH. Following the immersion period, the membrane unit was thoroughly rinsed with water to remove the chemical. This chemical cleaning protocol suggested by the membrane supplier (Mitsubishi Rayon) was able to recover intrinsic permeability 90–95%.

Besides on-line data, batch filtration tests were performed to determine the specific cake resistance (α) of the sludge samples. The test was conducted in a 400 mL unstirred filtration cell (Model 8400, Amicon, USA) using a 0.22- μm flat-sheet cellulose membrane filter (GVWP 09050, Millipore, USA). The cell was filled with 200 mL of mixed liquor sample and a constant pressure of 30 kPa was applied by pressurized nitrogen from a gas cylinder. The filtrate was continuously recorded by an electronic balance connected to a notebook using WINWEDGE software. The specific cake resistance, α (m/kg) was calculated (9) by

$$\alpha = \frac{2000A^2\Delta P t/V}{\mu C} \quad (1)$$

where ΔP is the applied pressure (kPa), A is the filtration area (0.00418 m^2), C is the MLSS concentration (kg/m^3), μ is the viscosity of permeate ($\text{N}\cdot\text{s}/\text{m}^2$) and $[(t/V)/V]$ (s/m^6) is the slope of the straight portion of the curve that is obtained by plotting the time of filtration to the volume of filtrate (t/V) versus the filtrate volume (V).

RESULTS AND DISCUSSION

The shear intensity in the MBRs was quantified by the mean velocity gradient (G) using expressions (14) presented in Table 1 and values reported in Table 2.

Table 1. Power requirement and velocity gradient expressions

Expression	Unit	Formula	Remarks
Mechanical power (P_m)	W	$P_m = N_p \rho n^3 D^5$ $(N_R \geq 10,000)$ $N_R = D^2 n \rho / \mu$	ρ = Density of mixed liquor (1000 kg/m ³); n = mixing speed (rev/s); D = diameter of impeller (0.1 m); N_p = Power number for impeller ($N_p = 1.1$); N_R = Reynolds number
Pneumatic power (P_p)	kW	$P_p = p_a V_a \ln p_c / p_a$	p_a = Atmospheric pressure (kPa); V_a = air flow rate (m ³ /s); p_c = air pressure at the point of discharge (kPa)
Total power (P_T)	W	$P_T = P_m + P_p$	
Velocity gradient (G)	1/s	$G = \sqrt{P_T / \mu V}$	V = reactor volume (0.01 m ³); μ = dynamic viscosity (N-s/m ²)

According to Table 2, the pneumatic mixing due to the air supply remained constant while the mechanical mixing due to stirring varied resulting in G variation among the MBRs.

The MBRs were run in a steady-state condition over a period of 120 days and the values of all the parameters were averaged along with standard deviation. The MLSS concentration was maintained between 6–8 g/L with MLVSS/MLSS ratio of approximately 90% in the MBRs at sludge retention time (SRT) of 40 d. The COD removal efficiency of the MBRs was above 95% representative of effective biodegradation and physical separation by the HF membranes.

Filtration Behavior in the MBRs

The variable mixing intensities in the MBRs resulted in membrane filtration behaviors as shown in Fig. 1. It shows the rise in TMP for each MBR over

Table 2. Shear intensity (G) in the MBRs

MBR	Mechanical mixing (rev/s)	Pneumatic mixing (m ³ /h)	Reynolds number (N _R)	P _m (W)	P _p (W)	Total P (W)	Power/volume (W/m ³)	G (1/s)
MBR ₀	0.0	0.3	0	0.00	0.17	0.17	17	83
MBR ₁₅₀	2.5	0.3	10,000	0.17	0.17	0.34	34	117
MBR ₃₀₀	5.0	0.3	20,000	1.38	0.17	1.55	155	249
MBR ₄₅₀	7.5	0.3	30,000	4.64	0.17	4.81	481	439

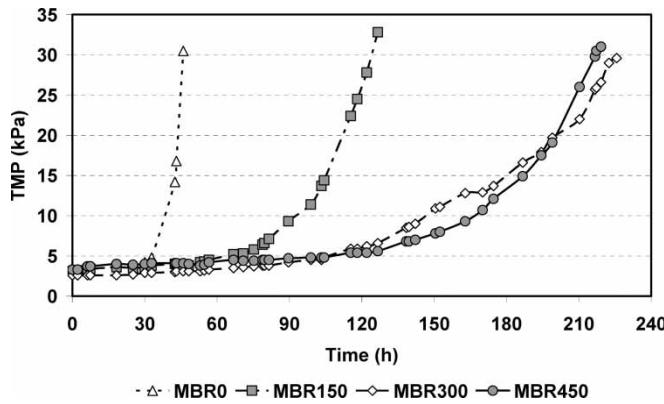


Figure 1. TMP variation versus operating time during the MBRs operation.

a typical filtration period. It was observed that the membrane in MBR_0 fouled rapidly followed by the one in MBR_{150} . However, membrane filtration in the MBR_{300} and MBR_{450} could be achieved up to five times the filtration period of MBR_0 . Taking into consideration the relatively similar biomass concentrations among the MBRs, MBR_{300} demonstrated minimum fouling in terms of the filtration duration. Moreover, the filtration duration could not be further increased in MBR_{450} with a higher G as compared to the one in MBR_{300} . In Fig. 1, all the TMP profiles exhibited two-stage process. Initially, linear gradual TMP rise was observed followed by sudden increase in the rate of TMP rise leading to need for membrane chemical cleaning.

The two-stage process in membrane fouling behavior has been extensively investigated and explained by Cho and Fane (15) and later by Zhang et al. (2). Prior to these two-filtration steps, a conditioning period has been observed due to the initial adsorption of colloids and organics mostly before cake layer formation initiates under sub-critical flux operation (2). After membrane conditioning of MBRs, the bioflocs initiate cake formation on membrane fibers and between fibers in low shear stress regions of the HF bundle during the first stage. Over time, the cake deposition worsens leading to depletion of effective pores resulting in TMP rise. At the end of this stage, exponential TMP increase is observed when the effective pores become critical and permeate productivity redistributes to the less fouled pores, for which local flux exceeds the critical flux (16).

There are two significant parameters during the first stage: the critical time (t_{crit}) over which the first stage is maintained and the fouling rate ($dTMP/dt$) during this stage (17). Figure 1 shows that the first stage of fouling was maintained until TMP reached 7 kPa in the four MBRs operation. The TMP profile data reveals that the t_{crit} was observed at approximately 30, 80, 130, and 140 h during operation of the MBR_0 , MBR_{150} , MBR_{300} , and MBR_{450} , respectively. The longer duration maintained in the

MBR₃₀₀ and MBR₄₅₀ for the first fouling stage can be mainly attributed to the high shear intensity on membrane fibers induced by mechanical stirring.

Membrane Fouling Rates

Based on the membrane filtration performance in the MBRs (Fig. 1), the membrane fouling rates ($d\text{TMP}/dt$) during the first and second fouling stages were determined. The first stage ranged from the start-up TMP of 3 kPa to 7 kPa and the second stage ranged from 10 kPa to the terminating TMP of 30 kPa. The fouling rates were determined by the slope of the linear curve from the TMP versus time plot as shown in Fig. 2.

The first stage fouling rates are representative of pore blocking, biopolymer deposition, biofilm attachment and growth, all contributing to steady TMP rise (2). Figure 2 shows that the first stage fouling rates in the MBRs decreased linearly with increase in the shear intensities. Indeed, the high shear stress exerted on membrane fibers retard biofloc deposition and avoids sludge accumulation between fibers, particularly in the central region of the bundle. Wicaksana et al. (18) found that increased fiber movement induced by high air flow rate appeared to reduce the rate of biofloc deposition on the membrane surface and slow down the rise of TMP at fixed flux. The second stage fouling rates, as expected, were found to be significantly higher than that in the first stage. However, the second stage fouling rate in MBR₄₅₀ operation was found to be higher than that in MBR₃₀₀ which was indicative of optimum shear intensity in the MBR₃₀₀ as shown in Fig. 2. At this point, it can be inferred that the mixing intensity of a certain extent is feasible to mitigate fouling, beyond which it becomes disadvantageous. The

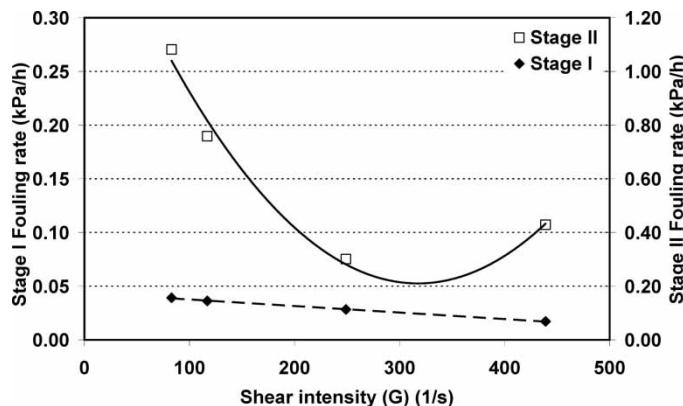


Figure 2. Fouling rates corresponding to shear intensities in the MBRs (G in MBR₀ = 83 s^{-1} ; G in MBR₁₅₀ = 117 s^{-1} ; G in MBR₃₀₀ = 249 s^{-1} ; G in MBR₄₅₀ = 439 s^{-1}).

sludge characteristics of the MBRs were investigated to determine the influence of the mixing intensity on deposited sludge cake properties.

Sludge Filterability Characteristics

Sludge filterability was characterized by the CST and the specific cake resistance (α). CST is a quantitative measure of the rate of water release from sludge and is indicative of the filterability and dewaterability of sludge. In order to minimize the effect of suspended solids (SS) on CST, the CST values are normalized by dividing with MLSS concentration for each sample. In contrast, specific cake resistance (α) is a more authentic and reliable parameter for measuring the fouling potential or filterability of sludge cake. Figure 3 shows the averaged specific cake resistance (α) and the normalized CST (CST_N) for sludge samples from the MBRs. The filterability of sludge improved with increase in shear intensity up to 249 s^{-1} (MBR_{300}) in terms of both specific cake resistance (α) and CST_N . However, the filterability deteriorated for the MBR_{450} sludge sample indicating that floc properties of MBR_{300} exhibited lowest fouling potential. It can be inferred from the low CST_N and specific cake resistance (α) values of MBR_{300} sludge that it was the appropriate hydrodynamic condition as well as the suitable sludge filterability characteristics that influenced the observed low fouling rates.

Particle Size Distribution (PSD)

Figures 4(a) and (b) shows the PSD of the sludge in the MBRs within range $0.05\text{--}750\text{ }\mu\text{m}$ and $0.05\text{--}20\text{ }\mu\text{m}$, respectively. The median particle sizes were

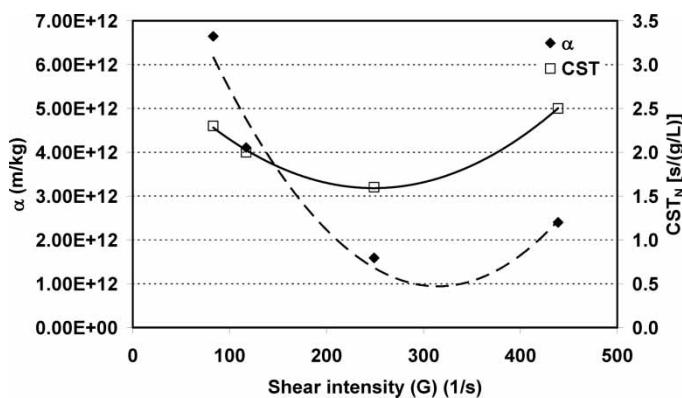


Figure 3. Specific cake resistance (α) and CST_N of the MBR sludge samples (G in $MBR_0 = 83\text{ s}^{-1}$; G in $MBR_{150} = 117\text{ s}^{-1}$; G in $MBR_{300} = 249\text{ s}^{-1}$; G in $MBR_{450} = 439\text{ s}^{-1}$).

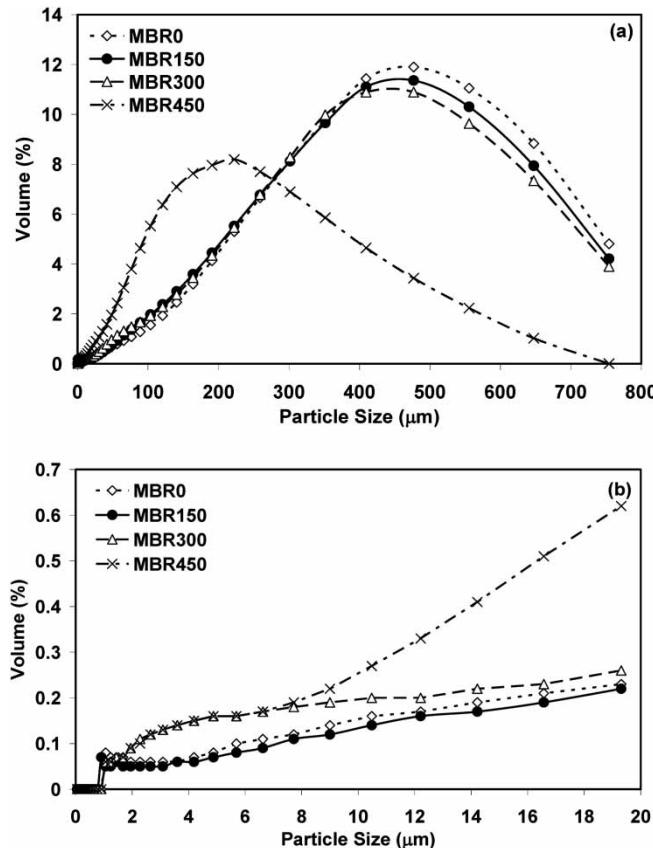


Figure 4. Particle size distribution of sludge suspensions in the MBRs (a) 0.05–750 μm and (b) 0.05–20 μm.

found to be 398, 379, 367, and 184 μm in the MBR₀, MBR₁₅₀, MBR₃₀₀ and MBR₄₅₀, respectively. Figure 4(a) shows that the bio-particles became relatively smaller with increase in mixing rate from MBR₀ to MBR₃₀₀ with similar extent of distributions. However, MBR₄₅₀ exhibited significant reduction in particle sizes as well as scattered distribution. Moreover, Fig. 4(b) shows that the bio-particle distributions from MBR₀ to MBR₃₀₀ revealed similar trends within the range of 0.05–20 μm with the exception of MBR₄₅₀ where increased percentage of particles greater than 10 μm suggested breakage of floc structure. The floc breakage into smaller particles under severe turbulent condition of MBR₄₅₀ could have induced the deterioration of the sludge filterability as depicted in Fig. 3. However, the improved fouling potential of MBR₃₀₀ sludge could not be explained with the PSD results.

Bai and Leow (19) studied the effect of mechanical mixing intensity on membrane fouling in a cross-flow microfiltration system and observed that

the finer particles ($<50\text{ }\mu\text{m}$) caused severe membrane fouling. However, Sombatsompob et al. (20) found that the membrane fouling improved in an attached growth MBR system in the presence of smaller bio-particles (17–33 μm) as compared to larger particles (65–226 μm) in a suspended growth MBR system. Similarly, Lee et al. (6) found that in a submerged MBR operation, the filtration performance enhanced with increase in air flow rate despite decrease in microbial floc size. Since activated sludge is a complex broth, it is difficult to explain the membrane fouling phenomenon explicitly on the basis of particle size.

Soluble and Bound EPS

The soluble and bound EPS concentrations were determined by the addition of carbohydrate and protein concentrations measured in respective soluble and bound samples of the MBRs as shown in Figs. 5 and 6, respectively. Figure 5 shows that the carbohydrate fraction of the soluble EPS was predominant among the MBR sludge. Moreover, soluble EPS concentrations in mechanically mixed MBRs were similar and slightly higher than that in the control system (MBR_0). Figure 6 shows that the bound EPS concentration in MBR_{450} was significantly higher as compared to that in the MBR_0 . The carbohydrate concentration in all the extracted samples was found to be similar but an increase was noticed in the protein levels of MBR_{450} . The high bound protein concentration in the MBR_{450} could be due to the bio-floc breakage releasing protein found at or outside the cell surface and in the intercellular space of microbial aggregate. The variation in the soluble and bound EPS concentrations, considered as major foulants, could not

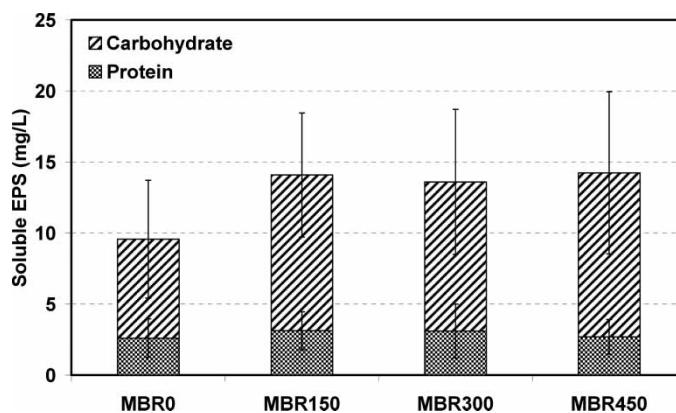


Figure 5. Soluble EPS in mixed liquor of the MBRs.

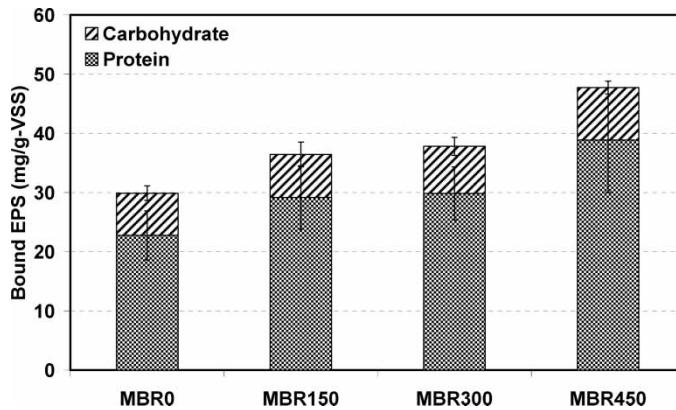


Figure 6. Bound EPS in mixed liquor of the MBRs.

adversely influence the fouling mitigation achieved by the mechanical mixing condition in the MBR_{300} .

Discussion

The optimum shear intensity of 249 s^{-1} in the MBR_{300} achieved low fouling rates in both stages. The first stage fouling was believed to be mitigated by the high shear intensity of mixed liquor turbulence inducing high fiber movement and slow deposition of biomass on the membrane fibers and between the fibers within the bundle. After cake formation in the second fouling stage, the

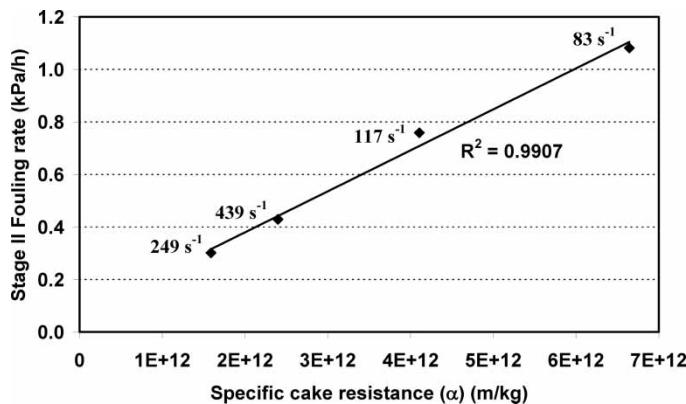


Figure 7. Relationship between Stage II fouling rate versus specific cake resistance (α) (G in $MBR_0 = 83\text{ s}^{-1}$; G in $MBR_{150} = 117\text{ s}^{-1}$; G in $MBR_{300} = 249\text{ s}^{-1}$; G in $MBR_{450} = 439\text{ s}^{-1}$).

fouling rate was improved by the high porosity and connectivity of deposited sludge cake depicted by the low specific cake resistance. The relationship between the specific cake resistances and the second stage fouling rates of the MBRs is shown in Fig. 7. The linear curve shows a strong relationship between the specific cake resistances and the second stage fouling rates with an r-squared value of 0.99 suggesting the dependence of the second stage fouling rate on the specific cake resistance of a deposited cake layer. Thus, it can be postulated that specific cake resistance can be a reliable parameter to predict the extent of the second stage membrane fouling rate in MBR filtration process.

CONCLUSION

The effect of sludge characteristics on membrane fouling were investigated in submerged MBRs operated at different mixing intensities. The membrane fouling behavior in terms of TMP variation with operating time was investigated. It was found that minimum fouling tendency was observed during MBR₃₀₀ operation with shear intensity (G) of 249 s⁻¹. Moreover, the CST_N and the specific cake resistance (α) of the sludge from MBR₃₀₀ were also found to be the lowest. Increase in G value beyond 249 s⁻¹ was not able to retard fouling any further and the fouling potential deteriorated indicating mechanical mixing of 300 rpm as the optimum. A slight variation was observed in the particle size distribution from MBR₀ to MBR₃₀₀. On the contrary, the bioflocs broke into smaller particles in MBR₄₅₀ under extreme shear stress conditions. Based on these results, it can be postulated that improved filtration performance of HF membranes can be achieved in submerged MBRs by physical modification of sludge properties and slow deposition of bioflocs on membrane surface induced by high mixing intensity. Moreover, increased fiber movement and homogeneous agitation by optimal shear intensity could avoid “dead zones” formation within the HF bundle.

Further investigation may be necessary into the microbial culture and activity and its impact on membrane fouling propensity to elaborate the present research findings of better MBR performance under appropriate mechanical mixing condition.

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