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The performance of immobilized membrane bioreactor with different membrane operation modes

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ABSTRACT

The immobilized membrane bioreactor (IMBR) was investigated for the removal of organic matter and its membrane fouling condition in treating food processing wastewater. The IMBR contains 5,000 mg/L mixed liquor suspended solids with hydraulic retention time of 24 h. The advantages of IMBR include high sludge retention time, improved removal of COD, ammonia nitrogen, and reduced membrane fouling frequency with much less production of soluble microbial products (SMPs). The results showed that the IMBR was an effective organic matter removal system because it achieved 96-97% removal of COD consistently. The concentration of total SMP in the IMBR was measured at 46.4 mg/L which included 24.8 mg/L of protein and 21.6 mg/L of carbohydrate. Steadily, approximately 33% of carbohydrate and 11% of protein were rejected by the microfiltration (MF) membrane. For this reason, it was concluded that carbohydrate poses a more significant impact on membrane fouling through formation of cake/gel layer than protein. Further, various operating conditions during membrane filtration were experimented which included continuous and intermittent filtration, aeration and non-aeration, and with fiber filter (FF) as pretreatment. It was discovered that while adding an additional FF filtration before MF might improve suspended solid retention, SMP was instead discovered to be the major cause of membrane fouling. In addition, aeration in the membrane tank could significantly improve membrane performance by scouring lightly attached particles from the membrane surface.

Keywords: Immobilized membrane bioreactor; Soluble microbial products; Membrane fouling; Protein; Carbohydrate

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1. Introduction

Membrane bioreactor (MBR) has been widely applied to wastewater treatment for decades. MBRs offer stable and better effluent quality, high organic removal efficiency, simple operation, and maintenance. However, membrane fouling and flux decline have remained as the major concerns of MBR system. In order to enhance the membrane process performance, Wu et al. [1] proposed a mixed filtration mode that was able to reduce mixed liquor suspended solids (MLSS) accumulation and fouling propensity on the membrane. In order to determine the optimized flux for better fouling control, Navaratna and Jegatheesan [2] evaluated the critical flux values by using prolonged flux-step method. Continuous and intermittent aeration were also employed to investigate the potential of membrane fouling reduction [3,4]. For instance, the relaxation refers to the periodical rest of the membrane filtration during which the foulants on the membrane surface could be more effectively scoured by aeration. Pore blocking is referred to irreversible fouling condition caused by inner channel blocking. This irreversible fouling often requires chemical reagents to restore membrane permeate flux.

It was reported that high level of biomass in conventional MBR would lead to faster membrane fouling [5,6]. To membrane operation efficiency, immobilized biological process (IBP) has been developed as an alternative to conventional MBR system. IBP offers many advantages over conventional biological treatment process such as higher sludge retention time (SRT), lower solids washout, and simple operation [7,8]. Recent studies indicated that IBP could achieve satisfactorily carbon and nitrogen removal simultaneously [9-11] and improved removal of micro-organic contaminants such as amine, phenol, and antibiotics [12–14]. Ng et al. [15] proposed bio-ball IBP that could effectively increase 25-30% membrane filtration flux and lower soluble microbial products (SMPs) production which were reported to be the major cause of membrane fouling than conventional activated sludge process. Membrane processes were coupled with IBP as moving bed biofilm reactor and bio-entrapped membrane reactor (BEMR) to combat membrane fouling [10,16]. Ng et al. [16] reported that the BEMR significantly improved and extended membrane filtration time by 39 days from conventional membrane reactor. As a result, the BEMR system could reduce operation and maintenance cost from less frequent chemical cleaning.

SMPs are the substances produced and released into the reactor by micro-organisms, which contribute to some soluble organic concentration in the effluent [17,18]. In addition, SMP had been recognized as the main cause to the membrane fouling. Previous researches proved that SMP can accumulate on the membranes to form cake/gel layer or penetrate into membrane pores and cause in-pore clogging [19-21]. Carbohydrates and proteins are two major components of SMP [15-17,19,20]. Studies of SMP characteristics had been investigated both in conventional MBR and immobilized membrane bioreactor (IMBR) systems [15,16,22,23], and SMP control through adjustments of operation conditions were also studied [16,24]. However, the species and the impact of these contents on membrane fouling remained unclear. The role of SMP in membrane fouling is rather complicated because of the divert sizes, molecular weight, hydrophobicity, and the operating conditions of the system [16,19,20,25]. Fenu et al. [26] established a model for SMPs to quantify the membrane fouling in dynamic environment, but the correlation between SMPs and fouling rates in full-scale MBR was still not well defined.

In this work, IMBR was applied for treatment of food processing wastewater. The IBP was constructed in a rectangular tank with parallel flat plates installed where activate sludge was immobilized at MLSS of 5,000 mg/L. Microfiltration (MF) membrane module was coupled with IBP to form an in-series membrane bioreactor system. The main goal of this research was to present the capability of IMBR on removing soluble organics and improving membrane operation.

2. Materials and methods

2.1. IMBR experimental set-up

The experimental setup is illustrated in Fig. 1. Fig. 1(a) shows the IMBR (IBP+membrane) with a working volume of 50 L. The membrane selected for this system was polyethylene (PE) flat sheet MF membrane, which was installed in a compartment immediately after the biological treatment. Fig. 1(b) illustrates the pretreatment of fiber filter (FF, TRIWIN WATER-TEC Co-MB-01) installed before MF. The IMBR was packed with the immobilized activated sludge on flat plates and the plate is made of polyvinyl chloride with size of $29 \text{ cm} \times 29 \text{ cm}$. The immobilized activated sludge flat plate was prepared by following the procedure of Yang et al. [8] and Ng et al. [15,16] and the characteristic of the IBP followed the previous work at Wang et al. [11]. A total of eight plates (each plate accounts for 5,000 mg/L of MLSS were prepared and installed in the biochamber (44 L) with packing ratio of 18.4%. The influent wastewater was taken from



Fig. 1. Schematic diagram of the experiment (a) without FF and (b) with FF.

treatment plant of the food processing factory. Influent was fed into the biochamber by a peristaltic pump and air flow of 5 L/min was supplied from the bottom of the biochamber by an air compressor. Air flow rate was controlled by a flow meter to maintain aerobic condition for the IBP system. Dissolved oxygen (DO) was maintained at concentration of 7–8 mg/L. The experiment was carried out with 24 h hydraulic retention time (HRT) and at temperature 25 ± 1 °C. The IBP system was operated for two weeks to ensure that the system reached steady-state condition, i.e. when the COD removal efficiency stays constant (>90%) [14]. Effluent from IBP tank was collected and pumped into the MF chamber with a constant flux of $40 \text{ L/m}^2\text{ h}$. The COD of food processing wastewater as influent was at the range of 800-1,300 mg/L. The suspended solids (SS) in the influent and effluent were measured at 66–210 and 16-66 mg/L, respectively. The samples were taken in the influent tank, IBP effluent tank, membrane tank, and membrane permeate reservoir.

2.2. MF membrane process

The PE flat sheet MF membrane used in this study for filtration process had an effective filtration area of 0.1 m^2 and nominal pore size of $0.4 \mu\text{m}$. The membrane permeate was withdrawn through the MF

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membrane by a suction pump. An average flux of $40 L/m^2 h$ was maintained and the evolution of the transmembrane pressure (TMP) was monitored. Membrane filtration was performed without backwash operation; however, the membrane modules were taken out for chemical cleaning at the end of membrane filtration run. Chemical cleaning was performed by soaking the membrane in solution of sodium hypochlorite (NaOCl) for 2h. Different operation modes of the MF process noted as P1, P2, P3, and P4 were designed. P1 was continuous membrane filtration. P2 was performed with intermittent membrane filtration of 9 min on and 1 min off. P3 was similar to P2 except this time, aeration was also applied on the MF system to reduce membrane fouling [3,4]. P4 was operated with FF as the pretreatment of MF. The effluent of IBP was collected prior to the FF in order to eliminate the SS from entering the IBP system. P1 to P4 operation modes were tested for their ability to reduce membrane fouling to increase membrane lifetime and thus decrease the operating costs. The membrane operation would be terminated after 2h of membrane filtration regardless of the recommended TMP limiting value, 45 kPa.

2.3. Analytical methods

Both the influent and effluent samples from IBP tank and MF were analyzed for COD and SMP compositions. DO and temperature in the reactor were recorded daily with a potable DO/pH meter (HACH HQ20). COD was measured by HACH closed reflux colorimetric method by spectrophotometer (HACH DR 2800). The samples were heated for 2 h with potassium dichromate (K₂Cr₂O₇) then measured at 620 nm with the detection range of 20-1,500 mg/L. SMP was analyzed for total protein and total carbohydrate contents which were regarded as the most important fraction of SMP materials. Total protein contents were determined according to the modified Lowry method with bovine serum albumin as protein standard [15,27], and the modified Anthrone method was used for total carbohydrate contents analysis with glucose as carbohydrate standard [15,28].

3. Results and discussion

3.1. Performance of IMBR on COD removal

The influent wastewater containing 800–1,300 mg/ L of COD was fed into the IMBR. Before membrane system was installed, a start-up period of 10 days was operated in order to assure that the bioactivity of the immobilized biomass in IMBR was in steady-state



Fig. 2. COD removal efficiencies of IMBR system (wastewater from food processing industry as influent).

condition. As illustrated in Fig. 2, IBP and IMBP removed up to 95% of COD under the studied 24 h HRT. The results indicated that there is no apparent benefit in using MF for COD removal since MF is primarily responsible for SS removal. The application of MF ($0.4 \mu m$) coupled with IBP increased less than 20% COD removal efficiency in the secondary effluent. If the organics are parts of the colloidal fractions, higher COD removal might be observed from MF since MF acts as the major component for SS retention. However, since IBP does not contain high colloids, MF in this case was not effective in improving COD removal. IBP coupled membrane system could achieve more than 90% COD removal consistently.

3.2. Membrane fouling under different operation conditions and filtration modes

TMP fluctuation was monitored as the main indicator of membrane fouling as illustrated in Fig. 3. IBP effluents were the feeding water to MF process. Continuous and intermittent membrane filtration (nine min filtration with one min relaxation) were compared and depicted in Fig. 3(a). As shown in Fig. 3(a), TMP increased to 45 kPa for continuous filtration (P1) within 40 min in all three cycles, while intermittent filtration (P2) extends membrane operation period to 50–70 min. The experimental results indicated that the intermittent filtration has a noticeable improvement on membrane fouling control (up to 50% improvement on operating period). This finding agrees with some of the previously published research paper



Fig. 3. Evolution of MF TMP at constant flux of 40 LMH. (a) Without aeration and (b) continuous aeration.

which also indicated positive impact from intermittent filtration pattern [1,2,29].

Wu et al. [1] proposed a mixed filtration mode (high and low filtration flux) and discovered that mixed mode was in fact effective at inhibiting cake layer formation and reducing membrane fouling. Further, it also reported that the hydraulic resistance of continuous filtration was about 4–7 times larger than that of intermittent filtration mode. The relaxation period provided an opportunity for aeration bubbles to alleviate membrane fouling [29]. Navaratna and Jegatheesan [2] reported that intermittent suction was effective in fouling control under low MLSS conditions (4,000–7,000 mg/L) in MBR system. Metzger et al. [30] showed the prolonged operation time of MBR with the assisted of membrane relaxation. Although intermittent membrane operation can reduce membrane fouling, this operating technique may not be economically feasible for large-scale MBR because it not only decreases membrane permeate flux productivity but also increases complexity in membrane operation.

The experimental results also showed the aeration significantly mitigated the fouling in MF process. Fig. 3(b) demonstrated the impact of continuous aeration to membrane operation. The TMP increased 17-26 kPa in two hours of membrane filtration. Intermittent membrane filtration with continuous aeration provides a physical mean to scour the particles away from membrane surface, especially during the relaxation period where zero suction was applied on the membrane surface. Additional, upflow air bubbles generated shear stress near the membrane surface, thus improved membrane operation by removing attached particles from the membrane surface [31,32]. The experimental results are supported by Ueda et al. [3], who reported that uplifting air flow was able to control cake layer formation. Coarse bubble aeration was also suggested to improve membrane performance [33,34]. While continuous aeration offers operational benefits, it brings obvious economical drawback when compared to non-aeration and/or intermittent aeration.

In addition, fiber filtration (FF) was also installed as a pretreatment of MF. However, FF did not improve MF operation as the reactor already contains significantly lower concentration of SS from IBP system (16–66 mg/L). As illustrated in Fig. 3(b), with FF as pretreatment of MF process, 12-18 kPa of TMP rise was observed in 2h of operation indicating that FF had only limited effect on fouling mitigation. This result was expected because concentration of SS in IBP effluent was not likely to cause serious fouling on membrane. The continuous aeration mode also abated the effect of SS on membrane fouling in the external tank. Ng et al. [15] inspected the potential causes of membrane fouling and concluded that SMP played a major role on membrane fouling while SS had negligible impact.

3.3. SMP characteristics

SMP was mainly composed of carbohydrate and protein [15–17,19,20]. Fig. 4 illustrates the averaged concentrations of SMP_C (carbohydrate fractionation) and SMP_P (protein fractionation) determined in effluents. Concentrations of SMP_P and SMP_C were 24.8 and 21.6 mg/L, respectively, indicating that protein was the major component of SMP. Similar SMP



Fig. 4. Carbohydrate and protein contents of SMP.

composition was also reported in previous studies [15,16,19,20]. Total SMP ($SMP_C + SMP_P$) in IBP effluent and MF permeate were also determined as 46.4 and 36.6 mg/L, respectively.

Ng et al. [16] showed similar results of the SMP composition using food and beverage processing wastewater as feed influent. Concentrations of SMP_P and SMP_C in IBP effluents were 22.1 and 14.5 mg/L, respectively. The effluent of SMP_P and SMP_C revealed that the SMP retained by membrane were mostly carbohydrate, while most protein contents in SMP were able to pass through the membrane. Therefore, it is evident that the carbohydrate is the major cause of membrane fouling in SMP. Linear correlation between membrane fouling and colloidal/soluble organic substance was reported by Lesjean et al. [22]. High carbohydrate concentrations will lead to the increase of filtration hydraulic resistance in MBR system. Cake/ gel layer formation on membrane surface and pore clogging are two typical types of membrane fouling. Studies also showed that carbohydrates were the major cause of cake/gel layer formation and main reason for the decline of membrane permeability [35,36]. Yigit et al. [37] also demonstrated similar results that carbohydrate fractions in SMP appeared to contribute more on membrane fouling than protein fractions.

Our previous reports [15,16] have discussed the membrane fouling behavior of IBP system. Longer SRT in the BEMR allows the development of slowgrowing micro-organisms which produce less SMPs. The high DO concentration and complete mixing environment in the IBP system would help to distribute nutrients across the entire IBP plate sheets. Therefore, it would reduce the chances of cell death and producing less biopolymers to membrane fouling.

4. Conclusions

The IMBR was investigated for the removal of organic matter and the reduction of membrane fouling. Under the experimental conditions, IMBR demonstrated consistent 96-97% removal of COD. The concentration of total SMP in the IMBR was 46.4 mg/ L which consisted of 24.8 mg/L of protein and 21.6 mg/L of carbohydrate. MF rejected 33% of carbohydrate and 11% of protein. Therefore, it was postulated that carbohydrate was the main foulant to membrane fouling than protein. The intermittent membrane filtration mode with 9 min on and 1 min off could improve membrane operation period by at least 25% because of fouling reduction. The IMBR system performed well with the use of FF as pretreatment to MF module. Aeration was required to reduce membrane fouling and improve overall membrane performance. Different standardized cycles of MBR relaxation should be suggested in future studies in order to provide more comprehensive membrane fouling experience.

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