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Submerged membrane adsorption hybrid system (SMAHS): process control and optimization of operating parameters

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Abstract

This study is to investigate the effect of operating parameters of submerged membrane adsorption hybrid system (SMAHS) such as preadsorption and powdered activated carbon requirement, aeration, filtration flux; There is an optimum value for each of the operating parameters and they are specific to the wastewater used. The duration and frequency of backwashing are vital parameters for successful long-term operation of a membrane system. The backwash duration and frequency required to remove the reversible component of the foulant layer increased with time during the operation of membrane and is not fixed. A new methodology introduced in this study to decide the appropriate duration and frequency of backwash based transmembrane pressure during the membrane operation. This method led to significant saving on the amount of backwash water and energy requirement.

Keywords: Submerged membrane adsorption hybrid system; Aeration; Preadsorption; Automation; Backwash; Powdered activated carbon; Transmembrane pressure (TMP)

1. Introduction

Membrane separation process is being emerged as an innovative wastewater treatment technology. However, its use at present is limited due to its high cost of installation and its long-term operational difficulty. Membrane fouling is a major obstacle to the successful

operation of the membrane separation process. The membrane processes such as reverse osmosis and nanofiltration can remove most of the pollutants, including dissolved organics, but their operational costs are high because of the high-energy requirements and membrane fouling. Micro or ultrafiltration is a cost-effective option, but they cannot remove dissolved organic matter due to their relatively larger pore sizes [1].

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The membrane fouling can be reduced by operating the membrane process under critical velocity and/or by combining the membrane processes with physicochemical and/or biological processes. In this study, a submerged hollow fiber membrane with powdered activated carbon (PAC) adsorption (adsorption membrane hybrid system) was investigated for the removal of organics from a synthetic wastewater representative of a biologically treated sewage effluent. The main aim of adding PAC to the system was to reduce the direct organic loading to the membrane surface. In this hybrid system, the organics are adsorbed onto the PAC, and the organic-laden PAC is eventually separated by the membrane. In long run, after the growth of microorganisms on PAC surface, the organic would be biodegraded by the microorganisms and thus the PAC can be used for long time. The membrane is also free from fouling (or very little fouling) and thus can be used for long time without cleaning. The membrane was submerged in a tank containing wastewater. A known dose of PAC was added to the tank. An air diffuser was used to keep the PAC in suspension and to provide dissolved oxygen for biological activity. The influent and effluent flows to and from the tank were maintained using pumps. The level of wastewater in the tank was maintained with the help of a level sensor. The membrane fouling was observed in a trans-membrane pressure (TMP) gauge.

This submerged membrane adsorption hybrid system (SMAHS) has many advantages. The PAC can be used for a long period. As adsorbed organics undergo biodegradation, more adsorption sites are created on the PAC surface. The submerged membranes do not become clogged as almost all organics are removed by PAC and the role of membrane is only to retain the PAC and other suspended solids. The energy requirement is very low (as low as 0.2 kw h/m³) and there is no major sludge problem. In this system, dissolved organic compounds which normally can pass through the

MF are preadsorbed onto PAC particles. The PAC together with adsorbed organics is then separated by the membrane filtration process. The previous researches showed that the addition of PAC could (i) provide better physical removal of natural organic matter (NOM) and Synthetic organic compounds (SOCs), (ii) reduce the direct loading of dissolved organic pollutants onto the membrane, and (iii) prevent membrane fouling [2–4]. Seo et al. [5] conducted an experimental study on the biological activated carbon microfiltration system for removing refractory organic matter (or persistent organic pollutants). The results showed that the system could remove 83% of total organic carbon (TOC) with 20 g/L PAC dose for 64 days. Furthermore, Kim et al. [2] found that the system could consistently remove more than 95% TOC with a PAC dose of 40 g/L for 40 days from a synthetic wastewater. It should be noted that this 40 g/L PAC is only added at the start of the experiment and the system has been running without any further addition of PAC. Nevertheless, they indicated that a high concentration of PAC could result in the formation of a cake at the membrane surface, thus reducing the effectiveness of physical cleaning by aeration and increasing the filtration resistance. Moreover, the drawback of PAC membrane system is the initial fouling due to rapid, irreversible adsorption of organic substances onto the membrane surface [6]. Consequently, preadsorption of organics prior to the wastewater passing through the membrane could avoid the initial membrane fouling.

The objective of this study was to study the effect of operating parameters such as filtration flux, initial PAC dose, preadsorption duration, aeration rate, etc.

2. Experimental

The schematic diagram of the submerged hollow fiber microfiltration system is shown in Fig. 1.

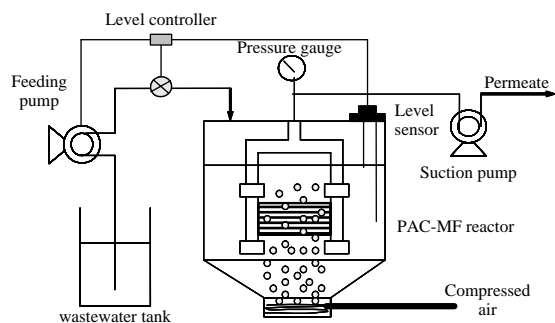


Fig. 1. Experimental set-up of SMAHS.

The details of hollow fiber membrane module are presented in Table 1. The experiments were conducted using a synthetic wastewater which contains persisting (less biodegradable) organic compounds, such as humic acid, tannic acid, lignin, lauryl, acacia gum powder, polysaccharide, beef extract, and peptone. This synthetic wastewater represents the biologically treated sewage effluent [5]. The TOC of the synthetic wastewater was between 3.8 and 4.2 mg/L and COD was 60–65 mg/L. Synthetic wastewater was pumped into the reactor using a feeding pump. The effluent flow rate was controlled by

Table 1
Characteristics of the hollow fiber membrane module

Items	Characteristics
Material	Polyethylene with hydrophilic coating
Nominal pore size	0.1 μm
Outer diameter	0.41 mm
Inner diameter	0.27 mm
No. of fiber	320 (16 \times 20)
Length of fiber	12 cm
Surface area	0.05 m^2
Membrane packing density	9858 m^2/m^3
Membrane manufacturer	Mitsubishi-Rayon, Tokyo, Japan

a suction pump. Level sensor was used to keep the wastewater volume constant in the reactor. The total volume of the membrane reactor is 6 L. A predetermined amount of PAC was added into the tank to adsorb the dissolved organic substances. A pressure gauge was used to measure the transmembrane pressure (TMP) and a soaker hose air diffuser was used to provide bubble aeration. The air bubbling in this system has a dual function of sweeping the membrane surface and mixing the PAC in SMAHS. In the long-term experiments, the air bubbles also helped in supplying oxygen to the microbial mass for biological activity. Since the membrane fouling in this study was mainly due to the organic matter, after each experiment, chemical cleaning was used to clean membrane in order to obtain reproducible results. The procedure of the chemical cleaning used is as follows: (1) the membrane was submerged in 2% citric acid for 2 h to remove iron, aluminium, and manganese attachments from the membrane; (2) the membrane was then submerged in 0.4% NaOCl and 4% NaOH solution for 2 h to remove silica and organic matter.

3. Results and discussion

3.1. Effect of preadsorption duration

The recent study by the authors [7] showed that TMP increase was very high when no adsorbent was used. This suggests that the organic matter is the key factor affecting the membrane fouling. In this study, the effect of the duration of preadsorption was observed over the 10-h experiments at a filtration flux 48 $\text{L}/\text{m}^2 \text{ h}$. The TOC removal efficiency was slightly higher (83.5%) in the case of no preadsorption (Table 2). However, as shown in Table 2, without preadsorption, the TMP development through membrane increased by 50 kPa within 10 h. In the case of no preadsorption, the initial organic loading onto membrane surface was higher and could cause initial

Table 2

TOC removal efficiency of effluent and TMP profile at different durations of preadsorption [filtration flux = 48 L/m² h; total amount of PAC dose = 30 g; aeration rate = 9.6 m³/h m²_(membrane area)]

Duration of preadsorption	TOC removal efficiency (%)	TMP development (kPa)
None	83.5	50
1 h	80.2	22
2 h	79.8	48

irreversible fouling of the membrane. Thus, preadsorption can reduce the membrane fouling by reducing the organic concentration exposed to the membrane surface. One-hour preadsorption was found to be sufficient as it produced much lower TMP development and comparable TOC removal (as compared to 2-h preadsorption). When a longer duration of preadsorption was used (i.e. 2 h), the carbon removed the majority of organics during the first 2 h of operation. It left less adsorption sites on the carbon for further removing the organic matter when the membrane filtration was started at the beginning of third hour. After the 3-h run, the TOC removal efficiency of PAC with 1-h preadsorption declined slower than that with the 2-h preadsorption [8,9]. Therefore, in the subsequent experiments, 1-h preadsorption was used as an operation condition to mitigate membrane fouling phenomenon.

3.2. Effect of aeration rate

Aeration rate was varied from 8 L/min to 20 L/min to study the effect of aeration rates. Table 3 shows the experimental results. In case of aeration, rate range from 8 L/min to 16 L/min, there was an increase in removal efficiency with the increase in aeration rate. However, TMP development did not vary with different aeration rate. The efficiency was not

Table 3

TOC removal efficiency and TMP profile at different aeration rates (filtration flux = 48 L/m² h; PAC dose = 5 g/L; preadsorption = 1 h)

Aeration rate m ³ /h m ² _(membrane area)	TOC removal efficiency (%)	TMP development (kPa)
9.6	80.2	22
14.4	84.6	23
19.2	87.2	24
24.0	87.0	23

improved with any further increase in aeration rate from 16 to 20 L/min.

3.3. Effect of amount of PAC in the tank

The effect of PAC quantity dosage was studied in terms of TOC removal and TMP development (Table 4). There was no noticeable difference of organic removal when PAC dose increased from 2 g/L of tank volume to 10 g/L based on the short-term experimental results presented in this study. However, higher PAC dose could reduce the TMP development which is helpful in preventing membrane clogging (Table 4).

A long-term study [PAC dose = 5 g/L; filtration flux = 12 L/m² h; aeration rate = 14.4 m³/h m²_(membrane area); backwash frequency = 1 day;

Table 4

Effect of PAC dose on the TOC removal efficiency and TMP development (filtration flux = 48 L/m² h; preadsorption = 1 h; aeration rate = 19.2 m³/h m²_(membrane area); backwash frequency = 1 h; backwash duration = 1 min; backwash rate = 120 L/m² h)

PAC amount of tank volume (g/L)	TOC removal efficiency (%)	TMP development (kPa)
2	83.4	19.5
5	84.8	16.3
10	87.5	12.8

backwash duration = 2 min; backwash rate = 2.5 times of filtration flux] conducted by the authors indicated that the TOC removal efficiency could be maintained high at 84% even after 15 days of operation. It is noted that an addition of 5 g/L PAC was only made at the start of the experiments with a daily replacement of 2.5% of total amount PAC. It works out to be carbon dose of 70 g/m³ of treated water. This calculation assumed only 15 days of operation. This experiment was continued running for more than 45 days. The TOC removal efficiency remained high due to the biological degradation of organic matter adsorbed onto PAC.

3.4. Effect of filtration flux

The effect of filtration flux was studied by varying the filtration flux in the range of 24–48 L/m² h. Table 5 shows the relationship of filtration flux with HRT, filtration velocity, and organic matter flux. As expected, the lower filtration flux led to the highest TOC removal and the lowest TMP development.

3.5. Automation of backwash initiation in membrane systems

During the production interval between two successive backwashes, the membrane progressively fouls, leading to a permeability decrease. A backwash is able to remove the reversible component of the foulant layer, minimizing the

severity of these fouling problems. The irreversible foulant component is unaffected by the backwash and must eventually be removed with periodical intensive physical and/or chemical cleaning. However, continued control of the reversible foulant layer positively impacts on the degree of irreversible fouling.

As the interval between backwashing is extended, the cake layer on the membrane compacts and becomes increasingly difficult to remove during the backwash cycle. This effective transition of the foulant layer from reversible to irreversible causes significant problems over the extended operating duration of the membrane system.

If the filtration interval is too long, the progressive increase in irreversible fouling leads to the requirement of the membrane treatment process being stopped for intensive physical and/or chemical cleaning. On the other hand, if the production interval is too short, the wasted permeate unnecessarily used for the backwashing results in a decline in the net productivity of the membrane system. This also increases the energy requirement [10].

A new control system was developed in the study that utilizes a cumulatively increasing transmembrane pressure set-point for the initiation of the backwash. This study initially involved a series of experiments conducted at fixed timed backwashing frequencies and an investigation of the results in terms of fouling and flux recovery. A transmembrane pressure

Table 5

Effect of different filtration flux on the TOC removal efficiency and TMP development (PAC dose = 5 g/L; preadsorption = 1 h; aeration rate = 19.2 m³/h m²_(membrane area); backwash frequency = 1 h; backwash duration = 1 min; backwash rate = 2.5 times of filtration flux; total operation time = 10 h)

Filtration flux (L/m ² h)	HRT (h)	Organic matter flux (mg _(TOC) /h)	TOC removal efficiency (%)	TMP development (kPa)
24	5.0	≈5	89.8	1.7
36	3.75	≈7.5	88.6	4.7
48	2.5	≈10	83.2	16.3

increase of 50 kPa represents the maximum allowable pressure drop before the membrane separation process must be stopped for intensive physical and chemical cleaning. The results of the study found that if a production interval resulted in a transmembrane pressure drop of 3% of the maximum limit of the membrane, the backwash was effective in restoring the permeability of the system, resulting in minimal flux decline and pressure drop over the duration of the experiment. If the production interval was extended past this point, the transmembrane pressure drop increased and the backwash was less able to restore the operational transmembrane pressure and flux resulting in an extended duration of operation at a decreased permeability. The new control system was found to minimize membrane fouling and maximize the productivity and treated water capacity of the membrane system. The new control system developed in this study was also able to automatically optimize the backwash initiation in a membrane filtration system operating under unsteady foulant concentrations and at a nonconstant permeate flux.

Fig. 2 shows a comparison of the results of optimal fixed time periodic backwash (15 min of production followed by a 15 s backwash) was used together with the new control approach.

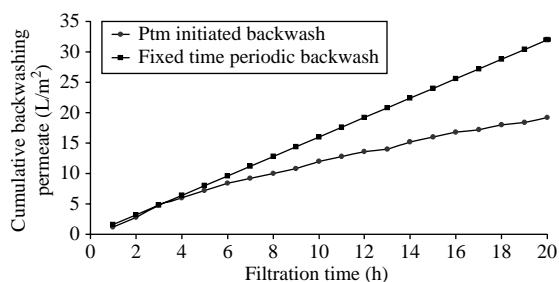


Fig. 2. Cumulative backwashing permeate required for the periodic backwash and the backwash initiation based on a 1.5 kPa P_{TM} increase using the closed loop configuration (synthetic wastewater TOC = 12.35 mg/L, initial permeate flux = 48 L/m² h and backwash flux = 96 L/m² h).

The new approach used a TPM increase of 3% of the maximum allowable value of 50 kPa each cycle to initiate a 15-s backwash.

While both the optimal fixed time backwash approach and the new TPM initiated backwash approach had similar permeate fluxes, the TPM method of backwash initiation resulted in a saving of permeate used for backwash of 40%. Fig. 2 shows a comparison of the cumulative amount of backwashing permeates required for each of the methods.

3.6. Action of backwash duration

A backwash of too short duration results in the failure of the complete removal of the reversible component of the foulant layer. On the other hand, a backwash too long effectively removes the majority or even the entire reversible component of the foulant layer. However, the additional permeate used unnecessarily reduces the productivity of the system in terms of permeate production. This also increases the energy requirement.

Fig. 3 indicates that the permeate flux achievable with the new approach is more or less the same as with the fixed 60-s backwash duration. This resulted in improvements in the productivity in terms of permeate production, as

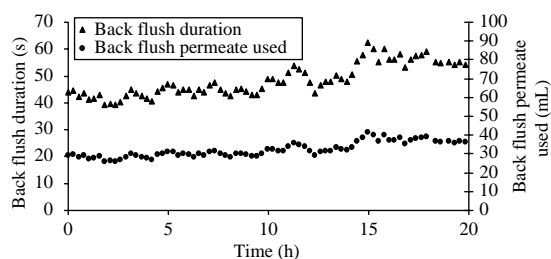


Fig. 3. Backwash duration required to reach the desired TPM profile over a 20-h period using the new control system (synthetic wastewater TOC = 12.35 ppm, initial permeate flux = 0.576 m³/m² d and backwash flux = 1.152 m³/m² d).

the new approach of variable backwash duration requires less time for the backwash. Fig. 3 shows the backwash durations and the amount of permeate required for each backwash. The backwash duration required initially was only about 42 s and increased slowly with the membrane operation. Even after 20 h of operation, it was less than 55 s. Thus, the permeate used for the backwash can be reduced by 25% using the new control system.

4. Conclusions

The preadsorption, PAC dose, aeration rate, and filtration flux had effects both on organic matter removal efficiency and TMP development. Thus, there is a need of optimizing these parameters with the specific water to be treated and the characteristics of PAC chosen. The preadsorption of 1 h prior to the membrane operation was important in mitigating the membrane fouling. The suitable aeration rate, filtration flux, and initial PAC dosing were 16 L/min, <24 L/m² h, and 5 g/L, respectively, for the wastewater used in this study.

The preadsorption of organics onto PAC could help to reduce the membrane fouling and to maintain a consistent permeate flux. The PAC replacement in PAC-MF reactor could stimulate both biological activity and adsorption, as well as optimize the operation of the hybrid system. With the PAC replacement, the system could keep TOC removal efficiency over 90% after 15 days run.

The duration of the backwash is a vital parameter for successful long-term operation of a membrane system. The backwash duration required to remove the reversible component of the foulant layer increased with time during the operation of the membrane and is not fixed. Using a backwash after 15 min of permeate production, the backwash duration required increased linearly from 44 to 55 s over the 20 h

of the experiment. This reduced the required amount of permeate for backwash by 25%. The organic matter removal was consistently high, in the range of 80–85%, irrespective of the variation in backwash duration.

The backwash frequency is a vital parameter for successful long-term operation of a membrane system. Under the experimental conditions used in this study, a 40% reduction in backwash water by automating the backwash initiation based on TMP control. Thus, with a selection of an optimum PAC dose and operational parameters such as filtration rate, PAC replacement rate, backwashing frequency, and duration, the adsorption membrane hybrid system has significant potential for long-term application in wastewater treatment for reuse.

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