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Membrane Fouling Control in an Anaerobic Membrane Bioreactor Coupled with Online Ultrasound Equipment for Digestion of Waste Activated Sludge

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In this study, an online ultrasonic equipment was employed to control membrane fouling in an anaerobic membrane bioreactor for waste activated sludge digestion. Four groups of ultrasound parameters were tested for the performance of membrane fouling control. The ultrasound power intensity of 0.18 W/cm^2 and timing of 3 min/h were considered to be optimal since that membrane fouling could be effectively controlled and no membrane damage was observed. The results of the analysis on the membrane fouling contributions and the scanning electron microscope observation of the fouled membrane indicated that the cake layer, which was the dominant membrane fouling, could be controlled effectively by ultrasound.

Keywords anaerobic membrane bioreactor; cake layer; membrane fouling; ultrasound; waste activated sludge

INTRODUCTION

In contrast to high strength soluble wastewater currently treated effectively with a variety of high-rate anaerobic reactors, an anaerobic membrane bioreactor (AnMBR) may particularly suit high strength particulate wastewater such as municipal sludge usually treated with conventional anaerobic digesters (1). For conventional anaerobic sludge digesters, the solids retention time (SRT) is identical to the hydraulic retention time (HRT). It results in large reactor volumes, since a long SRT (20–30 d) is required for effective volatile solids destruction (2). The AnMBR decouples the SRT from the HRT by almost complete retention of the solid from sludge suspension with membrane separation. The reactor volume of an AnMBR can be designed smaller and further degradation of the volatile solid may be achieved with a long SRT.

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However, membrane fouling is the main drawback of AnMBR, especially for the filtration of high strength particulate suspension. Membrane fouling is the result of adsorption of organic matter, precipitation of inorganic matter, and adhesion of microbial cells to the membrane surface (3). In general, membrane fouling in the AnMBR can follow the general two-pronged approaches aimed at

- reducing the rate of fouling, and
- cleaning a fouled membrane with a chemical or a physical method (1,4).

A cross-flow velocity of larger than 1.5 m/s should be maintained to reduce the rate of fouling in AnMBR with external membrane units (5). However, Padmasiri et al. demonstrated that an increase in cross-flow velocity resulted in poor anaerobic digestion performance (6). And also, the high energy consumption contributed to the cross-flow velocity must be taken into account. During physical cleaning such as backwashing used extensively in the current research, the filtration process must be shut down frequently to recover the membrane permeability for the effective operation of a membrane treatment plant, increasing the complexity of the membrane filtration process. Chemical cleaning requires the membrane to be taken offline. The disposal of contaminated water results in increased cost (4,7).

Ultrasonic cleaning technology has been developed for a long time. It has also been proved effective for the enhancement of membrane filtration by some researchers (8–11). In general, cavitation and acoustic streaming induced by ultrasonic waves were regarded as the mechanism for preventing the formation of a filter cake and enhancing membrane filtration rates (12). Although most of the present research focuses on membrane filtration of simple matter for a short term, online ultrasonic cleaning should be expected to be an alternative method for membrane fouling control during the long-term operation of AnMBR.

Our previous study has indicated that the membrane fouling could be controlled in an AnMBR coupled with ultrasound equipment where synthetic soluble wastewater was treated and mixed liquor suspended solids (MLSS) was below 6 g/L (13). Considering the practical application of AnMBR discussed above, the membrane fouling controlled in an AnMBR coupled with ultrasound equipment (US-AnMBR) when applied for digestion of waste activated sludge (WAS) was investigated in this study.

MATERIALS AND METHODS

Experiment Setup

The US-AnMBR system (Fig. 1) was composed of an 8 L volume of up-flow anaerobic bioreactor (4 L of upper sediment tank used to retain the suspended sludge decreasing the MLSS concentration into the membrane module), a stabilization tank for avoiding flow shock, and an external hollow fiber polythene membrane module (nominal pore size: 0.4 μm ; filtration area: 0.05 m^2 , Mitsubishi Rayon, Japan) embedded in an ultrasound cleaning bath (20 L \times 10 W \times 50 H cm³). Six piezo electric ultrasonic transducers were attached on the side wall of the bath. They were linked to an ultrasound generator with a frequency of 28 kHz and an adjustable power output of 0–300 W (JXD-03, Jinxing Co., Ltd, China). The distance between the ultrasonic transducer and the center of the membrane module was 3.5 cm. Mixed liquor was pumped from the stabilization tank into the membrane module and went back to the tank forming a cross-flow filtration. Mesophilic temperature 37°C was maintained through hot water recirculation. Another AnMBR without ultrasound was also built and operated in parallel. A low cross-flow velocity of 1.0 m/s was controlled. For the long-term operating of AnMBR, the continuous use of ultrasound is undesirable in terms of energy consumption. Therefore, ultrasound introduced into the AnMBR was irradiated intermittently in this study.

Inoculum and Feedstock

Two reactors were inoculated with anaerobic sludge obtained from A²O process at the Gao Bei Dian wastewater treatment plant in Beijing. The feedstock for systems was waste activated sludge collected from the secondary sedimentation tank of Qing He wastewater treatment plant in Beijing. It was prefiltered through a 1 mm screen to prevent clogging of the rough particle and then stored at 4°C before adding to the reactor. Two reactors were operated without the membrane filtration for three days. The temperature was increased gradually up to and maintained at 37°C.

Analytical Methods

Total solids (TS) and volatile solids (VS) in the reactor were determined according to the Standard Methods (14). The performance of WAS digestion was evaluated by the average efficiency of VS destruction for each operating condition.

To characterize membrane fouling, the total membrane filtration resistance (R_t , [m⁻¹]) were evaluated daily. Based on Darcy's law, it was calculated based on Eq. (1).

$$R_t = \frac{\text{TMP}}{\mu J} \quad (1)$$

where μ is the viscosity of the permeate estimated using the viscosity of pure water under the given temperature [Pa · s], TMP is the transmembrane pressure measured with a mercury manometer [Pa], J and is the permeate flux through the membrane [m³/m² · s].

At the end of Run 4 and Run 5, contributions of membrane filtration resistance were analyzed using the resistance-in-series model which was applied in many studies (3,15,16). According to this model, the total membrane filtration resistance (R_t) was separated into the intrinsic membrane resistance (R_m), the cake layer resistance (R_c),

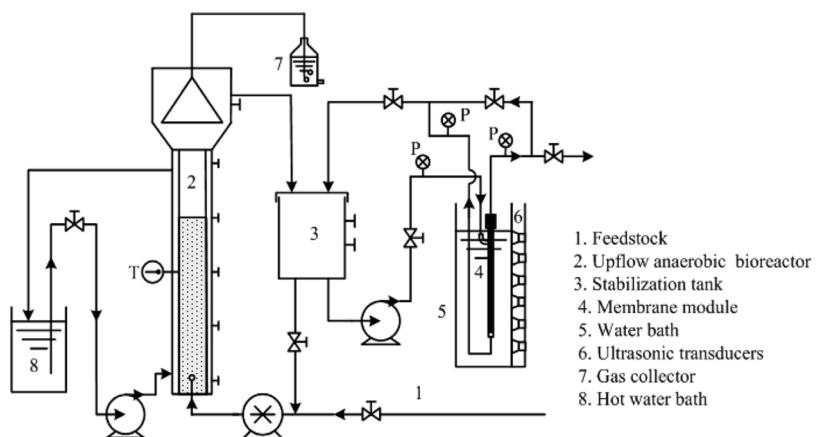


FIG. 1. Schematic diagram of US-AnMBR.

and the fouling resistance (R_f) (Eq. (2)). In this study, the fouling resistance was further divided into the organic fouling resistance (R_{orf}), inorganic fouling resistance (R_{inf}), and irreversible fouling resistance (R_{ir}) (Eq. (3)). The total filtration resistance was calculated with the mixed liquor of the reactor by Eq. (1). The intrinsic membrane resistance (R_m) was calculated with the initial clean water flux and TMP. The cake layer resistance (R_c), defined as the membrane fouling resistance which was able to be removed by water cleaning, was calculated by Eq. (4), where R_w was the membrane resistance with clean water after water cleaning. Then, the organic fouling resistance (R_{orf}) and inorganic fouling resistance (R_{inf}), defined as the membrane fouling resistance being able to be removed by alkaline cleaning (0.5% NaClO for 2 h) and subsequent acid cleaning (pH = 1.5 HCl for 2 h) respectively, was calculated by Eq. (5) and (6), where R_{alk} and R_{acid} were the membrane filtration resistance with clean water after alkaline and acid cleaning. Finally, the irreversible fouling resistance (R_{ir}) was the residual fouling resistance unable to be removed and calculated by Eq. (7).

$$R_t = R_m + R_c + R_f \quad (2)$$

$$R_f = R_{orf} + R_{inf} + R_{ir} \quad (3)$$

$$R_c = R_t - R_w \quad (4)$$

$$R_{orf} = R_w - R_{alk} \quad (5)$$

$$R_{inf} = R_{alk} - R_{acid} \quad (6)$$

$$R_{ir} = R_{acid} - R_m \quad (7)$$

Fouled membranes before and after cleaning were observed by scanning electron microscope (SEM, QUANTA 200, FEI, Holland). For the SEM analysis, samples were processed by gold sputtering on the surface with a sputter coater (SCD 005, BAL-TEC, Switzerland) and then scanned at various magnifications.

RESULTS AND DISCUSSION

Performance of WAS Digestion

During 77 days operation, the efficiency of the VS destruction in US-AnMBR achieved more than 52.1% with a short HRT of 6 days and a long SRT of 80 days, which was improved compared with that without ultrasound except Run 3 (as shown in Table 1). According to the digestion results, the performance of WAS digestion was enhanced for most cases by adopting ultrasound. For the sludge digestion process, the hydrolysis is the rate-limiting step. Ultrasound disintegration technology has been widely applied for cell lysis as a pre-treatment to improve anaerobic sludge digestion. It was assumed that the ultrasound adopted in this study could enhance the sludge disintegration.

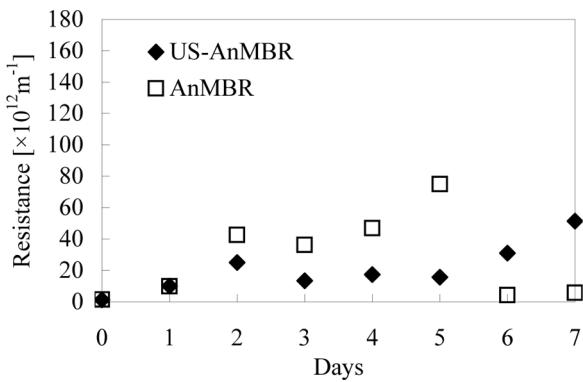
Performance of Membrane

At higher MLSS concentration condition (up to 32.8 g/L), the membrane in the system without ultrasound suffered heavier fouling compared with that in our previous study (13). It must be cleaned manually with pure water once the permeate flux is reduced below the designed value (as shown in Fig. 2, 3, 4, and 5). In the US-AnMBR, ultrasonic parameters, including the power intensity, determined simply from the output power of the ultrasound equipment per unit area of the transducer surface and the irradiation time for a period were adjusted according to the performance of the membrane filtration.

During operation of Run 1, the ultrasound was irradiated with a power intensity of 0.12 W/cm² and a timing of 5 min/h (operate 5 minutes for each hour) in the US-AnMBR. On day 1–5, the membrane resistance rapidly increased from 1.5×10^{12} m⁻¹ to 75.0×10^{12} m⁻¹ in the AnMBR, requiring offline membrane cleaning on the 6th day (Fig. 2). For the US-AnMBR, the filtration resistance was controlled at lower level in the range of $1.3\text{--}25.0 \times 10^{12}$ m⁻¹. However, the rapid increase of filtration resistance occurred on the 6th day and the cleaning procedure must be carried out on the 7th day for the US-AnMBR. Since the offline cleaning of the membrane in the US-AnMBR was almost as frequent as that without ultrasound, the

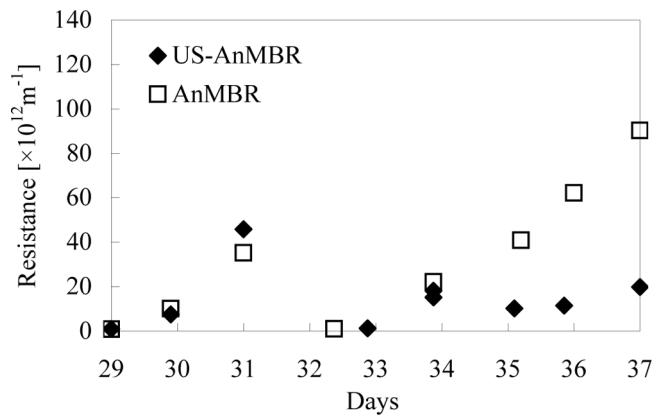
TABLE 1
Operation condition for the system and average VS destruction

Run	Days	Ultrasound parameters		MLSS concentration (g/L)		Average VS destruction (%)	
		Power intensity (W/cm ²)	Timing	US-AnMBR	AnMBR	US-AnMBR	AnMBR
1	1–7	0.12	5 min/h	5.2–10.7	8.3~14.5	59.7	54.2
2	8–28	0.18	5 min/h	5.0~16.3	9.7~16.5	72.8	54.9
3	29–37	0.18	1.5 min/0.5 h	16.5~29.8	16.7~28.9	57.9	63.2
4	38–62	0.18	3 min/h	21.4~33.1	18.3~32.8	52.1	44.2
5	63–77	0.18	3 min/h	0.04~12.4	0.07~12.7	55.0	49.7

FIG. 2. Membrane filtration resistance in Run 1 (0.12 W/cm^2 , 5 min/h).

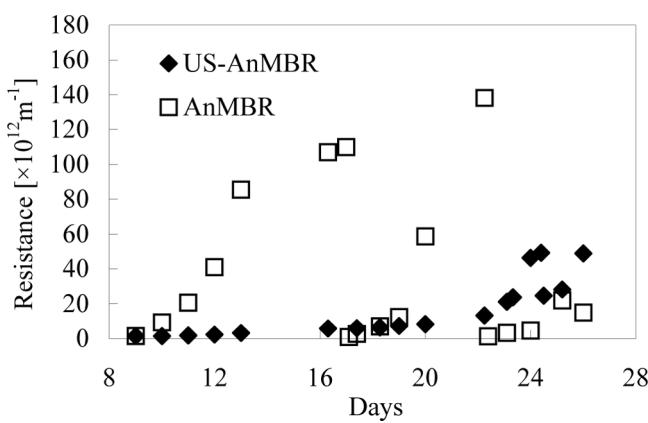
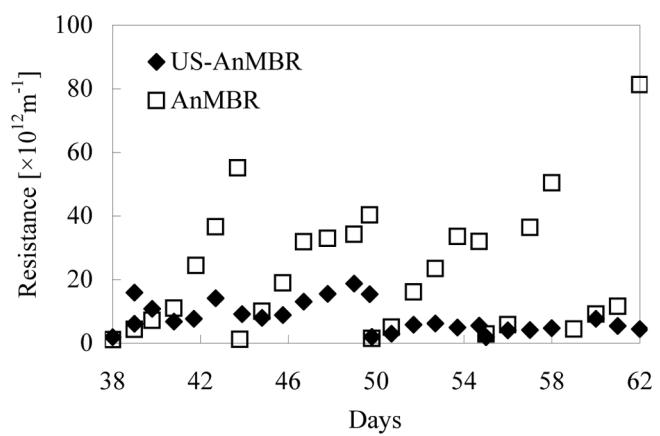
ultrasound condition in Run 1 must be adjusted. In general, an increase in power intensity will improve the membrane cleaning effect and provide the better flux (9,12). In the subsequent experiment, the ultrasound intensity of 0.12 W/cm^2 , lower than that adopted in most research, was increased up to 0.18 W/cm^2 .

When the power intensity of 0.18 W/cm^2 and the timing of 5 min/h were adopted in the US-AnMBR, the filtration resistance was controlled at below $8.2 \times 10^{12} \text{ m}^{-1}$ without any additional cleaning in the US-AnMBR during the previous 14 days while offline water cleaning was almost performed per week and the highest resistance of $138.2 \times 10^{12} \text{ m}^{-1}$ was obtained in the AnMBR (Fig. 3). However, the sludge floc appeared in the permeate from the US-AnMBR after the 22nd day, indicating that the membrane damage might occur. The membrane flux declined due to the reason that the permeate outlet of the membrane module was probably clogged with the sludge floc. Damage on the membrane has also been discovered in some studies, which was dependent on the membrane material, ultrasound power parameters, cross-flow velocity, the housing of the membrane module, and other factors (17,18). For

FIG. 4. Membrane filtration resistance in Run 3 (18 W/cm^2 , 1.5 min/0.5 h).

the long-term operation, the control strategy should be adjusted to avoid membrane damage, even though the membrane fouling control effect was significant under this condition. The approach was to reduce the ultrasound irradiation energy or to deliberately form a certain extent of fouling layer taken as a protective layer towards ultrasound irradiation.

Membranes were replaced at the beginning of Run 3 and the ultrasound irradiation time was reduced to 1.5 min/0.5 h. Moreover, instead of the beginning of the filtration process, the ultrasound irradiation started from the 34th day when a protective fouling formed characterized with a filtration resistance of $18.1 \times 10^{12} \text{ m}^{-1}$ (Fig. 4). Although the fouling control effect could be observed, the resistance increased up to $20 \times 10^{12} \text{ m}^{-1}$ in the US-AnMBR on the 37th day. The membrane was probably incapable of affording the long-term effective filtration without additional cleaning. The irradiation time of 1.5 minutes for a half hour was too short to effectively remove the fouling. The timing was adjusted as 3 min/h for the following operation. The ultrasound energy input remained constant.

FIG. 3. Membrane filtration resistance in Run 2 (0.18 W/cm^2 , 5 min/h).FIG. 5. Membrane filtration resistance in Run 4 (0.18 W/cm^2 , 3 min/h).

In Run 4 (day 38–62), as the operating strategy used on Run 3, the ultrasound was irradiated from the second day of this Run when a filtration resistance of $15.8 \times 10^{12} \text{ m}^{-1}$ was obtained. The substantial reduction of the filtration resistance achieved $9.8 \times 10^{12} \text{ m}^{-1}$ after the first ultrasound irradiation period, while $2.9 \times 10^{12} \text{ m}^{-1}$ was reduced at the former ultrasound condition (1.5 m/0.5 h). It indicated that the timing of 3 min/h was more effective on the fouling removal than 1.5 min/0.5 h with the same power intensity. On days 38–50, offline water cleaning was carried out per week in the AnMBR. It was observed that the membrane fouling could be controlled in the US-AnMBR (Fig. 5). However, similar to that in Run 3, the filtration resistance in the US-AnMBR increased gradually and achieved at a high value of $18.7 \times 10^{12} \text{ m}^{-1}$ on the 49th day. To be more effective on membrane fouling control, the fouling layer should be prevented with the ultrasound from the beginning of the filtration process. The fouling layer formed on the surface of the membrane possibly led to increase in attraction of biomass and solute on to the fouling layer on membrane surface. After the 51st day, the ultrasound was irradiated from the beginning of the operation with a cleaned membrane. From the results of days 51–62, the membrane fouling was controlled effectively in that the filtration resistance was kept below at $7.6 \times 10^{12} \text{ m}^{-1}$. In the AnMBR, the high resistance of $81.2 \times 10^{12} \text{ m}^{-1}$ was obtained and the frequency of water cleaning was reduced to once for every 4 days. In addition to the significant effect of fouling control, the membrane damage under this ultrasound irradiation condition did not occur in the US-AnMBR.

Due to excellent performance of membrane fouling control in Run 4, the same ultrasound parameters were used in Run 5 (Day 63–77). As anaerobic sludge digestion proceeded, suspended solids, especially inert matter, accumulated in reactors and thus partially digested sludge was discharged at the beginning of Run 5. The low MLSS concentration condition resulted in a better performance of the membrane filtration. In contrast to previous performance, the tendency of membrane fouling mitigated in the AnMBR until the 70th day when the membrane filtration resistance increased rapidly when the MLSS up to 8.9 g/L (Fig. 6). An increase in MLSS concentration would generally aggravate membrane fouling via increasing the opportunity of cake layer formation. Strohwald and Ross discovered a rapid decline in flux for $\text{MLSS} > 20 \text{ mg/L}$ (5). The results of Run 5 showed that when the MLSS concentration increased from 0.04 g/L to 12.4 g/L, the filtration resistance with the ultrasound irradiation maintained at a stable state, namely at below of $3.9 \times 10^{12} \text{ m}^{-1}$, hardly affected by the MLSS concentration. Although ultrasound seemed to be unnecessary to be used for membrane fouling control before the 70th day, the disparity of membrane filtration resistance enlarged gradually

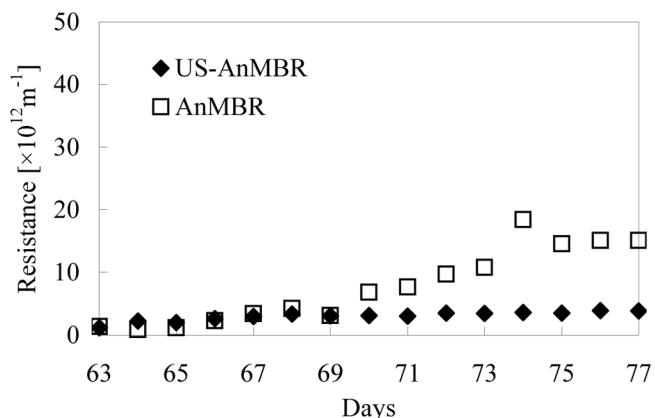


FIG. 6. Membrane filtration resistance in Run 5 (0.18 W/cm^2 , 3 min/h).

between the two systems and the effect of ultrasound appeared greatly as the filtration proceeding.

Considering the performance of membrane fouling control and avoidance of membrane damage, the ultrasound power intensity of 0.18 W/cm^2 and the timing of 3 min/h was the optimal condition to control membrane fouling in the AnMBR in this study. To keep resistance at a considerably low level, ultrasound should be imposed on the membrane at the beginning of filtration as long as no membrane damage occurred. For other circumstances, the optimal ultrasound condition obtained in this study might be adjusted since that the performance of fouling control by the ultrasound was affected by characteristics of feed liquid and other operational parameters.

Characteristics of Membrane Fouling Controlled by Ultrasound

According to the analysis on the fouling contributions, the cake layer resistance was up to $80.2 \times 10^{12} \text{ m}^{-1}$ and $13.9 \times 10^{12} \text{ m}^{-1}$ (Fig. 7) at the end of Run 4 and Run 5 respectively in the AnMBR, accounting for 98.8% and 92.0% of the total filtration resistance. It demonstrated that

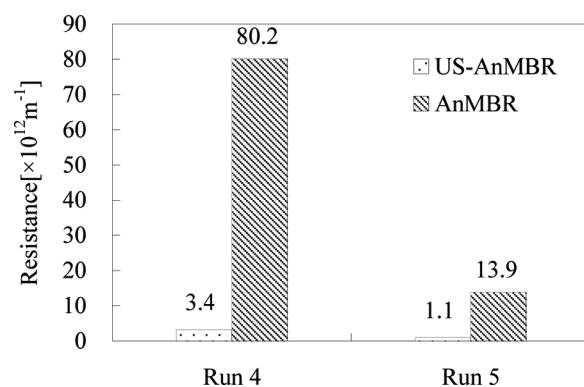


FIG. 7. Cake layer resistance for Run 4 (high SS) and Run 5 (low SS).

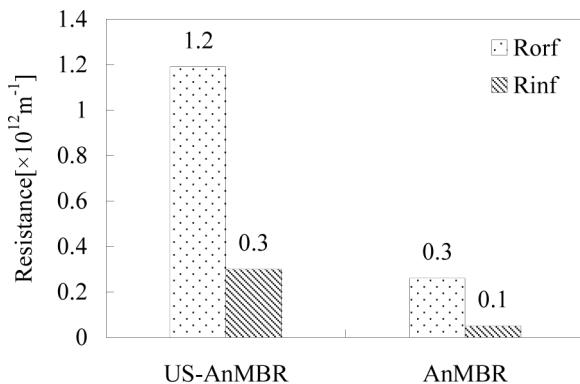


FIG. 8. Organic fouling and inorganic fouling resistance for Run 5.

the membrane fouling was dominated by a cake layer when AnMBR was applied for WAS digestion. The severe cake layer fouling was liable to form under the high strength MLSS condition. Zhang et al. had also observed that membrane fouling was dominated by a loosely attached fouling layer which could be removed by flushing in the AnMBR used for treatment of swine manure (4). Superior to the AnMBR, the low cake layer fouling resistance of $3.4 \times 10^{12} \text{ m}^{-1}$ and $1.7 \times 10^{12} \text{ m}^{-1}$ were obtained at the end of Run 4 and Run 5 respectively in the US-AnMBR, corresponding to the cake layer control efficiency of 95.8% and 92.4% respectively. The cake layer could be controlled effectively by the appropriate ultrasound irradiation. It was also approved by the SEM observation of the fouled membrane (Fig. 9a and 9e). The heavier cake layer covered on the surface of the membrane without ultrasound. The results of the mixed liquor property analysis indicated that the mixed liquor could not be modified by the ultrasound to control fouling. The fouling control by the ultrasound might be through directly preventing the cake layer formation or removing the cake layer.

However, ultrasound did not have much effect on the organic fouling and inorganic fouling control. In Run 4,

there was no obvious difference in the organic and inorganic fouling resistance between the two systems. At low SS condition for Run 5, the membrane even suffered more organic and inorganic fouling in US-AnMBR (Fig. 8) since there was not enough cake layer on the membrane resulting in direct exposure of the membrane against the feed liquid and more organic matter was released into the liquid phase due to the ultrasound. It was also established with the SEM images of the fouled membrane cleaned after water and alkaline cleaning (Fig. 9b, 9c, 9f and 9g). The fouled membrane in the US-AnMBR could be recovered almost completely with the cleaning procedures for Run 4 while the residual fouling resistance was unable to be removed for Run 5 was $0.5 \times 10^{12} \text{ m}^{-1}$ and $0.4 \times 10^{12} \text{ m}^{-1}$ more than that in the AnMBR. The heavier residual fouling for Run 5 could also be observed in Fig. 9d and 9h.

For anaerobic sludge digestion, the low MLSS condition like Run 5 appears rarely. Under high strength MLSS condition, severe cake layer fouling occurred and dominated the total membrane fouling. In spite of less effect on the organic, inorganic, and irreversible fouling control, the ultrasound was considered to be applicable for the membrane filtration enhancement in the AnMBR for the sludge digestion since it controlled the cake layer effectively.

CONCLUSIONS

The conclusions can be summarized as follows:

1. Efficiency of VS destruction in US-AnMBR achieved over 52% with a short HRT of 6 days. It was hardly affected negatively and even improved compared with that without the ultrasound.
2. Intermittent ultrasonic irradiation was able to control the membrane fouling efficiently in AnMBR when applied for WAS digestion. In this study, the power intensity of 0.18 W/cm^2 and timing of 3 min/h was regarded as the optimal ultrasonic condition in consideration of the efficiency of membrane fouling control

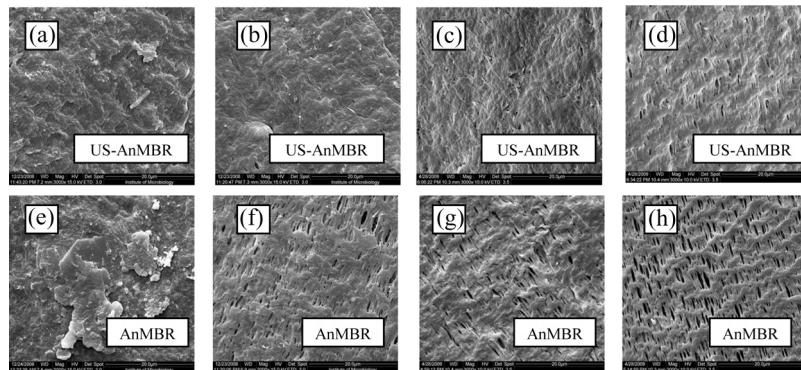


FIG. 9. The SEM observation (scale bar: $20 \mu\text{m}$) of the fouled membrane (a and e), the fouled membrane cleaned with pure water (b and f), NaClO solution (c and g) and HCl solution (d and h) in series for two systems at the end of Run 5.

and the lifetime of the membrane. To be more effective on the membrane fouling control, the ultrasound should be irradiated from the beginning of the filtration process.

3. The major membrane fouling controlled by ultrasound was the cake layer rather than other fouling contributions. During digestion of WAS in the AnMBR, a severe cake layer may form on the membrane surface at high SS condition and dominate the total membrane filtration resistances; therefore, ultrasound is effective and applicable for membrane fouling control.

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