Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2016.1180265

57 (2016) 29201–29211
December

Taylor & Francis
Taylor & Francis Group

Treatment performance and membrane fouling characteristics of inclined-tube anoxic/aerobic membrane bioreactor applied to municipal solid waste leachate

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Received 10 January 2016; Accepted 14 April 2016

ABSTRACT

This study investigated the performance of inclined tube anoxic/aerobic membrane bioreactor (itMBR) treating concentrated municipal solid waste leachate over 550 d at organic loading rates of 4.2–31.8 kg BOD/m^3 d. At optimum loading of 18 kg BOD/m^3 d, the itMBR could achieve high removals of 97% for biochemical oxygen demand, 94% for chemical oxygen demand, and 95% for total Kjeldahl nitrogen. Complete elimination of protein-like substances and partial removal of fulvic and humic acid-like substances were achieved. Major membrane foulants were found to be gel layers containing protein-like and polysaccharide-like substances and Ca-based inorganic scaling. Membrane cleaning using sodium hypochlorite followed by citric acid was found effective for the restoration of transmembrane pressure (TMP) while periodical chemical enhanced back-flushing at every 7–10 d could limit TMP increase rate and sustain the operation in long term.

Keywords: Chemical cleaning; Membrane bioreactor; Membrane fouling; Organic loading; Solid waste leachate

1. Introduction

A municipal solid wastes (MSW) is continuing to be one of main environmental pollutants in most countries. Solid waste transfer station (SWTs) is a common facility for handling MSW management during its transportation from the sources of generation to a final disposal site where leachate and odor pollution could create a serious annoyance condition to the nearby area. In tropical regions, seasonal variation has a great influence on the leachate volume and its characteristics [1]. Physical, chemical, and biological treatment process alone were found to be insufficient for the treatment of landfill leachate [2]. Freshly produced leachate is one of the most polluted wastewater due to its extreme containment of dissolved organic matter

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Presented at the 8th International Conference on Challenges in Environmental Science & Engineering (CESE-2015) 28 September–2 October 2015, Sydney, Australia

(DOM), inorganic macro-components, heavy metals, and xenobiotic organic compounds [3]. The treatments of fresh leachate were successfully reported by different wastewater treatment processes, i.e. expanded granular sludge bed reactor [4], up-flow anaerobic sludge blanket (UASB) reactor [5], etc. Most of them gave high efficiency, but required long hydraulic retention time (HRT) for the operation. In most cases, advanced post treatment processes would be required for polishing of effluent from biological treatment step [6]. Recently, submerged type aerobic membrane bioreactor (MBR) has been proposed for the treatment of leachate from SWTs operated under high organic loading rates (OLRs) of 2–10 kg COD/m^3 d [7] and the system could be successfully applied to remove chemical oxygen demand (COD) up to 97%. However, the MBR should be operated at relatively low permeate flux, i.e. less than 2.4 L/m^2 h in order to sustain the operation at high biomass concentration of more than 40 g/l. It was found that transformation of organic matter presented in leachate to humic-like substances could potentially foul the membrane surface during its rejection in MBR [8].

Novel type membrane bioreactor utilizing inclined tube separation in first stage reactor followed by second stage aerobic reactor has been developed [9]. The system has been successfully applied to the treatment of partially stabilized landfill leachate during long-term operation without sludge wastage [10]. Nevertheless, its application to highly concentrated leachate from SWTs and membrane fouling control strategy for its sustainable operation is still a challenging task and need to be explored. Therefore, prototype pilot-scale inclined tube anoxic/aerobic membrane bioreactor (itMBR) was applied to the treatment of fresh solid waste leachate generated from a SWTs in this study. The research was focusing on the treatment performance and membrane fouling control during increasing of OLR over long-term operation of more than 550 d. Fouling characteristics and chemical cleaning strategies for controlling membrane fouling were investigated in order to find appropriate operating condition for sustaining system operation at high OLRs in long term.

2. Materials and methods

2.1. Pilot scale itMBR operation

Pilot-scale inclined tube anoxic/aerobic membrane bioreactor (itMBR) as the schematic shows in Fig. 1 was installed at a solid waste management facility in Thailand. The system received fresh leachate drained from solid waste collection trucks before transporting MSW to its final disposal. It is comprised of a 4.0 m^3 of anoxic tank with an effective volume of 3.0 m^3 connected to an equal volume of an aerobic tank (effective volume of 3.0 m^3). An inclined tube module (0.15 m tube size, 60° inclination) of 0.45 m depth is installed inside the anoxic tank. The tank was purged with air intermittently at a flow rate of 50 l/min for 2 min followed by 15 min off in order to control dissolved oxygen (DO) level at 0.5 mg/l. In aerobic tank, two hollow fiber membrane modules (STERAPORE SADF™, PVDF material) with a total surface area of 12 m^2 and an average pore size of 0.4 μm are installed for solid–liquid separation. The operation was performed at a constant permeate flux mode. Intermittent membrane filtration was performed at 8 min on and 2 min off mode. The air diffusers are placed at the bottom of tank while providing an air flow rate of 416 L/min and DO concentration was maintained at 2.2–6.9 mg/l.

2.2. Water quality analyses

Three experiments were performed by varying HRT in anoxic and aerobic reactors between 12 and 20 h thus yielding OLR of 4.2–31.8 to the system (Table 1). Recirculation of mixed liquor from the aerobic to anoxic reactor was performed at 67% of feed flow rate. Chemical characteristics of influent and effluent of itMBR were monitored along the experimental period. The analyzed parameters performed according to Standard Methods for the Examination of Water and Wastewater [11] include biochemical oxygen demand (BOD), COD, total Kjeldahl nitrogen (TKN), suspended solid, ammonia nitrogen (NH_3-N), nitrite nitrogen (NO_2-N), nitrate nitrogen (NO_3-N), and total phosphorus (TP). For pH determination, a pH meter (Metrohm, 827pH lab) was used. The metal ions containing in the influent leachate i.e. Na^+, Ca^{2+}, Mg^{2+}, Fe^{3+} were analyzed by the inductive coupled plasma optical emission spectrometer (ICP-OES, Agilent 710-ES). The three-dimensional excitation–emission matrix (EEM) fluorescence technique was used to characterize DOMs in the water samples [8]. The water samples were scanned over excitation/emission wavelengths from 200 to 550 nm by the EEM spectra (Jasco FP-8200). Double distilled water was used to set background excitation/emission reading.

2.3. Membrane cleaning

Stepwise chemical cleanings with acid and alkali chemicals were examined for in order to remove foulants from membrane surface. The two modes of

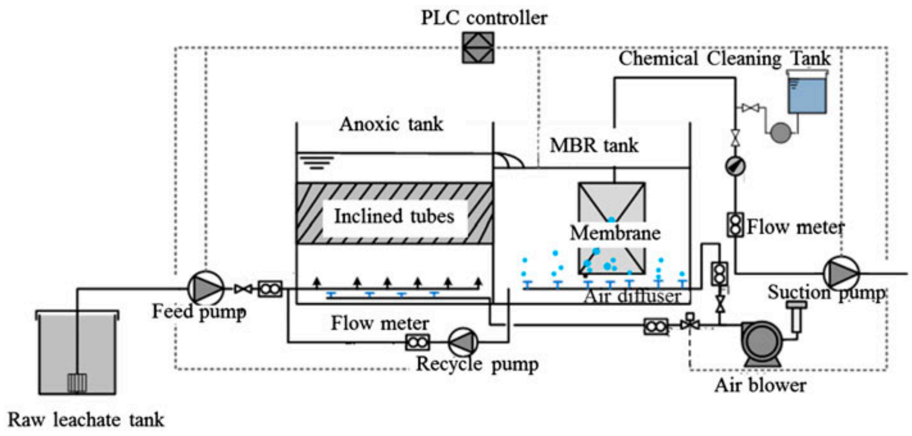

Fig. 1. Schematic of itMBR.

Table 1
Operating conditions of the itMBR

Parameter	Unit	Anoxic			Aerobic (MBR)		
		Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
Operating time	d	71	99	330	71	99	330
HRT	h	20	12	12	20	12	12
Aeration rate	l/min	–	–	–	416	416	416
Recirculation rate	ratio of feed rate	–	–	–	0.67	0.67	0.67
Permeate flux	l/m^2 h	–	–	–	13	21	21
MLSS	g/l	–	–	–	2.2–12.6	8.5–12.2	8.1–17.4
F/M	d^{-1}	–	–	–	0.2–1.6	0.4–0.8	0.5–1.4
OLR	kg BOD/m^3 d	4.2–9.0	10.8–18.0	12.0–31.8	1.4–5.4	3.6–7.5	6.0–15.6
DO	mg/l	0.2–0.8	0.1–0.8	0.3–0.7	4.9–6.4	4.6–6.9	2.2–6.6
Temperature	℃	30–35	30–35	30–35	31–35	31–35	31–35

chemical cleaning used were, i.e. (i) chemical cleaning-in-place (CIP) during which membrane modules were taken out from the itMBR. They were subjected to sequential chemical cleaning in the reagent tanks, and (ii) chemical enhanced backflushing (CEB) during which a cleaning reagent was injected in opposite direction of permeate flow while the membrane modules were still installed in the MBR. In order to achieve long-term steady efficiency, chemical cleaning agents and both CIP/CEB including cleaning frequency, were trialed for the membrane cleaning. Chemical reagents used for membrane cleaning were sodium hypochlorite (NaOCl) and citric acid in all cleaning tests. For the CIP method, membrane cleaning was performed when transmembrane pressure (TMP) was increased above 40 kPa. The membrane modules were immersed in NaOCl solution (0.3% w/v) for 6 h then soaked in a 2% (w/v) citric acid solution for 2 h. For the CEB method, the cleaning was performed using 2% (w/v) citric acid followed by 0.05% (w/v) NaOCl at every 7 or 10 d. The chemical injection rate was 2 l/m^2 of membrane surface area for 30 min followed by a 90 min holding time. It should be noted that the sequence of chemical cleaning in CEB was opposite to that of CIP due to the introduction of chemicals from the permeate side of the membrane module.

2.4. Membrane fouling investigation

For membrane characterization, the fouled membrane was sliced into small pieces and then soaked in 2.0% (v/v) of glutaraldehyde in 0.1 M phosphate buffer, pH 7.2 for 2 h. Thereafter, they were washed with de-mineralized water two times for 10 min and immersed for 1 h in 0.1 M phosphate buffer. This treatment was a fixing step prior to dehydration by ethanol. In order to characterize the foulants on the membrane, a piece of the fouled membrane was immerged into 0.3% (w/v) NaOCl for 6 h which represented as an alkali cleaned membrane sample. Then, the sample was immersed into 2% (w/v) citric acid for 2 h as an acidic cleaned membrane sample. The scanning electron microscopy (SEM, JSM-5410, JEOL) was employed to observe morphology of the membrane surface of all samples. The energy-dispersive spectroscopy (EDS, ISIS 300, Oxford) was used to identify deposited elements. The membrane pieces were dried at 50°C for 12 h for analysis via the Fourier transform infrared (FTIR) spectrometer (Spectrum One, Perkin–Elmer) in transmission mode in order to characterize the functional groups of the organic matters attaching on the membrane surface. Stretching or bending vibrational modes of the bonds of functional groups coupled to an IR excitation yielding the selective resonant absorption.

2.5. Biomass analyses

Biomass concentrations in the itMBR were monitored along the experimental period. Mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solid (MLVSS) were analyzed in aerobic reactor while accumulation of solid mass in terms of total solids (TS) in anoxic reactor was determined. In order to express the membrane fouling potential of mixed liquor in aerobic reactor, soluble microbial product (SMP) and extracellular polysaccharide substance (EPS) were analyzed. MLSS samples from aerobic reactor were centrifuged at 5,000 rpm for 15 min at 4°C. Then, the supernatant was filtered through 0.45 μm membrane filter for SMP analysis. For EPS determination, the solid fraction was mixed in 50 ml of 0.05% (w/v) NaCl and 0.3 ml of 37% (w/v) formaldehyde. Then, 20 ml of 1 N NaOH was added into the mixture. After stirring thoroughly, the mixture was centrifuged at 5,000 rpm, 15 min and the supernatant was filtered through 0.45 μm cellulose acetate membrane. Phenol-sulfuric method was employed for determining the carbohydrate content and Lowry's method was used for protein content of SMP and EPS [12]. Colorimetric measurement was performed using a spectrophotometer for both parameters.

Rheological property MLSS in MBR was determined by rotational viscometer (REDV-E, Brookfiled). The viscosity was measured at a shear rate 100 s^{-1}, at 20°C for 3 min to maintain suspension of the sludge [13].

The data were statistical analyzed using a SPSS 16.0 software (SPSS Corporation). Pearson correlations were used to determine the significance of relationships between viscosity, SMP, MLSS, and OLR variables; and between the SMP and OLR, MLSS. All correlations were considered statistically significance at a 95% confidence interval ($p < 0.05$).

3. Results and discussion

3.1. Treatment performance of itMBR

Fig. 2 shows the variation of influent and effluent characteristics and corresponding biomass concentrations in the anoxic and aerobic reactors during the whole operation period. The feeding leachate was acidic and contained high BOD and COD concentrations of 3,500–15,900 and 4,500–30,400 mg/l, respectively. In term of nitrogen, NH_3-N and TKN concentrations were 95–328 mg/l and 115–395 mg/l. Some major cations such as Ca^{2+} (338–475 mg/L), Mg^{2+} (175–250 mg/L), and Fe^{3+} (55–98 mg/L) were detected in leachate.

Table 2 shows the average chemical characteristics of influent and effluent of itMBR during three experimental runs (Run 1–3) as the OLR was stepwise increased. It was found that while BOD in feeding leachate was increased from 5,100 to 12,180 mg/l, the itMBR produced a relatively stable effluent with average BOD concentrations of 576, 189, and 584 mg/L in Run 1–3 equivalents to their removal efficiencies of 92, 97 and 95%, respectively. The COD removal efficiencies were also in range between 85 and 97%. Slightly lower COD removal than BOD removal was due to the presence of recalcitrant organic compounds in leachate exhibited by low BOD/COD ratio of 0.6 (Run 1 and 3). Notable improvement of organic removals was observed in Run 2 when the feeding leachate contained more biodegradable organic fraction (BOD/COD of 0.7). These results suggest that the itMBR system could handle the OLR up to 18.0 kg BOD/m^3 d by maintaining more than 90% removal efficiencies of biodegradable organic substances in most operating conditions.

Considering the removal efficiencies of each treatment unit, high fluctuation of inflow BOD and COD concentrations resulted in relatively unstable organic removal in first stage anoxic reactor. The average COD removal in the anoxic tank was 32, 44, and 49% during Run 1, 2, and 3, respectively. This lower removal efficiency was possibly due to short HRT maintained in anoxic reactor (12 h). Previous study has achieved 93% COD removal in UASB reactor operated at longer HRT of 2–3 d when applied to the treatment of fresh leachate at the same OLR range [5]. Nevertheless, fluctuation of organic concentrations in the effluent from the anoxic tank could be redeemed by the second stage aerobic reactor, which was effective in organic removals by giving the average COD

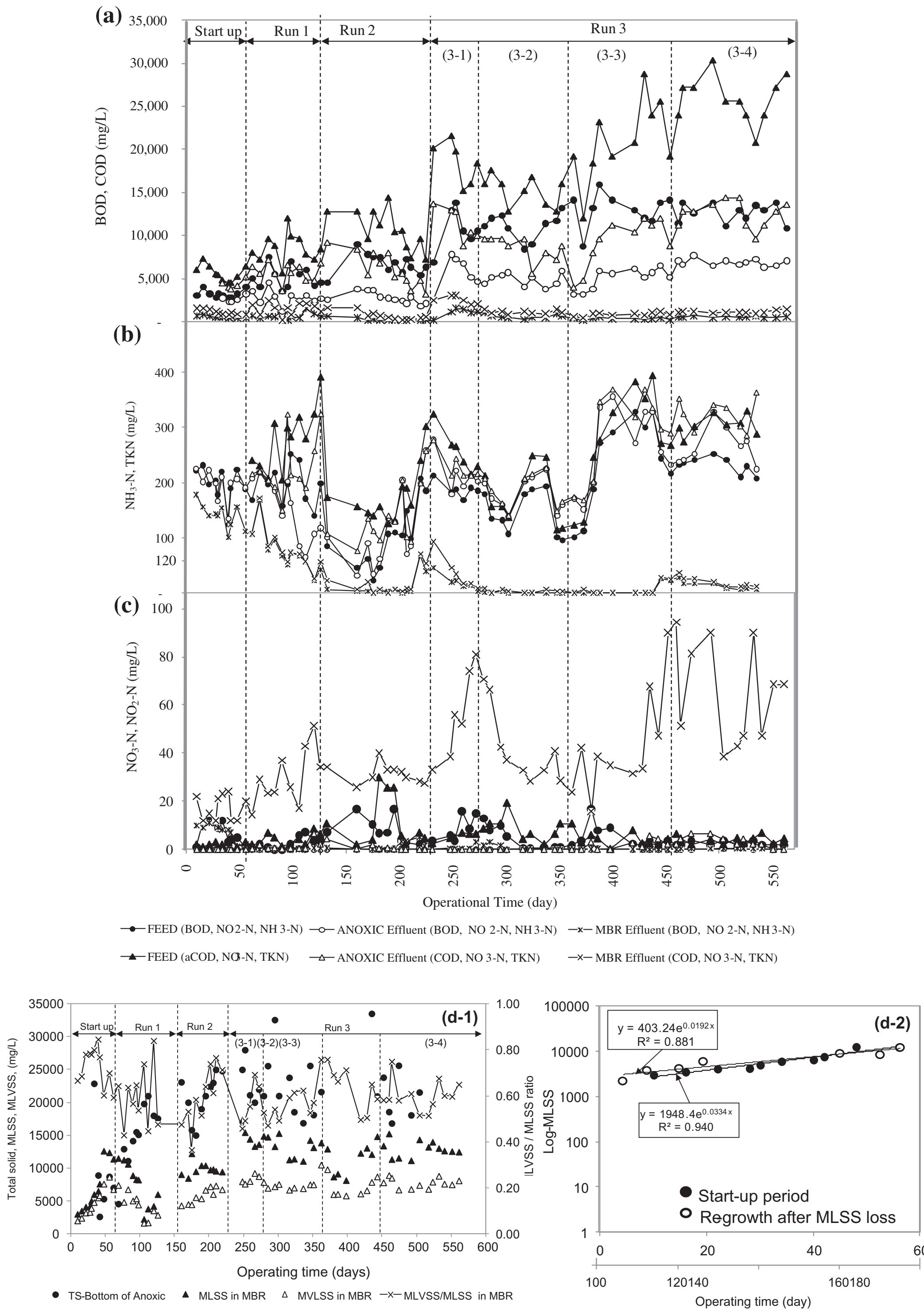

Fig. 2. Variation of water qualities and biomass concentrations (a) BOD and COD, (b) TKN and NH_3-N, (c) NO_3-N and NO_2-N, (d-1) MLSS, MLVSS in aerobic tank, TS in anoxic tank, MLVSS/MLSS, and (d-2) increasing pattern of MLVSS in aerobic tank.

Table 2
Water qualities and treatment performance of itMBR

Parameter	Influent (mg/l)			Anoxic—effluent (mg/l)			Aerobic—effluent (mg/l)			Overall removal (%)		
	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3	Run 1	Run 2	Run 3
pH	5.69 ± 0.57	5.21 ± 0.17	5.47 ± 0.59	6.42 ± 0.56	6.88 ± 0.27	7.29 ± 0.27	7.73 ± 0.23	7.81 ± 0.28	7.95 ± 0.27	–	–	–
BOD (mg/l)	5,100 ± 1,244	6,875 ± 1,011	12,180 ± 1,722	2,361 ± 890	2,738 ± 634	5,276 ± 1,267	576 ± 362	189 ± 91	584 ± 333	92 ± 3	97 ± 1	95 ± 4
COD (mg/l)	8,908 ± 2,039	11,227 ± 3,556	20,657 ± 5,194	6,058 ± 1,415	6,460 ± 2,835	10,324 ± 2,552	1,356 ± 687	646 ± 490	1,357 ± 777	85 ± 8	94 ± 4	93 ± 4
BOD/COD	0.6 ± 0.1	0.7 ± 0.1	0.6 ± 0.1	0.5 ± 0.1	0.5 ± 0.2	0.6 ± 0.1	0.4 ± 0.2	0.4 ± 0.3	0.5 ± 0.1	–	–	–
SS (mg/l)	355 ± 272	870 ± 431	1,219 ± 966	725 ± 630	839 ± 354	1,120 ± 732	ND	ND	ND	100	100	100
NH_3 (mg N/l)	196 ± 35	82 ± 38	201 ± 61	153 ± 54	94 ± 52	231 ± 59	75 ± 39	2.8 ± 1.9	11 ± 16	62 ± 18	96 ± 3	95 ± 7
TKN (mg N/l)	281 ± 57	157 ± 23	255 ± 74	227 ± 55	119 ± 36	259 ± 72	79 ± 38	7.6 ± 7.6	16 ± 21	69 ± 18	95 ± 5	94 ± 7
NO_2^- (mg N/l)	3.00 ± 2.70	7.12 ± 5.72	4.63 ± 4.55	0.06 ± 0.06	0.08 ± 0.06	0.08 ± 0.13	0.18 ± 0.25	1.38 ± 1.89	0.40 ± 0.74	–	–	–
NO_3^- (mg N/l)	4.7 ± 3.1	10.9 ± 11.3	8.0 ± 4.1	1.6 ± 2.0	0.3 ± 0.6	1.5 ± 2.1	30.3 ± 11.1	31.3 ± 4.0	52.8 ± 21.6	–	–	–
TP (mg/l)	12.8 ± 5.3	9.9 ± 2.3	13.4 ± 5.6	15.3 ± 3.6	11.4 ± 4.0	14.1 ± 5.6	2.0 ± 0.2	1.8 ± 0.2	1.2 ± 1.1	80 ± 4	86 ± 2	88 ± 6

Note: Remark: the numbers show average ± SD values.

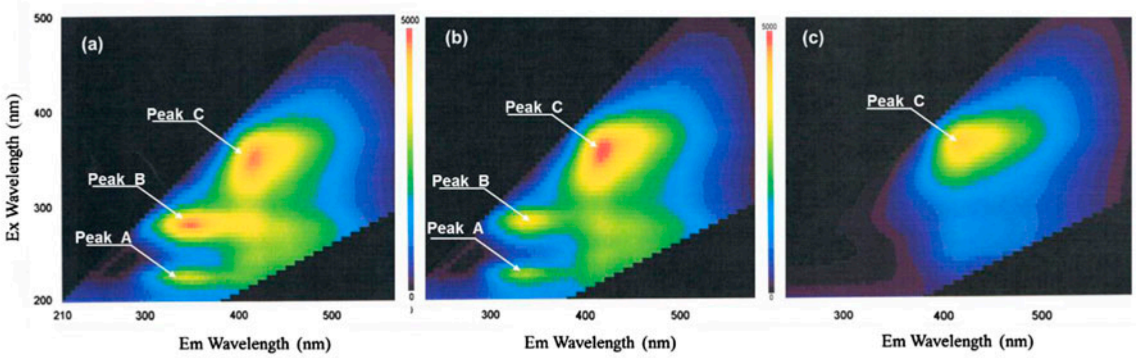

Fig. 3. Fluorescence spectra of (a) influent, (b) anoxic effluent, and (c) MBR effluent at the end of Run 3–2.

removals of 79–90% in all runs. To verify the organic matter transformation during the treatment, DOM presented in influent, mixed liquor, and effluent of itMBR was characterized by EEM spectra as shown in Fig. 3. The EEM spectra of raw leachate exhibited three main peaks. First, at the excitation/emission wavelength (Ex/Em) of 220–240/340–350 nm (Peak A), which was classified as a protein-like substance, second, at 270–290/330–360 nm (Peak B), represented a tryptophan protein-like substance associated with soluble microbial by-products and third, at 320–350/410–430 nm (Peak C) which was characterized as fulvic acid/humic-like substances [14]. In the itMBR, the fluorescence peaks of protein-like substance (Peak A, B) slightly changed during the treatment in anoxic tank and mostly disappeared after aerobic MBR tank. This observation suggests the removal of protein-like substance during the treatment in aerobic MBR, which would be expected to subsequently be deposit as a foulant layer on the membrane surface [8]. Whereas the humic-like substances (Peak C) were found at higher intensity in the anoxic tank as compared to that of the influent DOM. While lower intensity was found in effluent, it might be a result of membrane filtration. However, the remaining humic-like substances demonstrated the presence of hardly biodegradable organic substances which were not completely removed in the itMBR system.

In terms of nitrogen, the fed leachate had high fluctuation of ammonia/TKN concentrations. High removals (94–95%) of TKN were achieved in the itMBR. The efficiencies of anoxic tank for TKN removal were oscillated with the influent TKN concentrations (Fig. 2(b)). Higher NH_3 concentrations observed in anoxic effluent indicates the significance of a hydrolysis reaction of organic nitrogen compounds proceeded under first stage anoxic condition. Subsequently, a high degree of NH_3 and TKN removals were achieved in the second stage reactor as the ammonification and nitrification reactions proceeded in the aerobic tank maintained under high DO conditions. Comparison among the experimental runs, lower degree of NH_3 and TKN removals (62%) were achieved during the start-up period (Run 1) as the nitrifying organisms were slowly developed in the system [10]. In subsequent run, NH_3 and TKN removals increased up to 95% as the nitrogen loading rates were raised to 0.34 kg NH_3-N/m^3 d in Run 3. The anoxic tank provided high and stable efficiency similar to that observed in the previous study [10]. Meanwhile, nitrite concentrations were kept at relatively low concentration (<2 mg/l) while higher nitrate concentrations (30–53 mg/l) were observed in the effluent of aerobic reactor (Fig. 2(c)). Lower concentrations of oxidized nitrogen could be expected in the effluent, had the sludge recirculation been performed at a higher rate than that employed in this study (67% of feed flow rate). Previous study has reported that the sludge recirculation between aerobic and anoxic bioreactor was an influencing factor for the improvement of nitrogen removal [10]. Meanwhile, Guo et al. [15] explained that an increase in recycling rate from 50 to 100% could significantly reduce nitrogen removal from 32.4 to 12.4% due to an increase in DO concentration and deterioration of denitrification process in anoxic condition. Nevertheless, the recirculation rate was limited for biomass control purposes in this study.

3.2. *Variation of biomass concentrations*

During start-up, the aerobic tank was initially inoculated with 2.0 m^3 of sludge from an activated sludge process. During this start-up period, no sludge withdrawal was performed while the MLSS was allowed to increase as shown in Fig. 2(d-1). As a result, the MLSS concentrations increased from 3,000 up to 10,000 mg/l within two month during which an exponential growth pattern was observed as shown in Fig. 2(d-2). During the operation, average MLSS/MLVSS ratio was kept at about 0.6. The anoxic tank was then started-up by receiving excess sludge from the aerobic tank. The recirculation introduced during the operation of two-stage reactor initially affected the concentrations in the anoxic and aerobic reactor. Nevertheless, a more stable operation could be achieved when the recirculation rate of 66.7% of feed flow was employed leading to more stable biomass concentration in the aerobic reactor. The development of MLSS in the aerobic reactor was then subsequently proceeded and exponential re-growth was again observed. The MLSS concentration in the aerobic reactor was then maintained mostly between 10,000 and 15,000 mg/l while the accumulated solids in the anoxic reactor reached almost 25,000 mg/l after 550 d of operation without sludge wastage.

In itMBR, the control of biomass concentration in the aerobic reactor could be achieved by recirculation of excess biomass back to the anoxic reactor [10]. During long-term operation without sludge wastage, MBR sludge posed a good settling characteristic as indicated by low sludge volume index thus forming highly compacted sludge layer beneath the inclined tube while allowing much lower solid concentration in overflow from anoxic reactor to the anaerobic reactor [16].

3.3. Membrane fouling

The TMPs rising rates were determined under different OLR condition (Fig. 4). At relatively low OLR (Run 1), the MBR was operated at low permeate flux of 13 l/m^2 h. The TMP was gradually increased to 40 kPa with the rising rate of 0.56 kPa/d. This indicates deposition of foulants on membrane during 70 d of operation. From visual observation, there was no substantial accumulation of sludge on the membrane surface. Possibly, higher aeration intensity employed in this study helped preventing the accumulation of solids. When the TMP reached 40 kPa, chemical cleaning by CIP method was then applied. During Run 2, OLR was increased to 5.5 kg BOD/m^3 d with the membrane flux increased to 21 l/m^2 h. It was found that TMP increased rapidly (1.4 kPa/d) during this experimental period. At the end of Run 2, CIP was again performed. There were four cleaning procedures employed during Runs 3 in which OLR was gradually increase to 16.5 kg BOD/m^3 d. CIP was conducted for Run 3–1 during which the TMP was rapidly increased by three times (1.92 kPa/d) as compared to Run 1. Then, the TMP control was switched to CEB operation mode during Run 3–2 to 3–4. In Run 3–2, only NaOCl at 0.05% (w/v) was used for chemical cleaning at every 7-d interval. By this cleaning method, the TMP rising rate was 3.31 kPa/d. Subsequently, additional acid cleaning (2% w/v citric acid) prior to alkali cleaning was performed in Run 3–3 at every 7 d and Run 3–4 at every 10-d intervals. TMP rising rates of both runs were slightly different (3.35–3.49 kPa/d) and was found to be comparable to that of Run 3–2 even though OLR of these later two runs were higher than that of Run 3–2. From the results, it revealed that regular chemical cleaning using CEB technique at every 7–10 d could help maintaining TMP rising rate for sustainable operation of itMBR, even operated under high OLR condition. Wang et al. [17] reported that acid (mixture of 0.2% hydrochloric acid and 0.8% citric acid) and alkali (mixture of 1% sodium hydroxide and 0.2% NaOCl) agents were also effective to recover membrane permeability when it was mainly fouled by organic substances constituent in landfill leachate. In this study, the use of CEB cleaning technique helped restoring the TMP back to the same level as the initial condition after cleaning even after the itMBR has been operated for more than 300 d in Run 3.

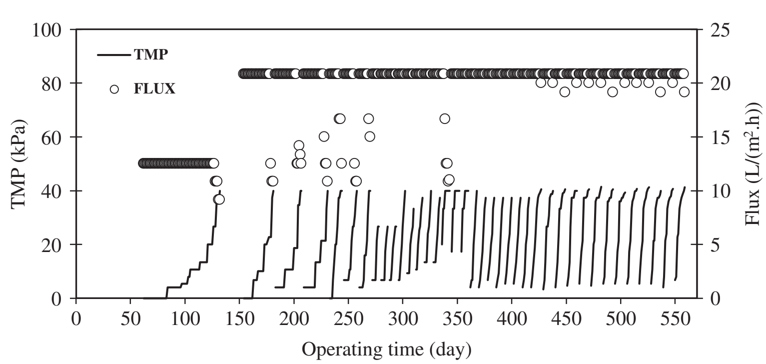

Fig. 4. Variation of membrane permeate flux and TMP in MBR.

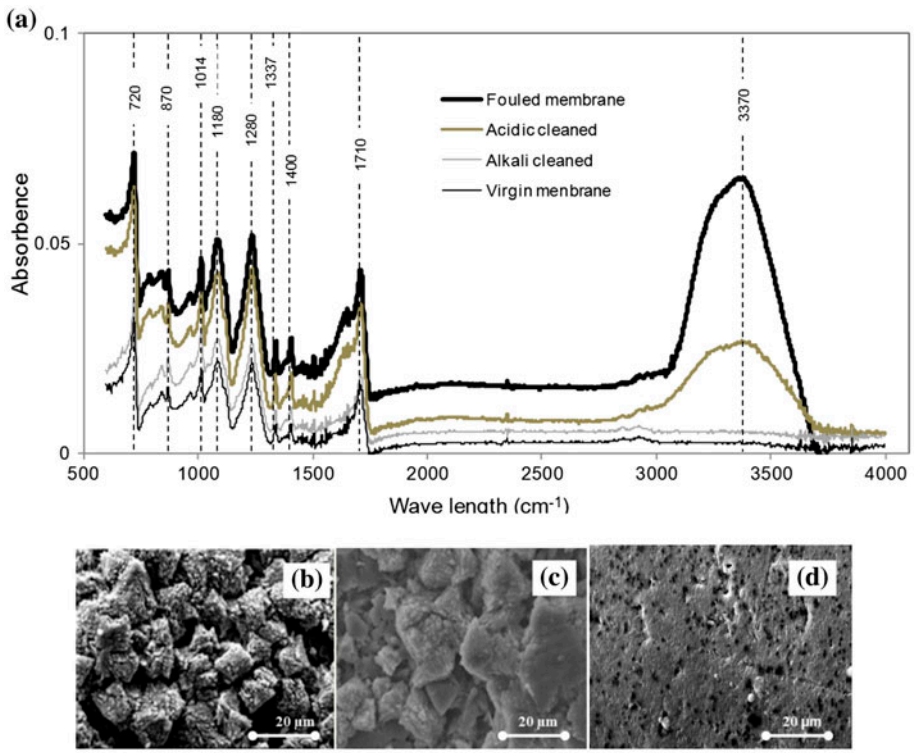

Fig. 5. FTIR spectra of fouled and cleaned membranes with different chemical cleaning reagents (a), SEM images of (b) fouled membrane (3000×), (c) alkali cleaned membrane, and (d) acidic cleaned membrane.

3.4. Membrane fouling and cleaning effectiveness

After the operation of itMBR for 550 d, the fouled membrane samples obtained from itMBR was investigated using FTIR techniques and they were subjected to chemical cleanings for evaluation of cleaning effectiveness. Fig. 5 shows the FTIR spectra of fouled and cleaned membrane. For fouled membrane (Fig. 5(a)), the functional groups of organic foulants deposited at membrane surface were characterized. A broadband peak (3,350–3,360 cm^{-1}) demonstrates an N–H stretch of an amine I/II, an amide; or an O–H stretch of alcohol and phenol. The chemical cleaning using sodium hypochlorite (NaOCl) could partially remove these compounds by reducing its concentrations. It helped removing C–F stretch (1,180/1,280 cm^{-1}) and C–H stretch (1,400 cm^{-1}), alkane/alkyl/halide/aromatic compounds (720 cm^{-1}), aromatic/amine I/II compounds (870 cm^{-1}); but partially for an N–O (1,337 cm^{-1}) and the symmetric and asymmetric C–O stretchs (1,014 cm^{-1}). Meanwhile, subsequent cleaning using citric acid could remove most of the remaining deposited foulants. From the above results, it revealed that organic fouling by protein and polysaccharide substances and alkane/alkyl/halide/aromatic compounds could be partially removed by using NaOCl but their bindings with the membrane surface were associated with inorganic scaling thus subsequent acid cleaning would be effective for their removals. The FTIR spectra of acid cleaned membrane also revealed that the cleaned membrane surface has been recovered closely to its original condition of the virgin membrane.

These results confirm that NaOCl was effective for the removal of organic foulants whereas citric acid mainly removed inorganic scaling from the membrane surface and they were in agreement with those reported in the literature [18].

The observations of deposited foulants via SEM images of membrane surfaces are presented in Fig. 5(b)–(d). It was found that there were slight differences between the images of fouled and alkali cleaning membrane. These results reveal that the organic fouling could not be eliminated after the alkaline cleaning, using NaOCl. Meanwhile, acid cleaning, using citric acid helped removing most of the remaining deposited foulants so the detachment of foulants which were bounded to the membrane by acid cleaning, was the crucial step for the membrane cleaning in this study. Furthermore, the examination of deposited foulants using EDS technique revealed that

Table 3
Elemental analysis of foulants deposited on the membrane surface and their concentrations in feeding leachate

	Found elements on membrane surface (% w/w)				Concentrations in leachate (mg/L)		
Elements	Virgin membrane	Fouled membrane	Acid cleaning	Alkali cleaning	Influent	Anoxic—effluent	MBR—effluent
C	43.56	11.33	46.63	16.31	–	–	–
F	56.44	ND	38.55	ND	–	–	–
O	ND	41.80	13.78	42.80	–	–	–
Na	ND	0.64	0.24	1.18	1,329 ± 279	3,618 ± 321	3,657 ± 249
Mg	ND	1.29	ND	1.00	209 ± 38	309 ± 33	234 ± 41
Si	ND	0.50	0.17	0.42	–	–	–
Cl	ND	0.51	ND	0.59	–	–	–
K	ND	0.39	ND	ND	–	–	–
Ca	ND	42.90	0.38	36.87	411 ± 56	683 ± 47	179 ± 26
Fe	ND	0.64	ND	0.46	72 ± 23	12 ± 2.4	6.0 ± 0.2
Al	ND	ND	0.25	0.38	–	–	–

Notes: Remark: ND: not detected; –: not analyzed, ion concentrations are shown as average ± SD.

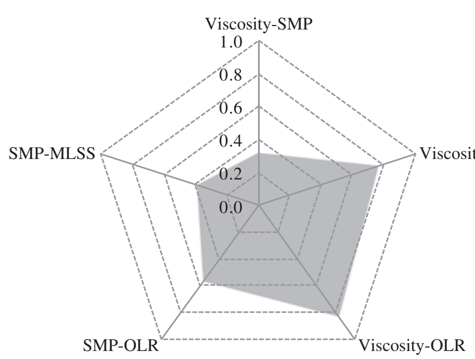

Fig. 6. Pearson correlation of viscosity, SMP, MLSS, and OLR (p-value < 0.05).

Ca (42.90%) appeared to the main element constituent in the fouling materials apart from the organic composition originated in membrane material (Table 3). Meanwhile, Na^{+}, Ca^{2+}, Mg^{2+}, and Fe^{3+} ions were found as the major inorganic elements in feeding leachate. These ions could be potentially formed into a foulant layer through their binding with humic acids and anion biopolymers [19]. Trzcinski and Stuckey [20] found that calcium in leachate precipitated as monohydrocalcite on the membrane surface causing severe fouling condition.

3.5. Factors influencing membrane fouling in itMBR

Pearson correlation technique was employed to determine the influencing factors affecting biomass characteristics, which could govern membrane fouling. Firstly, a correlation was developed between viscosity and SMP, MLSS and OLR. Linear relationship between viscosity and OLR appeared at highest positive correlation ($r_p = 0.829$). Correlation between viscosity and MLSS also showed a significant positive correlation ($r_p = 0.764$, $p < 0.05$). This is because the biomass, as expressed in term of MLSS, generally produced SMP, which leads to an increase in sludge viscosity in the aerobic reactor. Similar observation was reported [18] that viscosity had positive significant correlation with MLSS. At MLSS > 10 g/L, the viscosity of mixed liquor samples is sharply increased. Whereas, viscosity and SMP had relatively weak positive correlation ($r_p = 0.315$). The relationship between SMP and OLR and MLSS is shown in Fig. 6. It can be seen that SMP and OLR had a stronger relation than that with MLSS having r_p of 0.566 as compared to 0.390 respectively. Therefore, OLR and MLSS are considered as the key parameters governing the biomass characteristics, governing membrane fouling in this study. They should be properly controlled if the membrane fouling is to be mitigated during long-term operation.

4. Conclusion

High organic and nitrogen removals (>90%) were achieved during the treatment of concentrated solid waste leachate in inclined tube anoxic/aerobic membrane bioreactor (itMBR) operated at high OLR of 4.2–31.8 kg BOD/m^3 d. DOM in the forms of protein-like substances and tryptophan protein-like substances was successfully removed by combination of anoxic/aerobic treatment. OLR and MLSS concentration were the main parameters affecting biomass characteristics governing membrane fouling. Cleaning in place using sequential sodium hypochlorite and citric acid cleaning, was found effective for removing deposited organic foulant and scaling formed on the membrane surface. Meanwhile, CEB using the same chemicals at every 7–10 d, was also effective in limiting a TMP increase rate and sustain the MBR operation at high OLR.

Acknowledgments

This research was carried out under Research and Development of Water Reuse Technology in Tropical Region Project supported by Japan International Cooperation Agency (JICA) and Japan Science and Technology Agency (JST). The authors also appreciated financial support received from Kasetsart University Research and Development Institute (KURDI).

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