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Case Report

Moving bed anaerobic membrane bioreactor for low-strength wastewater treatment

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ABSTRACT

Anaerobic membrane bioreactors (AnMBR) combine biological processes with membrane separation in anaerobic conditions and have gained significant attention due to their high effluent quality, low energy consumption and sludge production, and biogas generation. A Moving Bed Anaerobic membrane bioreactor is an AnMBR in which particles are added to control the membrane fouling. In this research, two Moving Bed AnMBR with polyethylene glycol (PEG) granules added at around 10–15% of the reactor volume were applied for low-strength wastewater treatment at ambient temperature, for high efficiency and easy control of membrane fouling. After 240 days of operation, the COD removal rate was higher than 90%, the specific methane production was 0.23–0.28 L/gCOD$_{removed}$, the CH_4 ratio in biogas was 90–92% and the membrane function remained without fouling. PEG particles were shown to be effective for controlling membrane fouling and recovery of the membrane flux. The results suggest this AnMBR configuration could be a promising solution for municipal wastewater treatment.

1. Introduction

Environmental pollution is a serious problem in all developing countries. In Hanoi, the capital of Vietnam, all rivers and canals inside the city are highly polluted because more than 600,000 m^3/day of untreated municipal wastewater is discharged to the inner lakes and rivers. It is estimated that only 25–30% of the wastewater generated in Hanoi is treated [1]. In Vietnam and other developing countries, the wastewater treatment rate is low due to the high construction and operation costs of wastewater treatment plant; additionally, the sewage system has not been fully developed [1,2]. Finding or developing advanced wastewater treatment technologies that have low construction costs, energy demand, and plant footprint and also allow water reuse and resource recovery from treated wastewater is thus an essential task [2–4] (see Table 1).

Municipal wastewater (MWW) is considered a low or medium-strength waste stream, characterized by low organic strength (COD range between 300 and 1000 mg/L) and high particulate organic matter content [2,5]. This organic content means biogas could be produced from domestic or MWW by anaerobic processes. Anaerobic digestion comprises four key biochemical phases, defined as hydrolysis → acidogenesis→ acetogenesis → methanogenesis. The profile of these key phases determines the performance of the anaerobic degradation process [4]. Anaerobic processes strongly depend on operational temperature. However, heating of reactors requires energy and capital expenditure [6,7]. Hence, the advantages of anaerobic treatment can be maximized by operating at ambient temperatures, defined as temperatures of 5–30 °C, to minimize the internal or parasitic energy demand of the digestion process [3,8–10]. Low operational temperatures and low wastewater strength imply low biomass growth rates. Moreover, the hydrolysis, breakdown, and solubilization of complex organic matter to soluble substrates were inhibited and the biomass growth was also reduced in cold conditions [11,12]. Under such conditions, biomass concentration is difficult to maintain, especially when considerable amounts of biomass can be washed-out from the reactor. The AnMBR provides an effective solution to the above problems, as it is a combination of the anaerobic biological wastewater treatment process and membrane filtration. This approach has gained significant attention due to the high effluent quality, accurate control of microorganism retention in bioreactors, lack of need for aeration, low sludge production, and methane energy recovery [5,11,12]. High chemical oxygen demand (COD) removal efficiencies and methane conversion rates have been

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Table 1
Experimental set-up schematic.

Phase	Day	Objective	Rector	HRT	*COD (mg/L)*
Start-up	0–32	Batch experiment, acclimate inoculum to the reactor	AnMBR1 AnMBR2		
EP -1	32–74	Continuous experiment, Couple both reactors to establish a common baseline for the next stage	AnMBR1	4h - 2 days	400–500
			AnMBR2	4h - 2 days	400–500
EP -2	74–116	Continuous experiment, Reduce the HRT,	AnMBR1	1 day	400–500
			AnMBR2	1 day	400–500
EP -3	116–184	Evaluation of HRT effect on reactor performance	AnMBR1	12–15h	400–500
			AnMBR2	12–17h	700–800
EP -4	184–240	Reduce the HRT, increase the OLR, Evaluation of HRT, OLR effect on reactor performance; Increase the proportion of particles, evaluation of it on membrane flux	AnMBR1	7–9h	400–500
			AnMBR2	8–10h	700–800

achieved under various conditions when using AnMBR for municipal wastewater treatment at ambient temperatures [3,9,10,12,13]. Submerged AnMBR with flat sheet membranes, which have simple cleaning and replacement methods and good efficiency for commercial usage, could be applied for MWW treatment to reduce plant area and energy demand [7]. Lin et al. [13] conducted a feasibility evaluation of a submerged AnMBR for municipal secondary wastewater treatment at 25 °C and found the efficiency of COD removal was 88%. A cost analysis based on their lab-scale tests showed that the operational cost of an AnMBR could be only 1/3 that of an aerobic treatment process, and the energy generated from methane production can theoretically balance the energy required for the membrane biogas scouring [13].

Membrane cleaning is an essential part of the operation of MBRs and AnMBRs, which significantly influences membrane performance [14–18]. It is well-accepted that membrane cleaning can be categorized into physical and chemical cleaning. The main strategies to control membrane fouling in AnMBR include the application of crossflow for external membrane filtration and of biogas scouring for submerged membrane filtration systems [15,16,18]. Other methods tested for membrane fouling control include using ultrasonic techniques, vibrating membranes, and adding chemicals or adsorbents, such as powder-activated carbon, to improve the filterability of the mixed liquor or reduce the concentration of soluble membrane foulants [14]. Physical cleaning by relaxation and particle/carrier addition is widely applied to control membrane fouling in AnMBR because it can do this easily with little or no impact on the anoxic conditions and susceptible anaerobic micro-organisms. It has been reported that a combination of on/off mode in membrane operation, biogas scouring, and plastic particle addition increased the membrane flux from 10% to 50% and reduced the cake resistance/filtration resistance ratio from 95% to 62.1% [19].

In this research, AnMBR were applied for low-strength synthetic wastewater treatment and biogas production at ambient temperature. A combination of on/off mode in membrane operation, biogas scouring, and the addition of PEG plastic particles was applied to control membrane fouling and recover the flux of the membrane.

2. Materials and methods

2.1. Materials, experimental setup, and operating conditions

Two Anaerobic Membrane Bioreactors with an active volume of 8L were operated with the addition of PEG particles, which were cylindrical in shape and had a specific gravity of 1.01–1.15 kg/L and 4x4 mm apparent size, at 10% and 15% of the reactor working volume. A PVDF flat sheet membrane module (Kubota, Japan) with a pore size of 0.4 μm and an area of 0.11 m^2 per module was used in the reactors. The experimental set-up was as shown in Fig. 1. Both AnMBRs were run at 20 ± 1 °C - psychrotrophic anaerobic conditions. A timer was used to give a combination of on/off modes in membrane operation: 8′ on, 2’ off. Vacuum pumps and pipe diffusers were used to provide gas and liquid circulation inside the reactor.

2.2. Inoculum and characteristic of feed wastewater

Both reactors were inoculated with digestate from a mesophilic digester treating MWW biosolids at Millbrook wastewater treatment plant in Southampton, UK operated by Southern Water Plc. Prior to inoculation, the digestate was passed through a 1 mm sieve and diluted with tap water to the desired solids concentration. The initial Mixed Liquor Suspended Solid (MLSS) content of both reactors was around 5000–6000 mg/L.

Synthetic wastewater (SWW) with COD: 400–800 mg/L was prepared by mixing a ratio of dry yeast, urea, milk powder, sugar, diammonium phosphate, and tap water (1 L) for feeding the reactors. The pH of the SWW was around 6.9–7.4. The ratio of COD: TN: TP was 100: 8.94: 2.34.

2.3. Sample collection and analytical methods

Influent, effluent (treated permeate), and mixed liquor were taken regularly twice per week from the feedstock tank, permeate-draw pumps, and sampling ports of both reactors, respectively. Net permeate fluxes were determined by weight-time measurement method daily. Suction pressure was recorded by the data acquisition system. Produced biogas was collected through the gas bags which were replaced every day. The supernatant samples were obtained by centrifuging the reactor-mixed liquor at 13,000 rpm for 30 min.

The biochemical characteristics and stability of the reactors were assessed based on COD conversion, volatile fatty acids (VFA) concentration, specific methane production (SMP), and pH. Standard methods [20] were adopted for the measurement of COD and pH. Mixed liquor pH was measured with a pH meter (Jenway 3310, UK) calibrated with buffers at pH 4, 7, and 9.2 (Fisher Scientific, UK). Gas production was measured using a weight-type gasometer and reported at standard temperature and pressure of 0 °C and 101.3 kPa [6]. Biogas composition was measured using a gas chromatograph (Varian GP-3400, USA) and compared to standard biogas of 36% CO_2 and 64% CH_4 (v/v) (BOC, UK). Reported SMP values were only for methane produced in the headspace. Each experiment was conducted in duplicate.

The COD removal efficiency of the AnMBR, E (%), was calculated as follows:

$$E = \frac{C_I - C_E}{C_I} \times 100 \quad (1)$$

Where: C_I and C_E (mg/L) are the concentrations of COD at influent and effluent points of the biofiltration system.

The specific methane production (SMP) per gram of COD removed was determined using Equation (2). The term SMP in this work will always refer to the SMP per gram of COD removed unless noted.

$$\text{SMP} = \frac{(\text{Biogas Vstp})(\text{CH4\%})}{\text{CODremoved}} \quad (2)$$

Where: SMP is specific methane production per gram of COD removed (L CH_4/g $COD_{removed}$); CH_4% is methane fraction of τ (%).

Membrane performance was assessed by directly analyzing the membrane flux (permeate flow rate) at a constant transmembrane pressure.

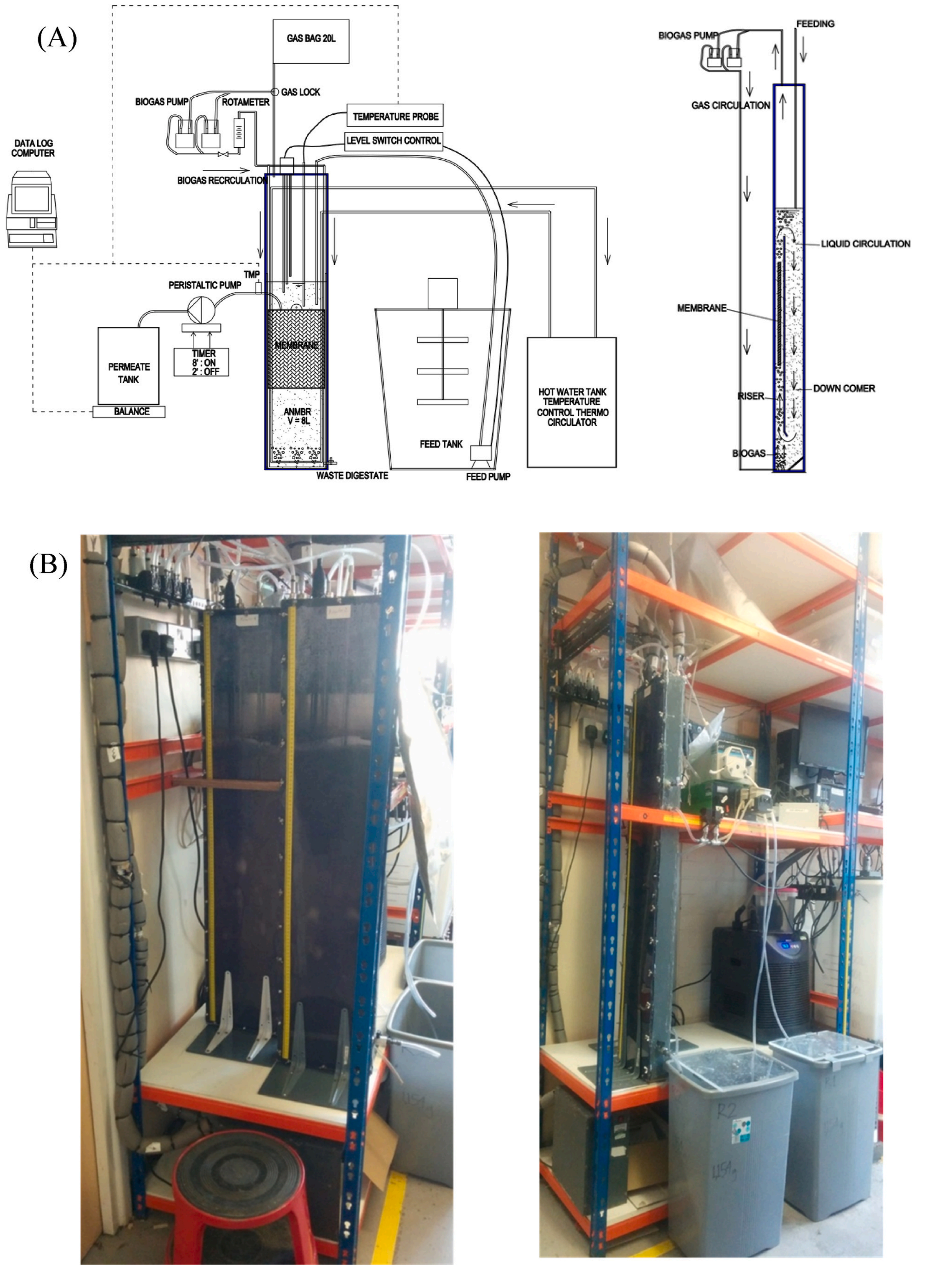

Fig. 1. (A) Schematic diagram and (B) photos of the lab-scale AnMBR system used in this study.

$$J = \frac{Q}{A_M} \quad (3)$$

Where: J = membrane flux (L/(m^2 hour); A_M = membrane area (m^2)

3. Results and discussion

The performance of the two reactors over the 240-day experimental period is shown in Figs. 2–5. Key parameters are discussed below in relation to operational changes during each of the four experimental phases.

3.1. Treatment performance of AnMBR

Fig. 2 shows the evolution of the pH and the total VFA of the two AnMBR reactors. pH and VFA are important indicators in anaerobic digestion, as they indicate any imbalances in the biochemical phases of the process due to differences in growth rates or metabolic capacities of anaerobic microorganisms [19,21,22]. Starting from pH 7.1, in the Start-up stage (batch experiment, acclimation of the inoculum to the reactor), the pH of both reactors tended to decrease quickly to 6.7. When moving to phase 1 and phase 2 (continuous experiment with long retention time), the pH of the 2 reactors gradually stabilized at about 7.0–7.2. In phase 3 and phase 4 when reducing retention time and increasing the influent COD and OLR, pH tended to decrease but was still stable in the range of 6.8–7.0, which is appropriate for anaerobic digestion.

The total VFA had a fast-increasing trend in the Start-up phase – Batch experiment. In AnMBR1 total VFA increased from 700 mg/L to 1800 mg/L, in the same trend the total VFA of AnMBR2 rose from 500 mg/L to 1300 mg/L; this implies the main biochemical phases of hydrolysis, acidogenesis, and acetogenesis were outpacing methanogenesis. In phase 1 and phase 2 - Continuous Experiment, the total VFA decreased quickly to below 100 mg/L, and the VFA was diluted and pushed out of the system along with effluent. When the OLR was controlled at a stable value, the AnMBRs worked well with high COD removal efficiency (Fig. 3) and stable VFA concentrations. In phase 3 and phase 4 when increasing the OLR, the total VFA of AnMBR2 initially jumped to 300 mg/L and 180 mg/L, at this time the COD removal efficiency was only around 70–80%. It should be noted that OLRs are also the factor that depends on other setting-up conditions for high-rate AnMBRs which most of the time VFAs play important roles, when the OLR increased, the deterioration of the system of performance due to the inhibition of microbial activity caused by VFA augmentation may occur [19,22]. After 3–4 weeks, the VFA fell and stabilized at under 50 mg/L at the end of Phase 3 and Phase 4. This suggested that when increasing the OLR, the number of microorganisms took a certain time to adapt to this change: usually, this process took several weeks, and this was a piece of valuable information for AnMBR operation. The low concentration of total VFA indicated the system was in methanogenesis.

The performance of COD removal is shown in Fig. 3. In phase 1 and phase 2 - Continuous Experiment, both reactors were operated with the same influent, COD of the influent increased from 250 to 600 mg/L and an average of around 450 mg/L. After 30 days of operation, the system had come into stability, the percentage of COD removal reached 80–90% and the COD of the effluent was under 100 mg/L (Fig. 3).

In phase 3, when the OLR of AnMBR1 was kept stable, the COD removal in AnMBR1 was also stable at around 90–94% and the effluent COD was under 50 mg/L. In this phase, the OLR of AnMBR2 increased from 0.45 to 0.9 g/(LD), the COD removal efficiency of AnMBR2 increased from 70% to 94% and the effluent COD went down to 50 mg/L at the end of phase 3. In phase 4, despite the increasing organic loading rate (AnMBR1: OLR = 0.45 → 0.9 g/(LD); AnMBR2: OLR = 0.45 → 1.4 g/(LD)) and decreasing HRT (from 10 to 8 hours), the COD removal percentages in both reactors were still around 90%, the effluent COD was under 75 mg/L and it conformed to the Vietnamese regulations for wastewater discharge QCVN 40:2011/BTNMT. The efficiency of COD removal in this study was close to and higher in some previous studies such as Lin et al., [13], (90%), and Watanabe et al., [9], (94%). The short HRT (8–10h) could reduce the required reactor volume and the construction cost of the wastewater treatment plant. These results suggested the use of anaerobic membrane bioreactors in municipal wastewater treatment with fluctuating influent COD at ambient temperature is very

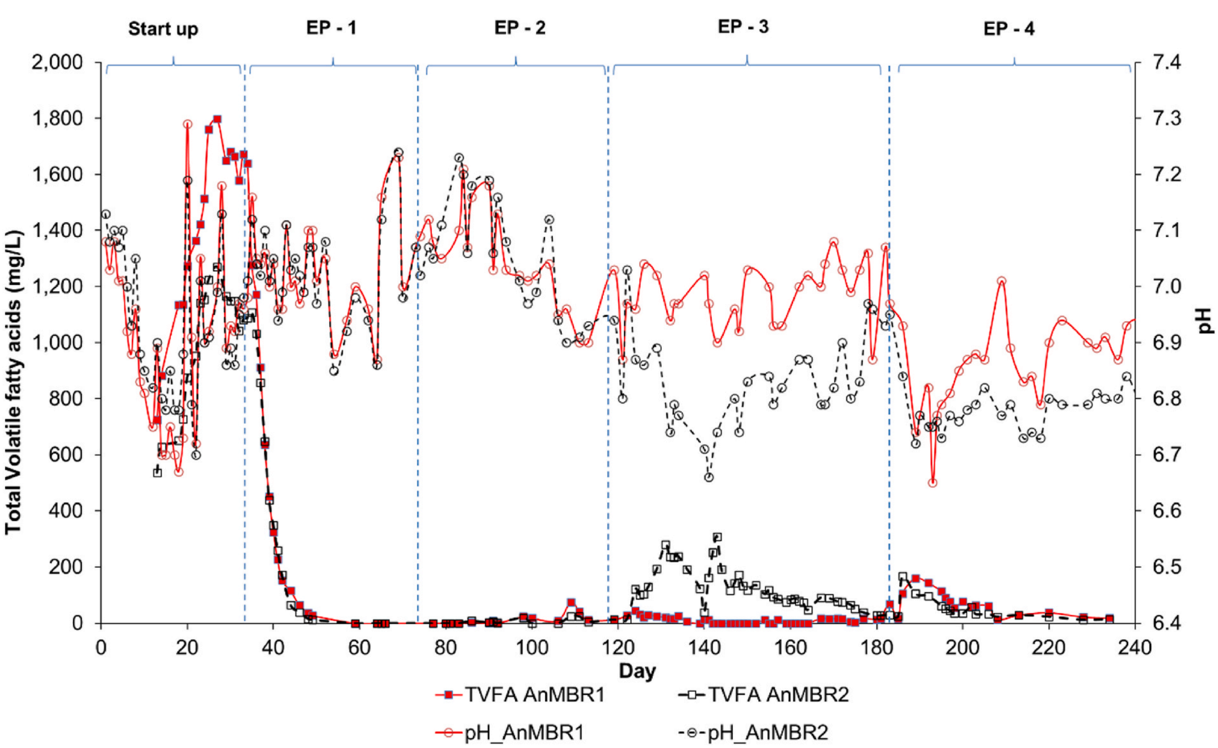

Fig. 2. pH and total volatile fatty acids (VFA) of both reactors.

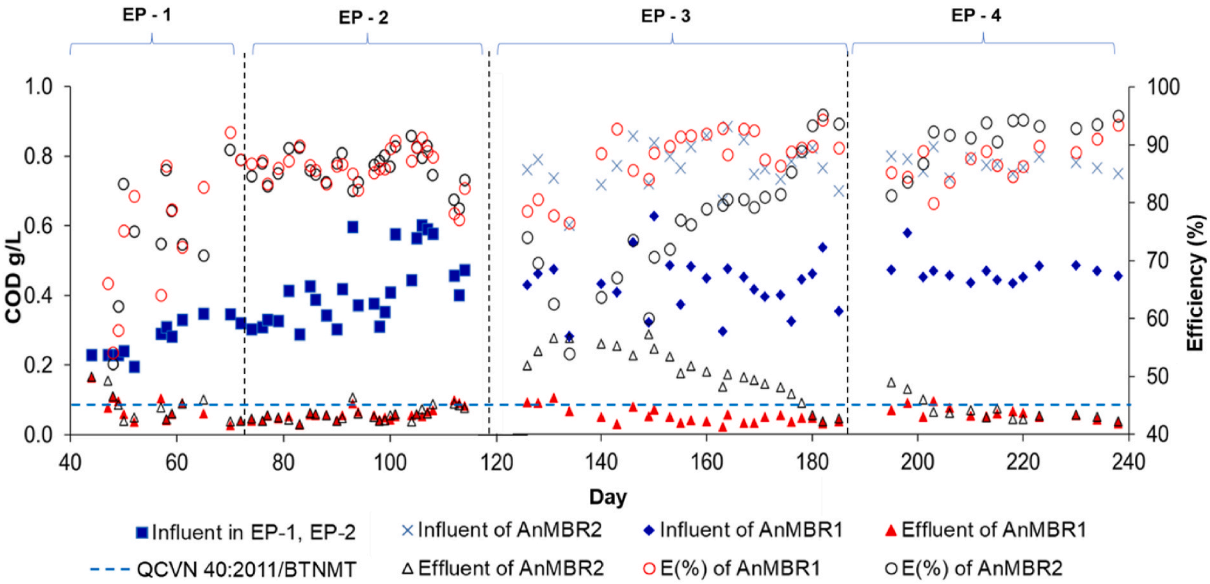

Fig. 3. The COD removal of both AnMBRs *(Note: QCVN 40:*2011/BTNMT*: National Technical Regulation on Industrial Wastewater)*.

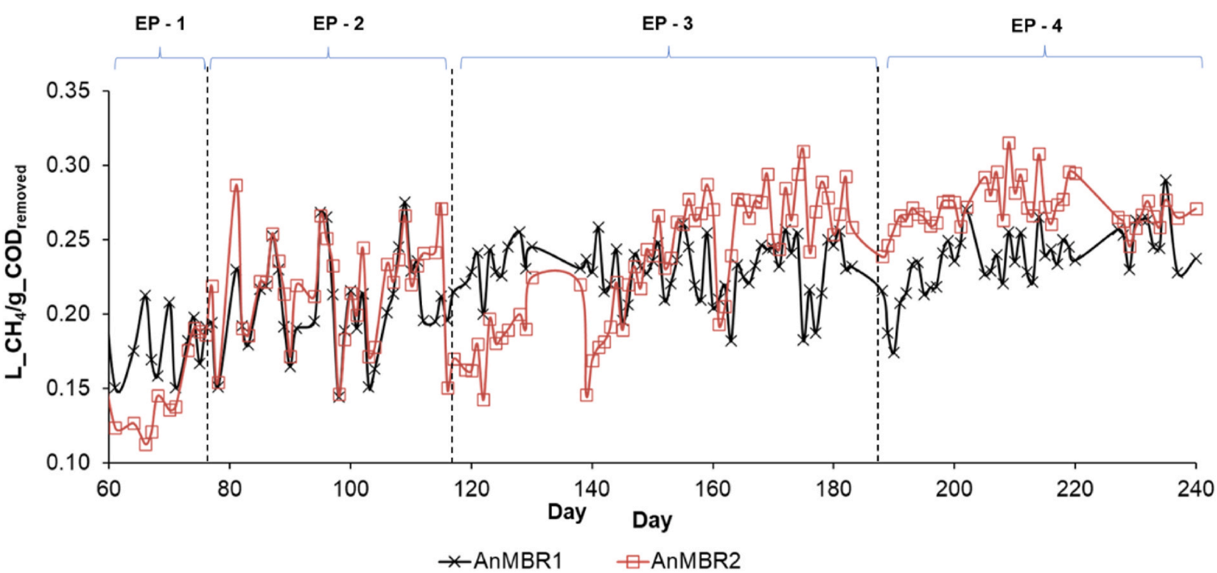

Fig. 4. The specific methane production.

promising.

The overall average methane yield of 0.23 and 0.28 L CH_4/g $COD_{removed}$) was nearly close to that in previous reports [8,18,23] in which the observed methane yield ranged from 0.24 to 0.30 L CH_4/g $COD_{removed}$ from AnMBR for MWW treatment. When the OLR increased to 0.9 g/(LD), the SMP of both reactors increased from 0.2 → 0.26–0.3 L CH_4/g $COD_{removed}$ at 20 °C. At a higher organic loading rate, the AnMBR had a higher rate of specific methane production [18]. All the COD that enters an anaerobic system ends up either in the methane gas, effluent, generated sludge, and dissolved methane [9,23]. In this study, the ratio of COD in methane gas was around 66–74%, it was a litter lower ratio than 75–77% in the report of Watanabe et al. [9].

The proportion of biogas production (CH_4+CO_2) increased from 60% to 82% in AnMBR1 and from 55% to 90% in AnMBR2 when the OLR increased. Moreover, working at a higher OLR, the AnMBR2 had a higher proportion of biogas production than the AnMBR1. At the end of phase 4, the proportions of biogas production were 82% and 90% for AnMBR2 and AnMBR1, respectively. The CH_4 ratio of both reactors was 90–92%, and the proportion of CO_2 was only 8–10% in biogas volume. The methane concentration rate in this study was higher than that reported by Martinez-Sosa et al., [8], (around 80%), Lin et al., [13], (around 70–90%), and Santiago et al., [23], (around 83–86%). Methane-rich biogas can be used for digester heating, electricity generation, or upgraded for fuel production.

3.2. Influence of particles on the membrane fouling

When extra PEG plastic particles were added to increase the proportion of particles from 10% to 15%, the flux of both membranes increased from 6 to 10 L/(m^2.h) and 5–8 L/(m^2.h) (increase of 60–66%). The PEG particles had a specific gravity of 1.01–1.15 kg/L, which is nearly the same as the specific gravity of water, so they are easily moved in the mixed liquor by gas and liquid circulation. Increasing the proportion of particles gave more contact between PEG granules, gas bubbles, and the surface of the membrane, thereby reducing the cake layer on the membrane while increasing the flux. Sriprasert [19] reported that

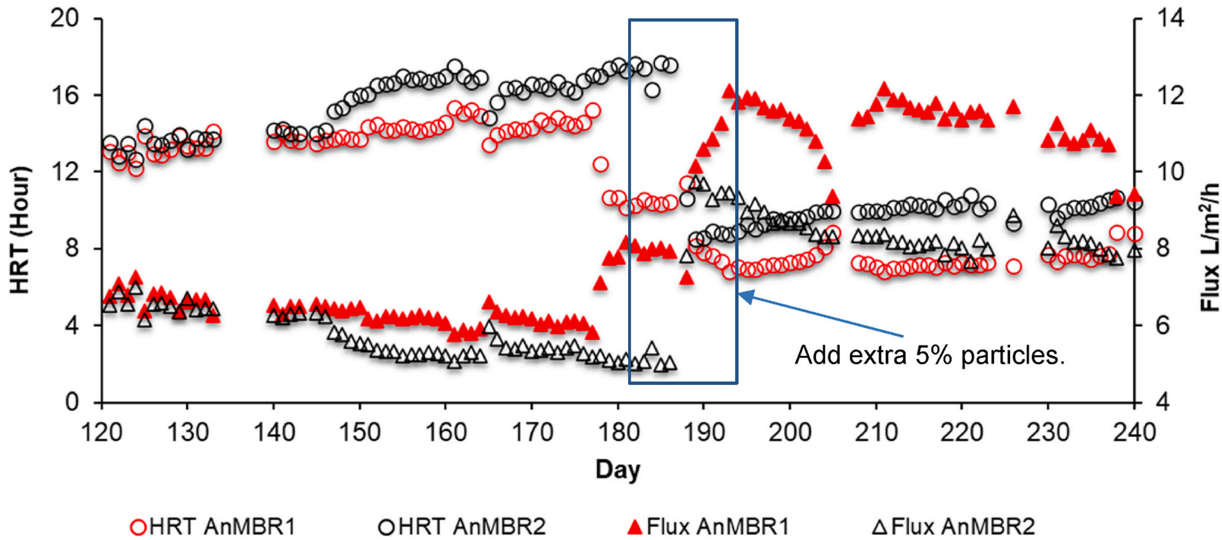

Fig. 5. The hydraulic retention time (HRT) and the flux of membranes of both reactors.

the addition of 10–20% carriers by volume could improve critical flux significantly, by up to 25–50% under the same sparging intensity, which implied that non-adsorbent particles can be applied in air-lift loop MBR as either a flux enhancer or an alternative method to reduce energy demand for sparging. The likely explanation is that the suspended particles/carriers mechanically scour the membrane surface, and the turbulence induced by the suspended particles/carriers can enhance the foulant back-transport away from the membrane surface [14]. In general, the first three mechanisms play a dominant role in membrane cleaning. Another study reported that adding PE carriers decreased cake resistance by 72.7% [17], and using PP granules could control fouling in AnMBR in long-term operation without any other cleaning at a higher flux 15–20 L/(m^2.h) [15]. Moreover, adding PEG particles did not have any negative impact on the anoxic conditions and the microorganisms compared with chemical cleaning. After cleaning, PEG granules could easily be removed from the reactor when digestate is taken out to control the solids retention time and reused several times. Using PEG to control fouling in AnMBR could therefore reduce the maintenance cost of this technology and thus help to promote the wider application of AnMBR systems.

4. Conclusions

The results of the study on the treatment of low-strength wastewater at ambient temperature using Moving Bed AnMBR with the addition of PEG particles showed that high and stable COD removal efficiency (more than 90%), specific methane production (0.26–0.3 L/gCOD$_{removed}$), and CH_4 ratio in biogas (90–92%) were achieved; and the membrane function remained without fouling. Adding PEG plastic particles were effective for controlling membrane fouling and recovery of the membrane flux by up to 66%. This improvement in the AnMBR system may lead to its widespread application of this technical and cost-effective anaerobic wastewater technology for municipal wastewater treatment with sustainable energy production.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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