LONG-TERM OPERATION OF THE PILOT SCALE TWO-STAGE ANAEROBIC DIGESTION OF MUNICIPAL BIOWASTE IN HO CHI MINH CITY

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ABSTRACT

A pilot-scale two-stage anaerobic digestion system, which includes a feed tank (0.4 m^3), a hydrolysis reactor (1.2 m^3) followed by a methane fermenter (4.0 m^3) was set up and run at the municipal solid waste landfill located in Ho Chi Minh City (HCMC), Vietnam. The feed that was separated from urban organic solid waste was collected at households and restaurants in District 1, HCMC. This study aimed to investigate the resource recovery performance of the pilot two-stage anaerobic digestion system, in terms of carbon recovery via biogas production and nutrient recovery from digestate. The average organic loading rate (OLR) of the system was step increased from 1.6 kg volatile solids (VS).m^{-3}.d^{-1}, 2.5 kg VS.m^{-3}.d^{-1} and

3.8 kg VS.m^{-3}.d^{-1} during 400 days of operation. During the long-term operation at three OLRs, pH values and alkalinity were stable at both hydrolysis and methanogenesis stages without any addition of alkalinity for the methanogenesis phase. High build up of propanoic acid and total volatile fatty acid concentrations in the fermenter did not drop pH values and inhibit the methanogenic process at high OLRs (2.5 - 3.8 kg VS m^{-3}.d^{-1}). The obtained total chemical oxygen demand (tCOD) removal performance was 83 - 87 % at the OLRs ranging from 2.5 kg VS.m^{-3}.d^{-1} and 3.8 kg VS.m^{-3}.d^{-1}, respectively. The highest biogas yield of 263 ± 64 L.kg^{-1} tCOD removed obtained at OLR of 2.5 kg VS.m^{-3}.d^{-1}. It is expected that a full scale 2S-AD plant with capacity of 5,200 tons day^{-1} of biowaste collected currently from municipal solid waste in HCMC may create daily electricity of 552 MWh, thermal energy of 630 MWh, and recovery of 16.1 tons of NH$_4^+$-N, 11.4 tons of organic-N, and 2.1 tons of TP as both organic liquid and solid fertilizers.

Keywords: *two-stage anaerobic digestion, biowaste, hydrolysis reactor, methane fermenter*

1. Introduction

Ho Chi Minh City (HCMC) daily generated 7,000 tons of municipal solid waste (MSW) that contained 60% biowaste in 2014 (Verma et al., 2015). About 85% of the MSW amount has been disposed to the sanitary landfills and the remains were sent to the composting plants (DONRE HCMC, 2016). Biogas production from biowaste for electricity generation was one of the prospective options for efficient waste recycling, which is a strategic development orientation in the Master Plan of solid waste management of HCMC to 2030 (DONRE HCMC, 2016).

Until now, anaerobic digestion (AD) has become an eco-friendly practice for municipal biowaste treatment due to various advantages such as effective organic pollution control, productive energy recovery through biogas production, and nutrient recovery through struvite precipitation (Bacenetti et al., 2019; Messineo et al., 2019; Lamnatou et al., 2019; van-

Staikenburg, 1997; Doyle et al., 2002; Chen et al., 2018; Ren et al., 2017). Deploying AD was highly suitable under the hot and humid region like Ho Chi Minh City (van Velsen et al., 1979). Two-stage anaerobic digestion system (2S-AD) which includes hydrolysis followed by fermentation can increase significantly biogas yield and stabilizing the methanogenic stage in comparison to the conventional one-stage anaerobic digestion (1S-AD) (Schievano et al., 2014; Li. et al., 2017). The fact that in the 1S-AD for high solid-containing organic wastes, the controlled pH values ranging from 7 to 8 are optimum for the growth of methanogenic bacteria and against the adverse effect of ammonia, but these values strongly inhibit that of acidifying hydrolytic bacteria which result in low whole AD performance (Sung and Tao, 2003; Buyukkamaci et al., 2004; Giovannini et al., 2016. Moreover, a strong accumulation of volatile fatty acids (VFAs) in the hydrolysis can significantly reduce the methane yield in the high-solids-1S-AD (Cho et al., 1995; Yang et al., 2013).

The physical separation of hydrolysis and methanogenesis in a 2S-AD system has been proven as an effective alternative to improve performance of hydrolysis as well as mitigating the inhibition of VFAs or ammonia (Schievano et al., 2014; Yu et al., 2016, Li, et al. 2017). Indeed, the hydrolysis phase can release methanogenic bacteria from high VFA accumulation and pH decrease (Zang et al., 2005). Furthermore, the 2S-AD system enhances the whole process stability by controlling solid retention time (SRT) of the hydrolysis stage to prevent organic solid overloading and the build-up of toxic materials such as sulfur compounds, nitrobenzene, biphenyl, and polynuclear aromatic hydrocarbon (Mizuno et al., 1998; Ng et al. 1999; Demirel and Yenigu, 2002; Walker et al., 2009). Biomass concentration in each stage can be easily controlled by adjusting different sludge retention times. Therefore, the 2S-AD system has given high performance in methane productivity and chemical oxygen demand (COD) removal in comparison to the 1S-AD system (Bouallagui et al., 2004; Demirer and Chen, 2005). In a lab-scale comparative study, Solmaz et al. (2014) demonstrated a 2S-AD

system stably ran at higher OLRs and shorter solid retention time than those of 1S-AD for organic waste from the food processing industry and municipal solid waste. Similarly, the high performance of the 2S-AD system for digestion of food waste and biotransformation of lignocellulose hydrolysate into biogas was also found (Zuo et al., 2013; Wu et al., 2018; Li et al., 2020). Some previous studies exhibited the 2S-AD process was able to produce higher methane yield than single-stage digestion. Massanet-Nicolau et al (2013) showed that the methane yield in the 2S-AD fed with food waste was enhanced by 37% in comparison with the single stage process. Similarly, Moestedt et al., (2015) reported that the biogas yield improved 12% and the methane content rose by 6% in the 2S-AD process.

One-stage anaerobic digestion has been widely used for biogas production from agricultural biomass such as animal manure, plant residues, organic solid and liquid wastes from agro-based industries such as natural rubber and cassava processing industries in Vietnam (Cu. et al., 2014; Watari et al., 2017; Fettig et al., 2013). Currently, municipal solid waste from cities or towns in Vietnam is dumped into landfills and that from a few cities are disposed of by incinerators and composting plants (Verma et al., 2016). Until now, full scale and pilot-scale studies on 2S-AD for the typical municipal biowaste generated from domestic activities of Ho Chi Minh City as well as other cities in Vietnam have been not studied yet. This leads to the lack of practices in the application of 2S-AD technology in typical conditions including characteristics of biowaste and the tropical climate of HCMC. Because of that, the long-term pilot study aimed to evaluate the performance of a 2S-AD plant for the treatment of municipal biowaste generated from HCMC.

Until now, studies on 2S-AD for the typical municipal biowaste in Vietnam have been much focused. Thereby, the long-term pilot study aimed to evaluate the performance of a 2S-AD plant for the treatment of municipal biowaste generated from HCMC.

2. MATERIALS AND METHODS

2.1. Pilot scale two-stage anaerobic digestion plant

The pilot-scale 2S-AD plant included a feed tank, a hydrolysis reactor, a methane fermenter, and a digested slurry tank (Figure 1, Figure S1). The working volumes of the feed tank, hydrolysis reactor, methane fermenter, and slurry container were 0.4 m^3, 1.2 m^3, 4.0 m^3, and 10 m^3, respectively. The sizes (diameter × height) of the hydrolysis reactor and fermenter were 0.55 m × 1.5 m and 1.8 m × 2.5 m, respectively. The heights of liquid volume and biogas containing compartment of the fermenter were 1.55 m and 1.15 m, respectively. An adjustable agitator (0.1 kW, 16-160 rpm) was equipped for the feed tank to provide a homogeneous feed mixture of the ground biowaste and the return digestate taken from the methane fermenter. A boiler (MIURA Company, Japan) with a capacity of 10 kg of steam.h^{-1} was used for supplying water steam to heat the feed mixture in the feed tank to about 55°C. After 2.5 hours of agitating, the whole volume of mixture ranging from 200 - 400 liters that was dependent on the desired organic loading rate was fed into the hydrolysis reactor during 5 - 10 minutes of pumping (a feed pump of 1.5 kW). The effluent of the hydrolysis reactor was automatically pumped into the methane fermentation at the ON:OFF mode of 5 seconds to 120 minutes. A liquid level control system was used to prevent the tank from overflowing. The digestate of the fermenter was manually pumped into the digested slurry tank when the liquid column in the fermenter was above 1.55-meter high. The hydrolysis reactor and the fermenter were equipped with a two vertical-paddle agitator (6-60 rpm) per each. An adsorption column containing 5% sodium hydroxide solution was used to remove hydrogen sulfide and carbon dioxide from the biogas generated from the fermenter.

Figure 1

2.2. Feed biowaste

The feed biowaste came from municipal solid waste (MSW), which was collected daily from households and restaurants at District 1 of Ho Chi Minh City and transported to the site by a

truck of Urban Environment Company (CITENCO). A separator (30 kW, Hitachi Zoshen, Japan) was used for separating plastic bags (containing the biowaste) from the biowaste and then cutting it into 5-mm biosolid debris. The feed biowaste included mostly wasted vegetables, fruits, and a small amount of spoiled rice, meat, etc. The non-biodegradable solid wastes (plastic waste, rubber, metal debris, cans, glasses, etc.) or hard solid wastes (bones, shells, woods, big fruit seeds, etc.) were manually separated before being fed into the separator to cut. The characteristic of the biosolids and the feed mixture are shown in Table 1.

Table 1

2.3. Seeding

To enhance the start-up phase, about 4 m^3 seed sludge containing 0.8% total solids (TS) and volatile solids (VS): TS ratio of 85% was fed into the fermenter. The seed sludge was collected from a biogas digester of a cow farm located at Cu Chi District. In the start-up period, the plant ran at 1.5 ± 0.4 kgVS.m^{-3}.d^{-1} (2.0 ± 0.7 kg COD.m^{-3}.d^{-1}) for 48 days. About 50-60 liters of seed sludge mixed with the biowaste was added into the feed tank during the start-up period when the digestate was not produced from the fermenter.

2.4. Operating condition

The biowaste and the returned digestate were fed into the mixing tank once per day or once every two days. The plant ran at organic loading rate (OLR) ranging from 0.7 - 5.5 kg VS.m^{-3}.d^{-1} ($3.5 - 8.0$ kg COD.m^{-3}.d^{-1}) that was scheduled into three periods with the step increases of organic loading rates: 1.5 ± 0.5 kg VS.m^{-3}.d^{-1}; 2.7 ± 0.6 kg VS.m^{-3}.d^{-1}; 3.7 ± 1.1 kg VS.m^{-3}.d^{-1}. The OLR was based on the total working volume of both the hydrolysis reactor and fermenter (5.6 m^3). The operating condition is presented in Table S2. The hydrolysis reactor and methane fermenter were run at the controlled operating temperature of about 32°C and 35°C, respectively, using water steam provided from the boiler. The pH value of the fermenter was adjusted from $7.4 - 7.5$ using a 6% NaOH solution in cases of pH value of

digestate less than 6.0. The weight ratio of the ground biowaste to the returned digestate ranged from 1.0:1.0 to 1.0:1.5.

Table S1

2.5. Analytical methods

Soluble and total COD concentrations, TS, VS contents, NH_4^+-N, total phosphorous (TP), and total nitrogen (TN) were weekly analyzed according to the water and wastewater examination standard methods (APHA, AWWA, 1998). The pH and temperature were measured on-site. VFAs were analyzed using gas chromatographic separation method 5560 B (APHA, AWWA, WEF, 1998) using GCMS-QP2010 SE (Shimadzu, Japan) with column TG – WAXMS 30 mm. The biogas composition was determined by a portable biogas meter Geotech 5000, UK.

2.6. Data analysis

SRT (solid retention time) of the 2S-AD system is the sum of SRTs of the hydrolysis reactor and methane fermenter. In this study, it is assumed that the SRT of the feed tank is not neglected (2.5 h/24h = 0.1 d). SRT (days) of each tank is defined as follows:

$$SRT = \frac{V_r}{Q_s + Q_r}$$

Where: V_r = Volume of the reactor (m^3),

Q_s = Daily volume of feed solids ($m^3.d^{-1}$),

Q_r = Daily volume of returned digestate ($m^3.d^{-1}$).

Whereas, OLR (organic loading rate, kg VS.$m^{-3}.d^{-1}$ or kg tCOD.$m^{-3}.d^{-1}$) of the whole system is determined as follows:

$$OLR = \frac{Q_s \times C_o}{V_h + V_f}$$

Where: Q_s = Daily of feed biowaste ($m^3.d^{-1}$),

C_o = VS or tCOD concentration of the feed biowaste (g.L^{-1})

V_h = Working volume of the hydrolysis reactor (m^3),

V_h = Working volume of the methane fermenter (m^3),

The relationship between SRT and OLR is illustrated as follows:

$$OLR = \frac{C_o}{SRT}$$

Hydrolysis efficiency (%E$_h$) was determined as follows:

$$E_h, \% = \frac{(pCOD_{inf} - pCOD_{eff})}{pCOD_{inf}} \times 100$$

Whereas: pCOD$_{inf}$ – Particulate COD (pCOD) of the influent.

pCOD$_{inf}$ – Particulate COD of the effluent.

3.   RESULTS AND DISCUSSION

3.1.   COD and VS

The pilot plant continuously ran for 400 days, except for when it stopped feeding biowaste for two weeks in Tet/Lunar New Year holidays (from day 55th to day 76th). Besides, after the first 16 days in the start-up period, pH values in the methane fermenter drastically decreased below 6.5. The biowaste feeding stopped for one week for the recovery of the fermenter performance. The pilot plant started producing significant biogas volume after 48 days of the start-up period at 1.5 ± 0.4 kg VS.m^{-3}.d^{-1}. Three average organic loading rates (1.6; 2.5 and 3.8 kg VS.m^{-3}.d^{-1}) were examined by changing the volume of the ground biowaste that was fed into the plant. Variation of VS and total chemical oxygen demand (tCOD) concentrations of the feed biowaste, effluent of hydrolysis reactor, and digestate during 400 days of operation was shown in Figure 2a and Figure S2a, respectively.

Figures 2b and S2b presented VS and tCOD removal rate of the whole pilot plant linearly increased as OLRs changing from 1.0 to 5.5 kg VS.m^{-3}.d^{-1} (1.5 to 7.5 kg tCOD.m^{-3}.d^{-1}), respectively. Nonetheless, after examining VS or tCOD removal of each stage, it is found that

the methane fermenter played a key role in organic removal, whereas, organic removal of the hydrolysis reactor was not happening at OLR higher than 2.0 kg VS.m^{-3}.d^{-1} (around 3 kg tCOD.m^{-3}.d^{-1}).

Figure 2

Figure 3a and Figure S3a showed that there was not much difference between VS and tCOD concentrations of the digestate as the OLRs stepping up from 1.6, 2.5 to 3.8 kg VS.m^{-3}.d^{-1}. The average VS and tCOD concentrations of the digestate were from 20 ± 8 g.L^{-1} and from 45 ± 14 g.L^{-1} (n = 55), respectively at three OLRs. High VS and tCOD removals of the pilot plant achieved at three OLRs. The obtained tCOD treatment performances were $87 \pm 3\%$ and $83 \pm 4\%$ at the average OLRs of 2.5 kg VS.m^{-3}.d^{-1} (SRT of 54 days) and 3.8 kg VS.m^{-3}.d^{-1} (SRT of 41 days), respectively. The performance was similar to the result of the previous study, which applied a laboratory-scale 2S AD for treating vegetable waste. The result showed that 87 % of organic matter was removed at operating conditions of overall OLR of 4 kg COD.m^{-3}.d^{-1} and SRT of 17 days (Bayr al et al., 1998).

Similarly, high total suspended solids (TSS) and volatile suspended solids (VSS) removals were obtained at all OLRs. Figure S3b addressed that VSS concentrations of digestate were 14.0 ± 5.0 g.L^{-1} (removal of 72%) and 16.1 ± 2.8 g.L^{-1} (removal of 82%) at the average OLR of 2.5 and 3.8 kg VS.m^{-3}.d^{-1}, respectively. The average ratio of VSS to TSS decreased from 0.78 for feed biowaste to 0.61 for digestate at OLR of 2.5 kg VS.m^{-3}.d^{-1}.

Unlikely the digestate, Figure S3b illustrated that tCOD, pCOD (particulate COD), VS and VSS concentrations of hydrolysate significantly increased at higher OLR or shorter SRT. The average VSS concentrations of the hydrolysate were 35 ± 12, 53 ± 11, and 76 ± 21 g.L^{-1} at the average OLRs of 1.6, 2.5 and 3.8 kg VSS.m^{-3}.d^{-1} (average detention times of 11 days, 7.2 days, and 5.5 days), respectively.

Figure 3b illustrates that the total hydrolysis efficiency (E_h) values which are the sum of E_h of

the hydrolysis reactor and that of the fermenter were not significantly different at three OLRs. The total E_h values of the whole pilot plant approximated $83 \pm 5\%$ and $79 \pm 7\%$ in terms of pCOD and VSS, respectively. However, as the OLRs increase, the E_h value of the hydrolysis reactor significantly decreased, but that of the methane fermenter was on the contrary. The E_h of the hydrolysis reactor obtained about $52 \pm 18\%$, $40 \pm 14\%$, and $32 \pm 16\%$ at the average SRTs of 11 days, 7.2 days, and 5.5 days, respectively. These SRTs of the hydrolysis were similar to those of 2S-AD systems that are fed with coffee wet wastewater (SRT of 5.5 days) and food waste (SRT of 6.7 days), respectively (Yans et al., 2014; Wang and Zhao, 2009). However, in comparison with the study of Yan et al. (2014), the lower E_h (by 32%) of the hydrolysis process at SRT of 5.5 days was found in the study. It is obvious that the larger amount of insoluble organic matter, like pCOD (107 ± 28 g.L^{-1}) or VSS concentrations (76 ± 21 g.L^{-1}) in the hydrolysis reactor, limited the activity of hydrolytic bacteria at OLR of 3.8 kg VS.m^{-3}.d^{-1}. Thus, the hydrolysis efficiency much depends on the size of the feed biowaste, temperature, and characteristics of the feeds, such as food processing waste or municipal organic waste (Solmaz et al, 2014; Ventura et al., 2014).

Figure 3

3.2. VFAs, pH, alkalinity, and ammonia

The change of pH, alkalinity, tVFA, and NH$_4^+$-N versus operating time is presented in Figure 4. The effluent pH values and alkalinity were stable after the start-up period (the first 48 days). pH values were not controlled in both hydrolysis reactor and fermenter after the start-up period. Thus, SRTs of the hydrolysis reactor and the fermenter (5.5 ± 0.8 d and 35 ± 5 d, respectively) at OLR of 3.8 kg VS.m^{-3}.d^{-1} were long enough to make stability of the 2S-AD process. The average pH value and alkalinity of the digestate were 7.6 ± 0.2 g.L^{-1} and 12.2 ± 2.8 g.L^{-1} as CaCO$_3$ at three OLRs (Figures 4a and 4b). pH values of 7.0 - 7.5 were suitable for the 2S-AD system to control the toxicity of high free ammonia (FA) which strongly

inhibited methanogenesis bacteria growth (Ward et al., 2008; Nathao et al., 2013; Handous et al., 2017). Similar to the fermenter, pH values and alkalinity of the hydrolysis reactor were not much changed at all OLRs. The average pH value and alkalinity of hydrolysate for 400 days of operation were 5.7 ± 0.3 and 5.78 ± 1.76 g.L^{-1} (n = 55) as $CaCO_3$, respectively. These pH values were in the range of 5.5-6.1, which was proper for acidifying bacteria that efficiently produce VFAs (Wang and Zhao, 2009; Giovannini et al., 2016). High alkalinity in the hydrolysis reactor, which provided from digestate return with the biowaste to digestate ratio of 1.0:1.0-1.5, can improve stability of acetogenesis.

Figure 4

Figure 4c shows that tVFA concentrations of hydrolysate increased rapidly even at low OLR (1.6 ± 0.8 kg VS.m^{-3}.d^{-1}), whereas those of the digestate only increased at higher OLRs. This is due to the acidogenesis process in which soluble organic compounds were fast converted into VFAs by acidifying bacteria in the hydrolysis reactor, while methanogenic bacteria used VFAs to convert to methane gas and CO_2 at low OLR (1.6 kg VS.m^{-3}.d^{-1}) in the fermenter. At high OLR (3.8 kg VS.m^{-3}.d^{-1}), short SRT of the hydrolysis reactor (5.5 ± 0.8 days) caused a large amount of un-hydrolyzed solid organic matter washed out from the reactor and then they were solubilized in the fermenter.

The fact that the mean tVFA concentrations of the digestate progressively increased from 0.89 g.L^{-1} at OLR of 1.6 kg VS.m^{-3}.d^{-1} to 9.08 g.L^{-1} at OLR of 3.8 kg VS.m^{-3}.d^{-1} (Figure 5). Furthermore, the mean tVFA:sCOD ratio of digestate distinctly increased from 0.07 at OLR of 1.6 kg VS.m^{-3}.d^{-1} to 0.43 at OLR of 3.8 kg VS.m^{-3}.d^{-1}. High VFA:COD ratio can inhibit methanogenic bacteria growth (Wei et al., 2015). Even though high VFA:COD ratio as well as high tVFA concentration presented at high OLR (3.8 kgVS.m^{-3}.d^{-1}) in the fermenter, pH values still maintained good stability for methanogenesis (7.54 ± 0.13). This can be explained by sufficient buffering capacity thanks to high alkalinity (12.7 ± 2.8 g.L^{-1} as $CaCO_3$) that well

limits pH decrease and then restoring a balance between acidogenesis and methanogenesis co-existing in the fermenter at the appropriate high OLR (Wang et al., 2005; Handous et al., 2017).

High ammonium (NH_4^+) and free ammonia (NH_3) concentrations can seriously inhibit methanogenesis (Magdalena et al., 2019). Figure 4d presented that total ammonia concentrations (TAN) of both hydrolysate and digestate rapidly increased from low OLR (1.6 kg $VS.m^{-3}.d^{-1}$) to high OLRs (2.5 and 3.8 kg $VS.m^{-3}.d^{-1}$). The mean TAN concentrations of hydrolysate and digestate at OLR of 3.8 kg $VS.m^{-3}.d^{-1}$ were 3.63 ± 0.26 and 2.85 ± 0.25 $g.L^{-1}$, respectively. Observably, these high TAN concentrations did not cause negative impacts on both stages of the pilot plant. Those were lower than 4 $g.L^{-1}$, which was widely claimed as an inhibitory value for the anaerobic digestion (Fujishima et al., 2000; Garcia et al, 2009). Otherwise, high ammonia concentration was useful for biomass conversion and VFAs production in 2S-AD (Angelidaki and Ahring, 1994; Massé et al., 2014).

Figure 5

It is recognized that VFA is a relevant parameter for evaluating stability of anaerobic digestion. Figure 5 and Figure S4 showed the concentration and distribution percentage of VFAs of hydrolysate and digestate at three examined OLRs. The large gaps between tVFA concentrations of hydrolysate and that of the digestate adduced the high performance of VFA conversion of the methanogenesis into methane in the fermenter. No considerable difference between tVFA concentrations of hydrolysate at three OLRs happened (Figure 5). The average VFA yields produced in the hydrolysis reactor at three OLRs were 677 mg VFA.g^{-1} $VS_{removed}$ or 513 mg VFA.g^{-1} $COD_{removed}$ or 121 mg VFA.g^{-1} $tCOD_{feed}$ which is lower than that of study of using microalgae biomass as a substrate for the AD process (300 mg.L^{-1} VFA-COD.g^{-1} $tCOD_{feed}$ (Magdalena et al., 2019). This may be due to the much smaller size of microalgae biomass than that of ground feed biowaste with a size of 5 mm.

Total VFA (tVFA) of hydrolysate considerable decreased from 15.5 ± 3.7 g.L^{-1} to 6.5 ± 3.6 g.L^{-1} in digestate (removal of 58%) at OLR of $2.5 - 3.8$ kg VS.m^{-3}.d^{-1}, while tVFAs of hydrolysate and digestate were 16.3 ± 5.9 g.L^{-1} and 1.03 ± 0.4 g.L^{-1}, respectively (removal of 94%) at OLR of 1.6 kg VS.m^{-3}.d^{-1}. However, concentrations of sCOD as well as tCOD of digestate at three OLRs were not much different (Figure S3a). Ratios of tVFA-COD to sCOD at OLRs of 1.6 kg VS.m^{-3}.d^{-1} and 2.5 - 3.8 kg VS.m^{-3}.d^{-1} were 7.7% and 37.7%, respectively. This exhibited tVFA did not significantly contribute to sCOD of digestate at low OLR, which sCOD is predominant non-VFA compounds like humid and fulvic compounds.

Figure S4 illustrated that acetic and propanoic acids, as the main components of VFAs, accounted for 31-41% and 17-24% of tVFAs production in the hydrolysate and digestate at higher OLRs (2.5 and 3.8 kg VS.m^{-3}.d^{-1}), respectively. While acetic and butyric acids were the most abundant products obtained at low OLR (1.6 kg VS.m^{-3}.d^{-1}), which corresponded to 39% and 20-25% of tVFAs produced in the hydrolysis reactor and methane fermenter, respectively. Various VFAs in the hydrolysis reactor and the fermenter may have different effects on performance of acetogens and methanogens. The fact that propionate inhibits more severely than butyrate and acetate in the methanogenic fermenter, whereas butyrate can be decomposed more easily than the others due to its higher energy production during the digestion (Rajagopal et al. 2013). In comparison with other VFAs, digestion of acetate can obtain the highest methane yield because of its one-step degradation (Wong et al., 2008). A significant increase in tVFA concentration in the fermenter at higher OLRs was attributed to acidogenesis of the insolubilized particulate solids washed out from the hydrolysis reactor. Wang et al. (2009) illustrated that no considerable inhibition of methanogens at acetic acid concentration of 2.4 g L^{-1} and butyric acid concentration of 1.8 g L^{-1}, whereas a propanoic acid concentration of 900 mg L^{-1} significantly inhibited the methanogens. The average acetic, propanoic, isobutyric and butyric acids in digestate at OLR of 2.5 - 3.8 kg VS.m^{-3}.d^{-1} were

2.6 ± 1.4 g.L^{-1}, 1.3 ± 0.9 g.L^{-1}, 0.76 ± 0.48 g.L^{-1} and 0.78 ± 0.62 g.L^{-1}, respectively. This showed high propanoic acid build-up in the fermenter did not drop pH values and inhibit the methanogenic process at high OLRs (2.5 - 3.8 kg VS m^{-3}.d^{-1}). This may be explained that high acetic acid concentration (2.6 ± 1.4 g.L^{-1}) and high ratio of acetic acid to propanoic acid (1.93 ± 0.6) in the fermenter resulted in stimulation of methanogens activity (Moestedt et al., 2020).

3.3. Methane recovery efficiency

Figure S5 showed the time course of biogas production during the experiment. About the first 90 days of start-up operation at the average OLR of 1.5 kg VS m^{-3}d^{-1}, the volume of biogas insignificantly produced. This concurrently happened with low VS and tCOD removals (Figure 2a and Figure S2a). Figure 6 illustrated that the highest biogas production yield (263 ± 64 L.kg^{-1} tCOD removed) obtained at OLR of 2.5 kg VS$\bullet$m^{-3}.d^{-1}. The biogas production yield decreased at OLR of 3.8 kg VS$\bullet$m^{-3}.d^{-1} (189 ± 60 L CH$_4$.kg^{-1} tCOD removed). This might be attributed to incomplete conversion of VFAs to methane in the fermenter at high OLR. Indeed, the average tVFA concentrations of the digestate rapid increased from 3.55 ± 2.32 g.L^{-1} at OLR of 1.6 kg VS m^{-3}.d^{-1} to 9.08 ± 2.49 g.L^{-1} at OLR of 3.8 kg VS$\bullet$m^{-3}.d^{-1} (Figure 6).

Figure 6

Lower biogas yield (202 ± 65 L CH$_4$.kg^{-1} COD removed or 238 ± 85 L CH$_4$.kg^{-1}VS$_{fed}$) was obtained at lower OLR (1.6 kg VS$\bullet$m^{-3}.d^{-1}). This can be explained by organic carbon and VFA losses in the hydrolysis reactor operated at long SRT (11 ± 3 days). While higher biogas yield (around 578 ± 280 L CH$_4$.kg^{-1}VS$_{fed}$ and 258 ± 94 L CH$_4$.kg^{-1}VS$_{fed}$) achieved at higher OLRs (2.5 kg VS.m^{-3}.d^{-1} and 4.3 kg VS.m^{-3}.d^{-1}, respectively) and was much higher than those from studies on the 1S-AD process. The biogas yield obtained from the study of Schirmer et al (2014), using the 1S-AD process for treating fresh municipal solid waste was 75 L.kg^{-1}

VS_{fed}. In another study, higher efficiency at 166 $L.kg^{-1}$ VS_{fed} obtained as treating the different substrate (Amon et al, 2006). Figure S6 shows that no significant gaps in methane and CO_2 distributions in the biogas produced at three OLRs. The average share of CH_4, CO_2, O_2, H_2S and other gases in the produced biogas at OLR of 2.5 – 3.8 kg $VS.m^{-3}.d^{-1}$ was 67 ± 3 %, 25 ± 2%, 0.6 ± 0.5 %, 4.1 ± 3.9 ppm and 7.6%, respectively. The content of the on-site measured biogas components using portable biogas meter Geotech 5000 at three OLRs was described in detail in Table S2.

3.4. TN and TP

Figures 7a and 7b presented the variation of TKN and TP of the feed biowaste, hydrolysate, and digestate in whole the experiment, respectively. The average TKN and TP concentrations of the fed biowaste were 8.22 ± 2.45 $g.L^{-1}$ (n = 41) and 1.89 ± 0.40 $g.L^{-1}$ (n = 32), respectively, whereas the average TKN and TP concentration of the digestate were 3.14 ± 1.47 $g.L^{-1}$ and 0.40 ± 0.07 $g.L^{-1}$, respectively.

Figure 7

Digestate obtained from the 2S-AD system has decreased VS contents (21 ± 8 $g.L^{-1}$), elevated pH values (7.5 ± 0.4) and alkalinity (11.5± 0.4 $g.L^{-1}$ $CaCO_3$), high TKN (3.1 ± 1.5 $g.L^{-1}$) and medium TP (0.40 ± 0.08 $g.L^{-1}$). The high ammonium (NH_4^+) to total nitrogen (N) ratio (63 ± 8%) reveals digestate as an immediately bioavailable material for plant growth. The characteristic of the digestate in this study is similar to those from biogas plants fed with animal manures that usually use as fertilizer in Germany (Möller and Müller, 2012). Furthermore, some studies showed that digestate could be utilized as fertilizers for vegetable cultivating farms (Liu et al., 2009), which require a large amount of quick-release fertilizers (Möller and Müller, 2012).

Figure S7 shown that the mean TKN loss from the 2S-AD system at the OLR of 1.6 kg $VS.m^{-3}.d^{-1}$ was high (69 ± 12 %), whereas TKN losses at high OLRs (2.5 and 3.8 $VS.m^3.d^{-1}$)

were lower (53 ± 10 % and 54 ± 5 %, respectively). TP losses at three OLRs were not much different and the average value ranged from 75 to 82% (Figure 7b). TKN and TP losses might be attributed to the formation of struvite (Hidalgo et al., 2015) and ammonium carbonate (Möller and Müller, 2012). A small amount of free ammonia (NH_3) may be stripped into the biogas stream at low pH values (<1%) (Kirchmann and Witter, 1989). On the contrary, UNL Water (2020) claimed that considerable NH_3 amount volatized during anaerobic digestion at high pH value and temperature. Indeed, the slightly high pH values (7.59 ± 0.17) and high alkalinity (13.3 ± 2.7 g L^{-1} as $CaCO_3$) may increase free ammonia stripping at OLR of 2.5 – 3.8 kg VS $m^{-3}.d^{-1}$. Moreover, the returned digestate containing high ammonia concentration (2700 ± 560 mg L^{-1}) was mixed with biowaste at temperature of $55^{o}C$ that enhanced strong volatilization of free ammonia. Besides struvite precipitation, TP loss might be due to the precipitation of metal phosphate such as calcium phosphate, ferric phosphate under high alkalinity in the fermenter (Hjorth et al., 2009).

3.5 Perspectives of the potential application of 2S-AD digestion in solid waste disposal in HCMC

DONRE of HCMC (2018) reported that HCMC daily generated about 8,900 tons of MSW that consisted of 65-90 % of biodegradable matter. The annual MSW generation rate of HCMC was 7 - 8%. About 7,200 – 7,500 tons of MSW were collected daily from households and public facilities and transported to the solid waste disposal facilities consisting of landfills (75% of the collected MSW), composting plants (15%), and incinerators (5 - 10%) (DONRE HCMC, 2018). However, the current MSW disposal facilities have caused serious problems such as the generation of malodorous smell from composting plants and landfills, air pollution from incinerators, and large land use of landfills.

The use of anaerobic digestion for degradable organic solid waste can produce remarkable biogas as a renewable energy source that is efficiently replaced for fossil fuel (Nguyen et al.,

2014) and the digestate can be used as fertilizer for agriculture (Möller and Müller, 2012). The practical operation of the pilot 2S-AD plant (for 400 days) illustrated the high performance in terms of organic removal, biogas yield and nutrient recovery for biowaste separated from MSW in HCMC. The achieved results showed that the average VS removal, biogas yield, NH_4^+-N, TKN and TP recovery of the pilot plant operated at OLR of 2.5 - 3.8 kg VS.m^3.d^{-1} at temperature of around 35°C were 87 $\pm$ 5 %, 52 $\pm$ 19 Nm^3 biogas ton^{-1}, 2.7$\pm$ 0.6 kg ton^{-1}, 4.3 $\pm$ 0.7 kg ton^{-1} and 0.40 $\pm$ 0.07 kg ton^{-1} of wet weight biowaste (n = 30), respectively. Beneficial biogas utilization in terms of environmental protection and economic perspectives has been demonstrated in the practice. The fact that biogas has been potentially used for both heat energy and electricity productions and as vehicle fuel (Persson et al., 2006; Holm-Nielsen, et al. 2008). Murphy et al. (2003) evaluated the asset value of 1 m^3 biogas that may produce electricity of 2.04 kWh and the met energy of 2.33 kWh. Thus, application of 2S-AD process for biowaste of Ho Chi Minh City can generate about the average electricity of 106 kWh ton^{-1} and thermal energy of 121 kWh ton^{-1} (total energy of 227 kWh ton^{-1}). This value is lower than those (404 or 785 kWh ton^{-1}) of 1S or 2S-AD processes fed with food waste, respectively (Chu et al., 2008; Thi et al., 2016). This may be attributed to difference of carbon content of the feed waste, experimental scale or operating conditions such as temperature, OLR or biomass concentration in the digester.

To evaluate quantitatively beneficiaries of the 2S-AD process to be used for MSW disposal in HCMC, analysis of material balance of a large scale 2S-AD plant with capacity of 5,200 tons d^{-1} of biowaste is presented in Figure 8. It is assumed that all quantity of biowaste (5,200 tons day^{-1}) is disposed of using the 2S-AD plant to produce electricity and fertilizers, and the plant will be run at OLR of 2.5 - 3.8 kg VS m^{-3}.d^{-1} and at the other operating conditions (temperature, ratio of digestate to biowaste and SRT) that are similar to those of the pilot study. The calculation of its material balance (water, solids and nutrients) and energy

production are based on results obtained from this study. The use of the membrane up-concentration process using ultra or micro filtration (UF or MF) followed by reverse osmosis process is a proficient option for nutrient recovery in the 2S-AD plant (Malamis et al., 2013; Drosg et al., 2015). Figure 8 showed that the plants can daily generate about electricity of 552 MWh, thermal energy of 630 MWh and produce 1100 tons of both organic solid and liquid fertilizers containing 16.1 tons of ammonium-N, 11.4 tons of organic-N, 2.1 tons of TP and 89 tons stabilized organic matter (non biodegradable VS).

Figure 8

4. CONCLUSION

The pilot 2S-AD plant illustrated the high performance of COD and VS removal for biosolids separated from MSW in Ho Chi Minh City. The high biogas production yield of the pilot plant obtained at OLR of 2.5 - 3.8 kg $VS.m^{-3}.d^{-1}$ and temperature of $33 - 35^{o}C$ was much higher than that of the 1S-AD process from the previous studies. High build-up of tVFA concentration at OLR of 3.8 kg $VS.m^{-3}.d^{-1}$ in the fermenter did not inhibit the methanogenesis process. High ratio of NH_4^+-N to the feed TKN and high ammonia concentration coupled with high stabilized organic matters shown the potential use of the digestate as valuable fertilizer or materials for soil conditioning.

Advanced Waste Treatment Technology, VNU-HCM.

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Table 1. Characteristic of the feed biowaste

Parameter	Feed biowaste
Volatile solids (VS), % dry weight	20.1 ± 4.1, n = 49
Total solids (TS), % wet weight	82.5 ± 6.3, n = 55
VS:TS ratio, g.g^{-1}	0.82 ± 0.06, n = 49
Total chemical oxygen demand (tCOD), g.L^{-1}	233 ± 70, n = 55
tCOD:VS ratio, g.g^{-1}	1.37 ± 0.42, n = 49
Total Kjeldalh nitrogen (TKN), g.L^{-1}	8.22 ± 2.45, n = 41

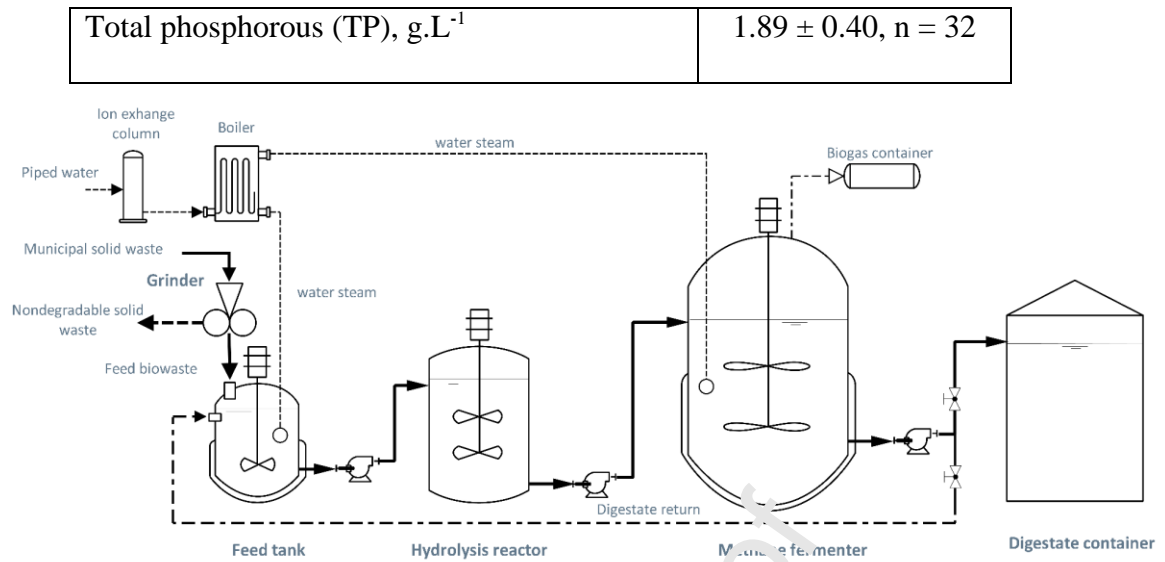| Total phosphorous (TP), g.L⁻¹	1.89 ± 0.40, n = 32

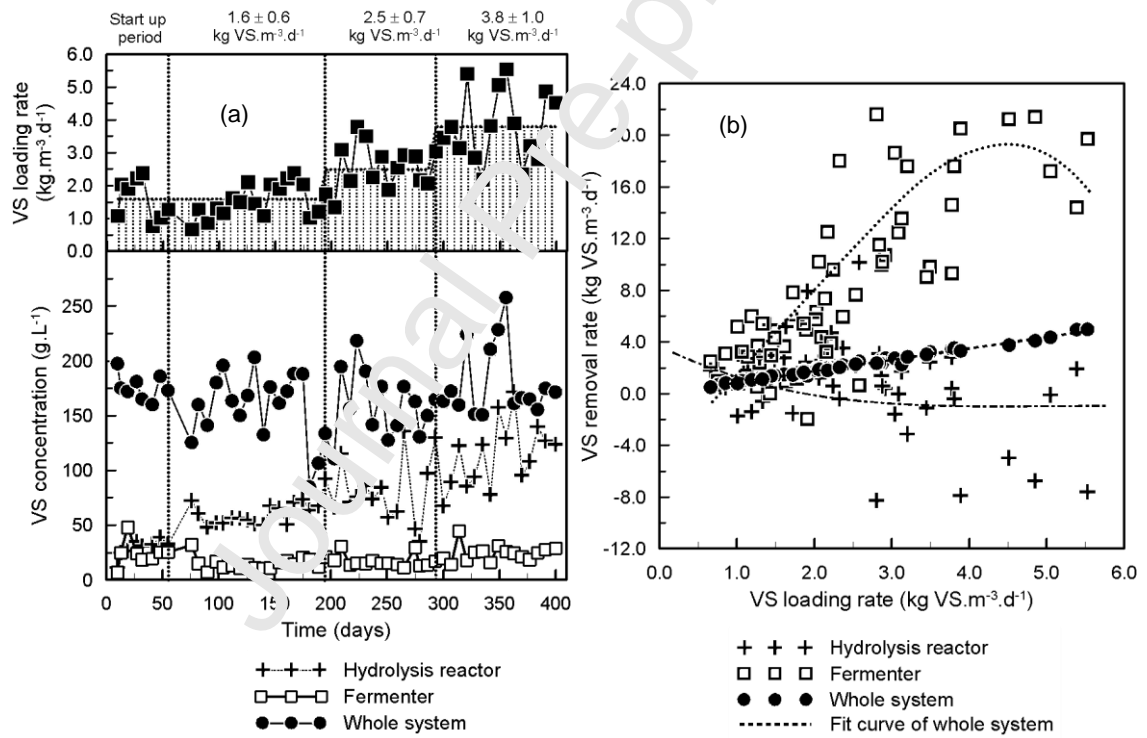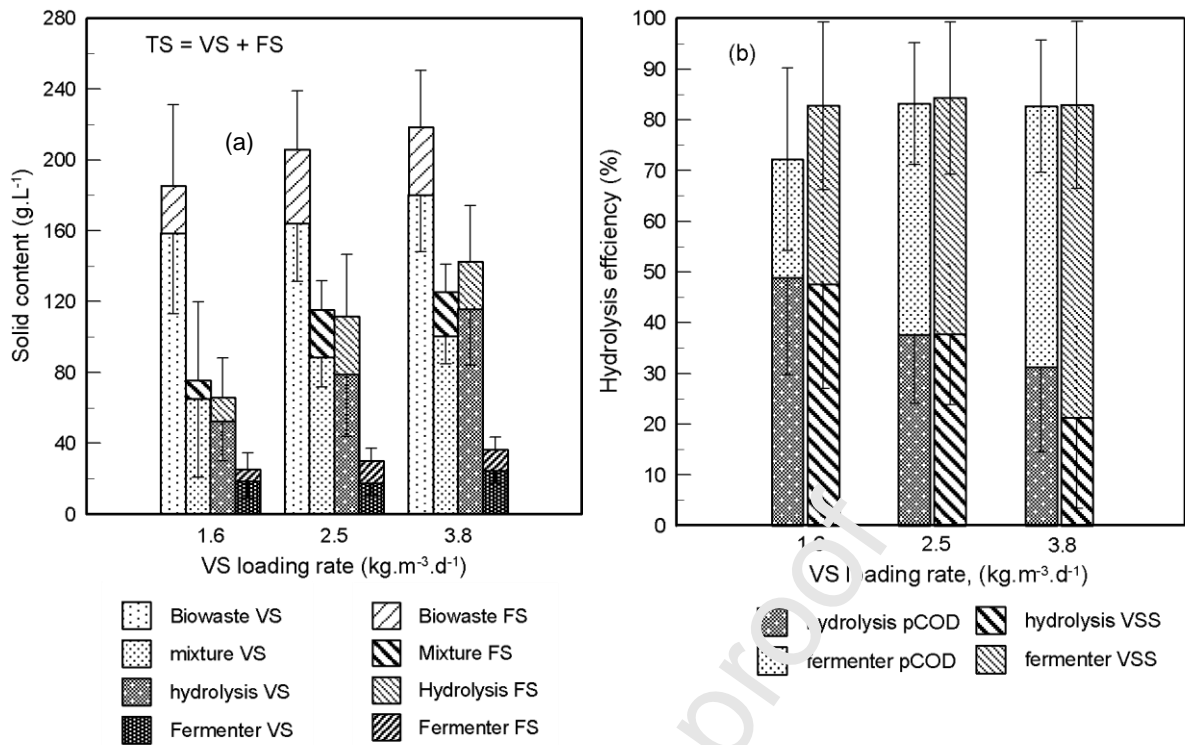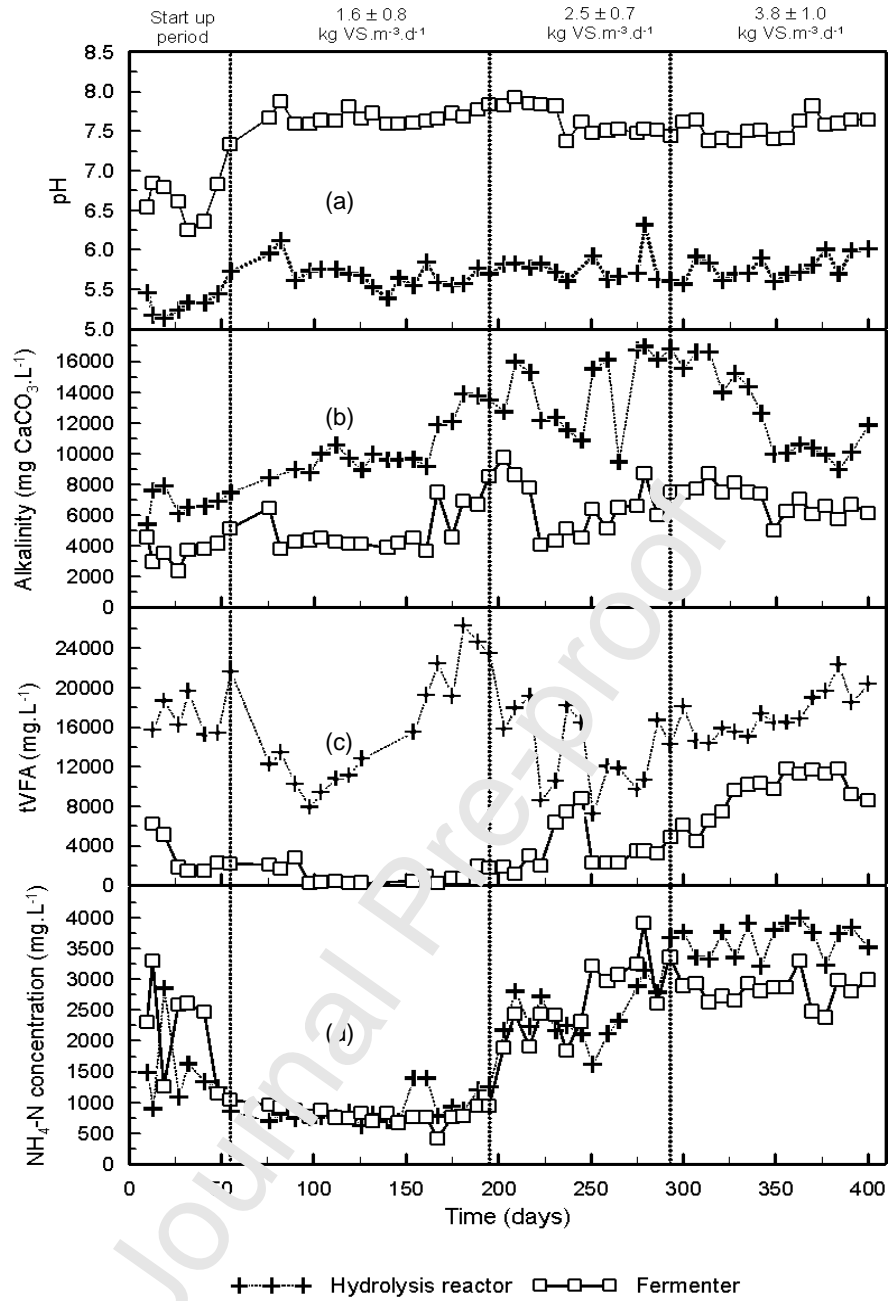**Figure 1**. 2S-AD pilot plant for municipal biosolids

Figure 2. Time course of (a) VS and (b) average VS and FS at different OLRs

Figure 3. (a) TSS and VSS concentrations and (b) Hydrolysis efficiency of hydrolysis reactor and methane fermenter at various average OLRs.

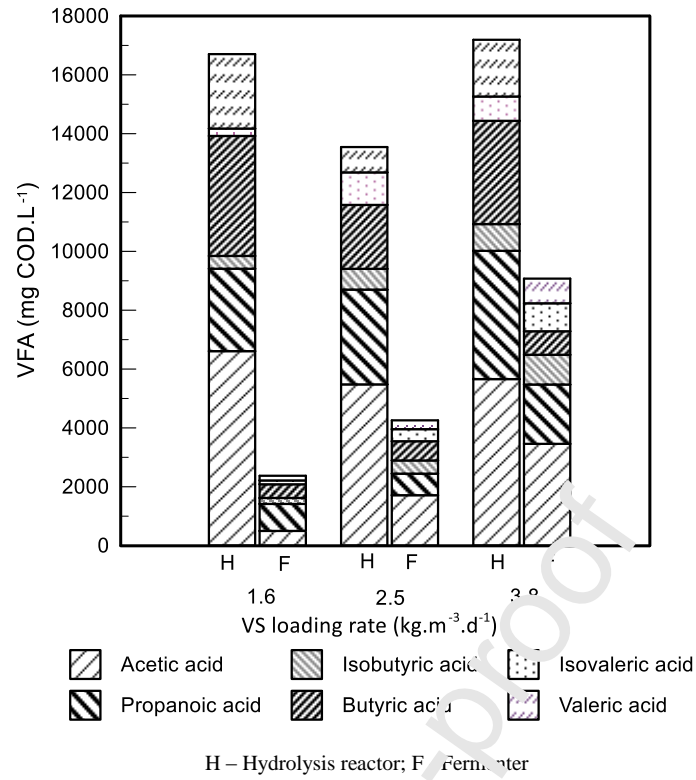**Figure 4.** Time courses of (a) pH, (b) alkalinity, (c) tVFA and (d) NH_4^+-N

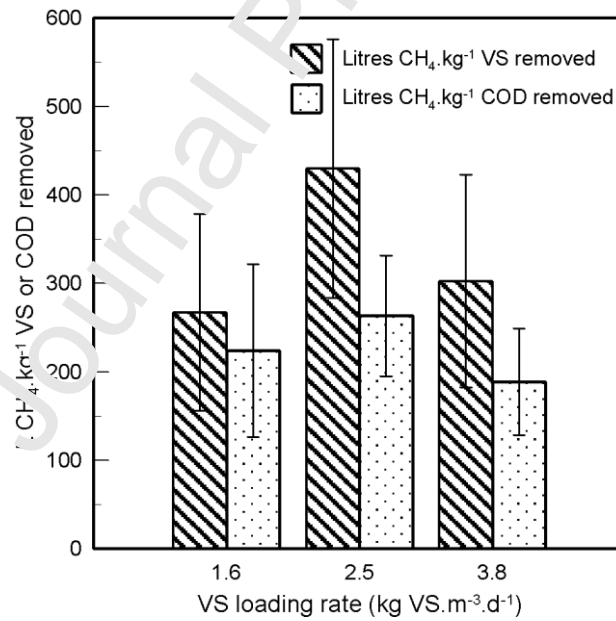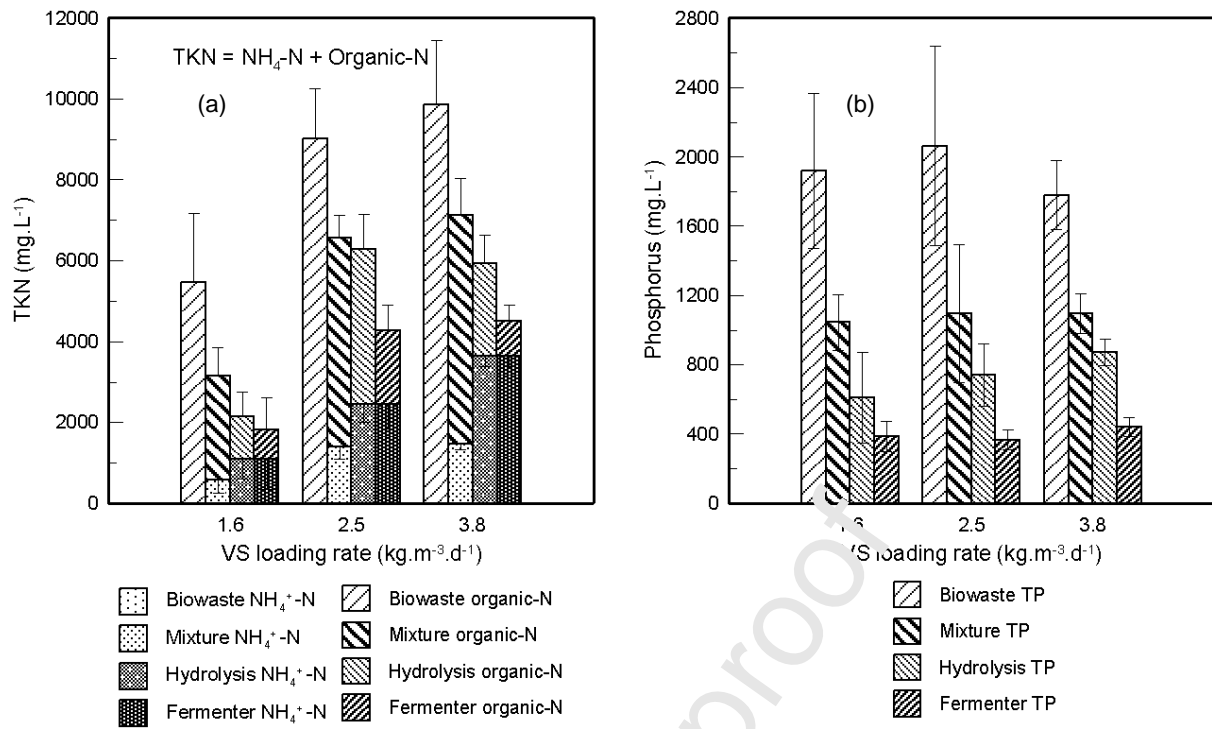**Figure 5.** Change of VFAs concentration at various average OLRs.
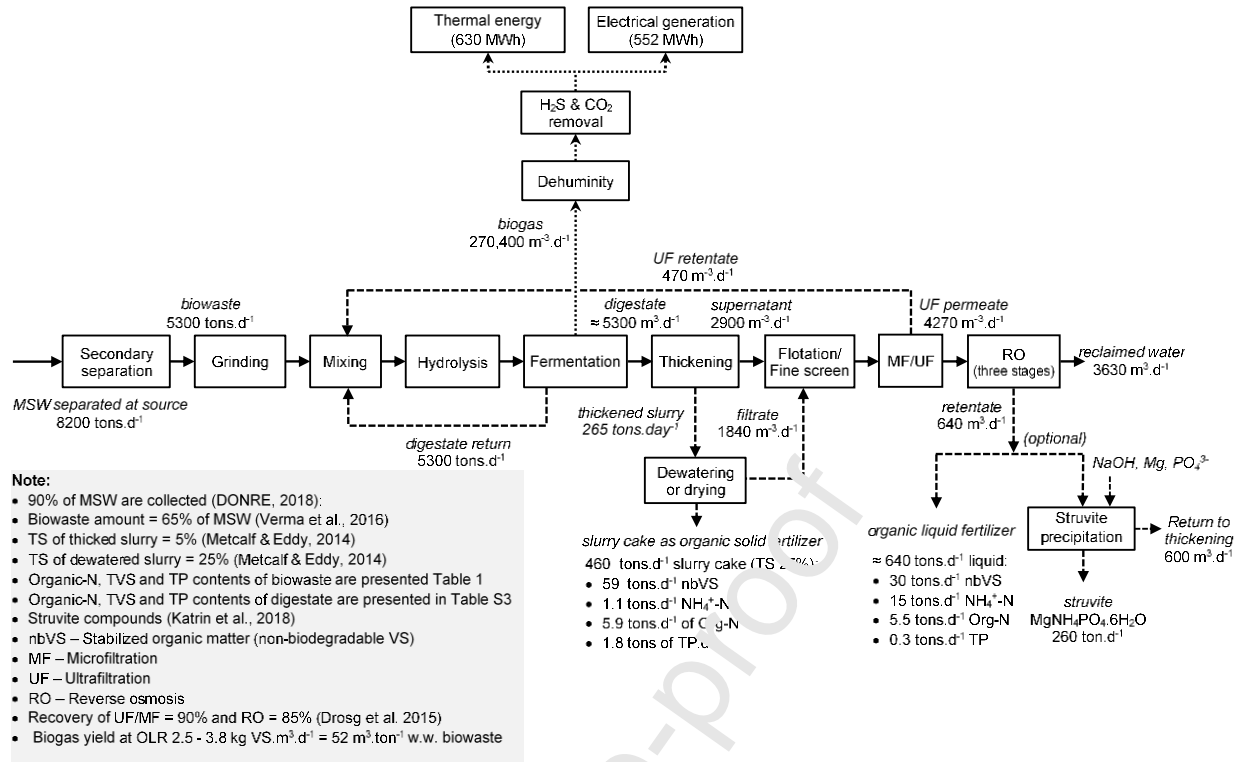

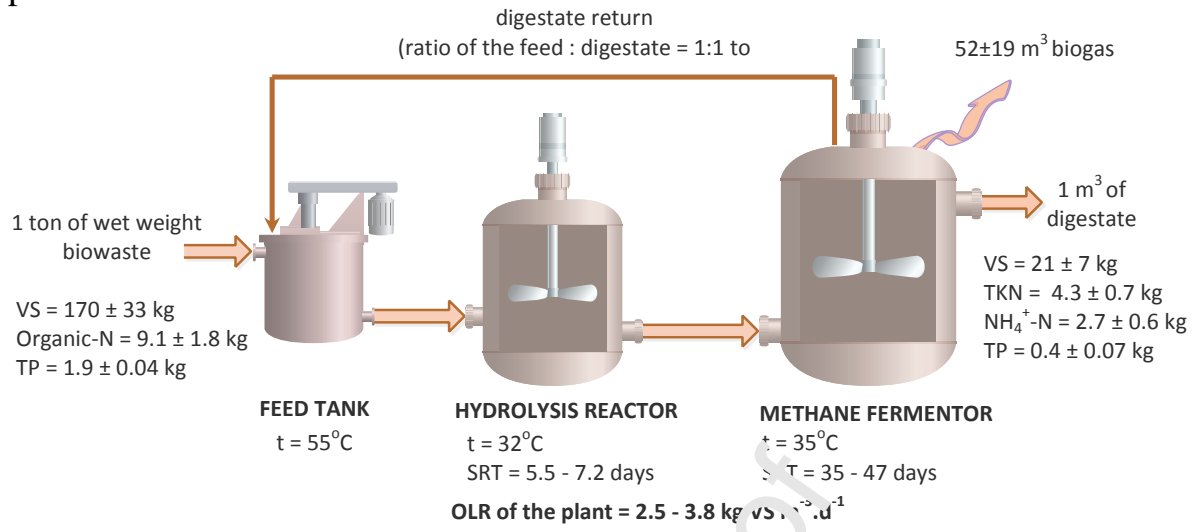**Figure 6** Biogas production rate versus the various OLRs

Figure 7. TKN (a) and total phosphorous (b) concentrations of feed, mixture, hydrolysis and digestate at various OLRs

Figure 8. Schematic diagram of the proposed large scale 2D-AS plant for municipal solid waste disposal in HCMC

Graphical abstract

digestate return
(ratio of the feed : digestate = 1:1 to

52±19 m^3 biogas

1 ton of wet weight
biowaste

VS = 170 ± 33 kg
Organic-N = 9.1 ± 1.8 kg
TP = 1.9 ± 0.04 kg

FEED TANK
t = 55°C

HYDROLYSIS REACTOR
t = 32°C
SRT = 5.5 - 7.2 days

METHANE FERMENTOR
t = 35°C
SRT = 35 - 47 days

1 m^3 of
digestate

VS = 21 ± 7 kg
TKN = 4.3 ± 0.7 kg
NH$_4^+$-N = 2.7 ± 0.6 kg
TP = 0.4 ± 0.07 kg

OLR of the plant = 2.5 - 3.8 kg VS m^{-3}.d^{-1}

HIGHLIGHTS

- High build-up of total propanoic acid concentration and total volatile fatty acid in the fermenter of the 2S-AD process did not cause failure of the methanogenesis.

- The average biogas yield obtained from the pilot 2S-AD plant using biowaste of Ho Chi Minh City was higher than that of the previous studies of 1S-AD process.

- The 2S-AD plant potentially produces the thermal energy and electricity, and organic fertilizer from recovery of nutrients in the digestate.