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*Title page

Effect of organic loading rate on anaerobic digestion of pig manure: methane production, mass flow, reactor scale and heating scenarios

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Abstract: The effect of organic loading rate (OLR) with total solid (TS) control (3%-8%) on the performance of anaerobic digestion of pig manure (PM) using completely stirred anaerobic reactor was investigated. Based on the lab data, how OLR affects mass flow, construction scale and heating requirement in a farm-scale biogas plant was calculated. And three scenarios of typical reactor-heating technology were comparatively analyzed. The optimal OLR was 1.89 g volatile solid (VS) /(L·d) with methane yield of 438.38 mL/gVS in the lab condition. The lower OLR, the larger amount of water and energy consumption, lower methane production and larger amount of liquid digestate was observed. Thus, the reactor with low OLR was suitable in tropical regions with the main target of disposing PM and fertilizer production. High OLR has advantage in the investment, but owns risk of instable process for a long-term run. In our study, among the three heating supply scenarios, biogas boiler was the best option for the designed biogas plant with the given breeding scale under moderate OLR. Combined heat and power (CHP) has potential advantage for the biogas plant under high OLR.

Key words: anaerobic digestion; pig manure; organic loading rate; mass flow; biogas plant

*Cover Letter

Dear Editor,

I would hereby like to submit our manuscript entitled "**Effect of organic loading rate on anaerobic digestion of pig manure: methane production, mass flow, reactor scale and heating scenarios**" for possible publication in ***Journal of Environmental Management*** as an original research paper.

All authors have read and approved this version of the article, and all the authors mutually agree that it should be submitted to ***Journal of Environmental Management***. The manuscript was not previously submitted to ***Journal of Environmental Management*** and no part of this paper has been published or submitted elsewhere. It is the original work of the authors. The language has been checked by a native tongue speaker with expertise in the field.

Important original contributions include the following:

- This work for the first time investigated how the effects of different OLRs on the mass flow and energy requirement observed in lab-scale operation are interpreted in large-scale without building a real large-scale reactor.
- Results showed that The optimal OLR was 1.89 gVS/(L.d)(5%TS) with high methane production and limited digestate production, stable process, moderate construction scale and cost, low operation cost and moderate self-heating energy compulsion. The reactor with low OLR was suitable in tropical regions with the main target of disposing PM and fertilizer production. High OLR has advantage in the investment, but owns risk of instable process for a long-term run. Among the three heating supply scenarios, biogas boiler was the best option for the biogas plant with the given breeding scale under moderate OLR. Combined heat and power (CHP) has potential advantage for the biogas plant under high OLR.
- This study may offer appropriate considerations for design of the commercial biogas plant

The current manuscript consists of 5 figures and 3 tables.

We believe that the current study would be an interesting topic for ***Journal of Environmental Management*** readers. I look forward to receiving your evaluation.

Sincerely yours,

Na Duan

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Dear Editor,

Thanks for yours and reviewers' constructive comments on our manuscript entitled "Effect of organic loading rate on anaerobic digestion of pig manure: methane production, mass flow, reactor scale and heating scenarios" **(Manuscript ID: JEMA-D-18-03867).** We have carefully considered all comments and have made extensive revisions accordingly. Please find our detailed response and revisions as following; each change is marked in blue color.

Responds to the reviewer's comments:

Reviewer #1:

The manuscript is of general interest; however, some important revisions are required before it can be considered for publication as follows:

1. There are plenty of grammatical and syntax errors throughout the manuscript which is a turn-off. Examples are, Page 5, L 11 "However, there is few studies linking…", P5, L15 "Thus, this is extreme important because …", and many more. A thorough English revision is required. This is a Must should the paper be accepted for publication.

Answer: We have carefully polished the language in the revised manuscript.

2. Please include a photo of the experimental set-up used, a clear schematic presentation will also do.

Answer: Thanks for your constructive suggestion. I have added some information about the schematic of the reactor in the revised paper.

The schematic of the reactor was the same as previously described (Zhang et al., 2018). (Page 6, Line 21- Page 7, Line 1)

Zhang, D.J., Duan, N., Tian, H.L., Lin, C., Zhang, Y.L., Liu, Z.D., 2018. Comparing two enhancing methods for improving kitchen waste anaerobic digestion: Bentonite addition and autoclaved de-oiling pretreatment. Process Saf. Environ. 115, 116-124.

3. Please define abbreviations the first time they are used; Combined Heat and Power (CHP).

Answer: We have defined the abbreviation of CHP at the first time in the revised

manuscript (Page 11, Line 4-5).

.

4. Please change "4. Conclusion" to "4. Conclusions".

Answer: We have changed “4. Conclusion” to “4. Conclusions” in the revised manuscript.

5. Please also report on the biochemical methane potential (BMP) of the substrate used. The following article should be used:

Jingura, R.M. and Kamusoko, R., 2017. Methods for determination of biomethane potential of feedstocks: a review. Biofuel Research Journal, 4(2), pp.573-586.

Answer: Thank you for your suggestion. We have calculated the biochemical methane potential using the method described in the literature as follows. We also added the results in the revised manuscript (Page 11, Line 12).

Jingura, R.M., Kamusoko, R., 2017. Methods for determination of biomethane potential of feedstocks: a review. Biofuel Res. J. 4(2), 573-586.

6. Three scenarios (i.e., solar energy, biogas boiler, and CHP) were analyzed and conclusions were drawn. However, the issue of sustainability has not been well discussed. Environmental sustainability is of utmost importance in the growing biofuel industry and should be included in the manuscript and discussed. The following articles should also be included and discussed:

Lynch, J.M., 2017. The Sustainability of Biofuels. Biofuels and Bioenergy, pp.261-272.

Rosen, M.A., 2018. Environmental sustainability tools in the biofuel industry. Biofuel Research Journal, 5(1), pp.751-752.

Ali, S., Xu, H., Al-amin, A.Q. and Ahmad, N., 2018. Energy sources choice and environmental sustainability disputes: an evolutional graph model approach. Quality & Quantity, pp.1-21.

Answer: We have analyzed the sustainability and added the three references in the revised manuscript.

Sustainability is a vital factor while selecting the heat supply scenarios, which mainly include economic sustainability and environmental sustainability (Lynch, 2017). In terms of the economic cost, the investment of the biogas boiler, solar collector and electricity generator under 5%TS was about $\$0.63\times10^4$, 1.88×10^5 and 3.14×10^5, respectively. Environmental sustainability has been widely investigated using

different assessment methods, such as life cycle assessment, energy, exergy (Rosen, 2018). Specifically in energy system, energy production and utilization should avoid energy-environment conflict (Ali et al., 2018). Considering the environmental sustainability, the three scenarios are all have less environmental impact in comparison with heating using fossil energy. CHP has great conversion efficiency, thus, can achieve maximum greenhouse gases emission saving. However, the environmental costs caused by electricity generation using fossil fuel were often ignored, and the environmental benefits of electricity generation using biogas were also covered by its economic profit. Therefore the environmental value of electricity generation using biogas is not fully reflected and the technology competitiveness is not fully realized in China. Thereby, considering the energy efficiency and cost, biogas boiler was the best option for the biogas plant with the given breeding scale under moderate OLR. And the rest of the biogas can be used as domestic fuel for pig farms and neighboring households. However, for the CHP, because of the high efficiency and high profit from electricity, so in the long run, it may be a good choice for the biogas plant under high OLR. The solar collector can be used in tropical areas to enhance the heat availability and reduce the energy requirements, simultaneously reduce the collector areas and cost.

Lynch, J.M., 2017. The Sustainability of Biofuels, in: Love, J., Bryant, J.A. (Eds.), Biofuels and Bioenergy, John Wiley and Sons Ltd. UK, pp.261-272.

Rosen, M.A., 2018. Environmental sustainability tools in the biofuel industry. Biofuel Res. J. 5(1), 751-752.

Ali, S., Xu, H., Al-amin, A.Q., Ahmad, N., 2018. Energy sources choice and environmental sustainability disputes: an evolutional graph model approach. Qual. Quant. https://doi.org/10.1007/s11135-018-0775-9

Reviewer #2:

1. The manuscript must be improved in terms of English language.

Answer: Thanks for your precious suggestion. We have carefully reviewed the language and clarity throughout the revised manuscript.

2. Abstract: "The effect of organic loading arte …" Please mention here the OLRs you tested. It helps readers, who only read the introduction, to better understand what you are talking about.

Answer: We have revised the sentence as you suggested:

"The effect of organic loading rate (OLR) with total solid (TS) control (3%-8%) on

the performance of anaerobic digestion of pig manure (PM) using completely stirred anaerobic reactor was investigated." (Page 2, Line 1-3)

3. Abstract: "how OLR affect mass flow, heating requirement and …" should change to "how OLR affects …"

Answer: We have written the sentence as you suggested:

"Based on the lab data, how OLR affects mass flow, construction scale and heating requirement in a farm-scale biogas plant was calculated." (Page 2, Line 3-5)

4. Abstract, Line 4: What do you mean by supply change here?

Answer: We have revised the sentence to make it clear.

"Based on the lab data, how OLR affects mass flow, construction scale and heating requirement in a farm-scale biogas plant was calculated. And three scenarios of typical reactor-heating technology were comparatively analyzed." (Page 2, Line 3-6)

5. Abstract: "… biogas boiler was the best option…"It should be mentioned here this option is suitable for the plant you designed. A large number of researchers have already shown that CHP would be the best alternative in biogas plants because it can satisfy plant's heating requirement and simultaneously generate electricity which can substitute fossil based electricity.

Answer: A good suggestion. This point was proposed in this study, and we have rewritten this point as follow:

"In our study, among the three heating supply scenarios, biogas boiler was the best option for the designed biogas plant with the given breeding scale under moderate OLR."

6. Introduction needs to be rewritten to better present the idea behinds the work and the novelty of this research.

Answer: We have revised the introduction part in the revised manuscript, and two references added as follows:

Li, J., Kong, C.X., Duan, Q.W., Luo, T., Mei, Z.L., Lei, Y.H., 2015b. Mass flow and energy balance plus economic analysis of a full-scale biogas plant in the rice-wine-pig system. Bioresour. Technol.193, 62-67.

Dong, F.Q., Lu, J.B., 2013. Using solar energy to enhance biogas production from livestock residue-A case study of the Tongren biogas engineering pig farm in South

China. Energ. 57,759-765.

7. Page 3 line 4 "However, most farms do not have adequate manure management measures to handle .." change to "However, most farms do not have adequate manure management strategies to handle .."

Answer: We have written the sentence as you suggested:

"However, most farms do not have adequate manure management strategies to handle the large amounts of manure and prevent environment pollution." (Page 3, Line 4-5)

8. Page 3 line 9: "… produce valuable by-products such as clean energy (biogas) and high-quality fertilizers …" It depends on the substrate which is introduced to plant. For instance, if sludge or organic fraction of municipal solid waste are anaerobically digested the quiality of the digestate is a matter of issue.

Answer: We agree with you. The only substrate related in this study is animal manure; we do not have sludge or any other organic fraction of municipal solid waste. A large number of researchers have already shown that the digestate from animal manure AD system can be used as fertilizers.

10. Table 1. Some important parameters can be added to this table. For example how much were Total N and total ammonium nitrogen?

Answer: We have added the total Kjeldahl nitrogen (TKN) in Table 1. We did not measure the ammonium nitrogen of the substrate. However, we measured the ammonium nitrogen of the digestate in the AD process.

11. Page 6, line 13. Do you have Characteristics of inoculum? It can be added to Table 1.

Answer: A good suggestion. We have added the characteristics of inoculum in Table 1.

12. Page 6, line 20. Did you sieve PM before feeding the reactor?

Answer: We did not sieve the PM before feeding the reactor. Because we did not use pump to feed, we just feed manually.

13. Page 7, line 9: ' by multiplying the organic nitrogen by 6.25'. Please check this coefficient in the literature. It must be 0,625

Answer: We have checked the related literature. The coefficient is 6.25.

14. Section 2,3. Analytical method. Many parameters like TKN, COD, Trace elements, etc have been mentioned here but they have not been presented in the manuscript? Please provide such information in proper places within the text or tables.

Answer: We have measured TKN, lipids, crude fibers and element for the raw material (pig manure), and all the data has been listed in Table 1. We measured TCOD, SCOD and total ammonia concentration in the AD process. All the data has been stated in Fig.2 and Fig. 3. The analysis of the data was in the section of 3.1.2 and 3.1.3.

15. Eq.1. When you use TAN (total ammonium nitrogen NH4-N) you will have free ammonia nitrogen(NH3-N) and not free ammonia. Please correct it in the equation.

Answer: We have revised equation in the revised manuscript.

16. Page 9 line 1. Change ‘Heat losses’ to ‘Heat is lost …’

Answer: We have written the sentence as you suggested:

“Heat is lost through the biogas, the reactor and evaporation, which may increase the heat consumptions ” (Page 9, Line 6-7)

17. Table 2. ‘CH_4 production (mL/gVS)’ can be replaced by the term ‘Specific methane yield (mL/gVS)’.

Answer: We have used “Specific methane yield (mL/gVS)” replacing “CH_4 production (mL/gVS)” in Table 2.

18. Page 11 line 13. “The maximum methane yield …” this comparison is not correct. First of all, your substrate was PM while those referenced were swichgrass and pig slurry. On the other hand, all VS cannot change to biogas and it depends on the degradability of the substrate. For instance wheat straw has a high VS but due to its rigid structure it cannot be degraded without pretreatment. If the yield was expressed based on COD, the comparison would make sense.

Answer: Thanks for your suggestion. In general, when we compared the methane yield with different treatments or literatures, methane yield based on VS or COD are used. In this study, we will use the lab-experiment data to investigate how would be the effects of OLR on AD process performance in large-scale biogas plant. For large-scale biogas plant, VS is very easier than COD to measure. Thus, in present study, methane yield based on VS was used to compare with different treatments and

other literature.

19. Page 12 line 2. "Thus, the incomplete degradation and overload inhibited …" Please show the total VFA and pH changes in one graph. The selected OLRs were not so high that can cause inhibition. This graph can show VFA changes and pH changes during the experiment to see if any inhibition was occurred.

Answer: Thanks for your precious comment. We understand that VFA is an important index for anaerobic digestion. Although the VFA was not analyzed in this study, comparative analysis of other important parameters including ammonia, biogas production and composition, as well as organic matter reduction during the AD process have been studied. So that we can synthetically assess the AD performance through the above mentioned parameters

20. Page 13 line 6. In this study OLR was set by changing the TS of the substrate. When TS of the substrate increases, it is clear that the total N in the substrate increases also. Therefore, we expect to have higher TAN in the digestate but the TAN and FAN presented in the Figure 3 do not show any sign of inhibition. Specially, your reactors worked under mesophilic condition and under such circumstances the risk of free ammonia inhibition is lower. I suggest to present the TAN and FAN of the substrate and digestate. It helps reader to better understand what has happened in your reactors.

Answer: Thanks for your suggestion. As shown in Table 1, we have added the TAN of the inoculum and the TKN of the PM. Although the TAN and FAN of the PM was not analyzed in this study, the change of TAN and FAN in the whole process has been evaluated. Thereby the reader can obtain the changing trend of TAN and FAN concentration in the whole process with different OLRs.

21. Page 15 line 9. "Specifically, the total …" It is not clear why energy consumption of 3%TS was higher than others.

Answer: The conclusion can be obtained from Table 3. Table 3 has listed the heat requirements of different OLRs. The lower OLR, the larger amount of water consumption and liquid diegstate, as well as larger reactor volume was observed. Thereby, more energy will be consumed at 3% TS.

*Highlights (for review)
Click here to view linked References

Highlights

1. How organic loading rate affect anaerobic digestion of pig manure.
2. The optimal organic loading rate of pig manure was 1.89 gVS/L.d(5%TS).
3. Organic loading rate has significantly affected mass flow and heating requirement.
4. Different heating supply scenarios have been comparatively analyzed.
5. Low organic loading rate was suitable for operation in tropical regions.

*Manuscript
Click here to view linked References

Effect of organic loading rate on anaerobic digestion of pig manure: methane production, mass flow, reactor scale and heating scenarios

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Abstract: The effect of organic loading rate (OLR) with total solid (TS) control (3%-8%) on the performance of anaerobic digestion of pig manure (PM) using completely stirred anaerobic reactor was investigated. Based on the lab data, how OLR affects mass flow, construction scale and heating requirement in a farm-scale biogas plant was calculated. And three scenarios of typical reactor-heating technology were comparatively analyzed. The optimal OLR was 1.89 g volatile solid (VS) /(L·d) with methane yield of 438.38 mL/gVS in the lab condition. The lower OLR, the larger amount of water and energy consumption, lower methane production and larger amount of liquid digestate was observed. Thus, the reactor with low OLR was suitable in tropical regions with the main target of disposing PM and fertilizer production. High OLR has advantage in the investment, but owns risk of instable process for a long-term run. In our study, among the three heating supply scenarios, biogas boiler was the best option for the designed biogas plant with the given breeding scale under moderate OLR. Combined heat and power (CHP) has potential advantage for the biogas plant under high OLR.

Key words: anaerobic digestion; pig manure; organic loading rate; mass flow; biogas plant

1 Introduction

In China, the manure quantities increase rapidly in a few concentrated areas as a result of large-scale and intensive livestock production moving towards (Deng et al., 2017). However, most farms do not have adequate manure management strategies to handle the large amounts of manure and prevent environment pollution (Jiang et al., 2011). In response to the problem, the Chinese government has implemented a series of management and incentive policies. All these regulations recognize that biogas technology is an environment-friendly solution to reduce environmental pollutions and produce valuable by-products, such as clean energy (biogas) and high-quality fertilizers. Besides, because of the reliability of anaerobic digestion (AD) technology and growing awareness of environmental protection from farm owners, the number of large- and medium-scaled biogas plants built in livestock farms increases with the average annual growth rate of about 9.15% from 2001 to 2006 and 21.53% from 2006 to 2010 (Yang et al., 2012). In China, nearly more than 90% of the biogas plants are built up for manure management (MOA, 2011). Particularly, China is rich in resource of pig manure (PM) with pig rearing amount of 465 million heads in 2014 (Yu et al., 2015), thereby, due to its abundance, inherent buffering and containing a wide range of nutrients for the growth of anaerobic microorganisms, PM has a great potential for scaled biogas generation (Regueiro et al., 2012). In addition, a variety of anaerobic reactors have been developed and applied in China including continuous stirred tank reactor (CSTR), up-flow anaerobic sludge blanket (UASB), up-flow solids reactor (USR), and plug flow

reactor (PFR). Among which, the biogas plants with CSTR and USR technologies are prominent, comprising 65% of the total plants (Chen et al., 2012).

The AD process is complex, affected by many factors including the type and composition of the substrate, microbial composition, temperature, organic loading rate (OLR), pH and reactor configuration, etc. Many studies have reported that temperature is one of the most crucial factors influencing the microbial community, process kinetics, stability, substrate utilization rates, and methane yield (Bouallagui et al., 2009; Dela-Rubia et al., 2002; Riau et al., 2010). The AD process is commonly operated in the mesophilic temperature, considering the process stability, energy expense and microorganism sensitivity (El-Mashad et al., 2003; Fernandez et al., 2008; Ward et al., 2008).

OLR is also considered as an important operational parameter. The high OLR means high treatment capacity and methane yield, but it also may lead to overloading and thereby causing process instability, and even system failure. Li et al. (2015a) reported operating limits of OLR when livestock manure was mono-digested. Guo et al. (2013a) indicated that there might be a risk of acidification if the reactor operated below 38 ^{o}C at high OLRs. Thus, there is a need for a strategy to find a suitable OLR for achieving a stable and optimum anaerobic digestion of pig manure.

Regarding PM, its carbon to nitrogen (C/N) ratio was lower relative to the optimum C/N ratio (20:1-30:1) existing ammonium inhibition risk. Therefore, several studies, in batch or continuous experiments, have indicated the benefits of co-digestion with other

wastes (Astals et al., 2013; Ren et al., 2014; Zhang et al., 2014), however, PM is still commonly used as the sole feedstock to produce biogas for most farms in China. Actually, for co-digestion, the sources, transportation, and supply stability of the co-substances should be considered to keep a long run of the biogas plant.

Some existing biogas plants have been operating at low efficiency owing to various technical barriers, such as running at lower or higher OLR, fluctuant digestion temperature, lack of knowledge about maintenance and monitoring, etc.(Deng et al., 2017). Thus, the appropriate operating conditions are essential for the commercial and sustainable development of AD technology treating PM. Most of the AD studies are in lab-scale (Guo et al., 2013a; Ni et al., 2017; Tsapekos et al., 2017) or evaluat the operational performance of existed biogas plants (Dong and Lu, 2013; Li, et al., 2015b). The results and discussion from lab-scale indeed could provide insight into large-scale development. However, there are few studies linking the data from lab-scale to give design consideration for a farm-scale biogas plant, in particular, how the effects of different OLRs on the mass flow and energy requirement observed in lab-scale operation be interpreted in large-scale without building a real large-scale reactor has never been investigated. Thus, this is extremely important because it can (i) help to know the input, output and detailed mass flow and energy consumption, (ii) investigate different scenarios to provide appropriate considerations for the design of the commercial biogas plant.

In the present study, anaerobic digestion of PM was performed with different OLRs

operation using total solid (TS) control (3%-8%) at mesophilic temperature (35±1°C) in lab-scale. The data obtained was then used as input for validating the results in large-scale. The objective is to: (1) investigate how would be the effects of OLR on AD process performance in large-scale based on the simple modeling; (2) simulate the variations of mass flow, biogas plant scale and heating requirement under different OLR_S in large-scale using lab-scale data. (3) analyze different scenarios of heat supply and sustainability.

2 Materials and methods

2.1 Feedstock and inoculum

The fresh PM as the substrate for the biogas production was collected from Beilangzhong pig farm (Shunyi District, Beijing, China), and then was delivered to the laboratory and stored at 4°C until use. The PM had a high proportion of carbohydrates (over 60%TS), followed by the crude protein and crude lipids. The C/N ratio was approximate 10.64. The inoculum was obtained from a centralized biogas plant treating PM anaerobically and inoculated with PM for two weeks prior to use. The characteristics of the PM and inoculum are presented in Table 1.

2.2 Reactors and experimental set-up

Six identical 20-L semi-continuous stirred-tank reactors (CSTR) with a working volume of 17.5-L were used. Reactors were operated under a mesophilic condition at 35±1 °C by circulating water from a heated water bath (SY-200, Changfeng Yi Qi Yi Biao Company, Beijing, China) through a water jacket surrounding the reactor. The

schematic of the reactor was the same as previously described (Zhang et al., 2018). The reactors (R1-R6) were manually fed once per day with TS concentration of 3% to 8%, while their OLRs were set at 1.13 to 3.03gVS/L·d (Table 2). The PM was diluted with tap water to the target feeding loads and an equivalent volume of digestate was purged once a day. The reactor was stirred at 60 rpm and the hydraulic retention time (HRT) was set at 22 days. Each reactor was generally operated for nearly 2 HRTs reaching a steady state with stable daily methane yield, pH and COD removal.

2.3 Analytical methods

Total Kjeldahl Nitrogen (TKN), lipids and crude fibers were measured following the standard procedure of Kjeldahl method, Soxhlet method and cellulose analyzer (Model A220, ANKOM Technology Corporation, NY, USA), receptively. Protein was estimated by multiplying the organic nitrogen by 6.25 and carbohydrates were estimated by subtracting the amount of protein and lipids from volatile solids. Elemental analysis was conducted using an element analyzer (Vario MICRO Cube, Elementar Analysen systeme GmbH, Donaustraße, Germany).

The biogas yield was automatically measured using a gas flow meter (LML-1, Changchun Automotive Filter Co., LTD, Changchun, China) and biogas composition was analyzed every day using the gas chromatograph (Model 1400, Agilent Technologies, USA). The biochemical methane potential was calculated based upon the main organic compositions as described (Jingura and Kamusoko, 2017). Digestate samples were collected from the reactors every three days. The TS, VS, total chemical

oxygen demand (TCOD), soluble chemical oxygen demand (SCOD) concentration and total ammonia nitrogen (TAN) concentration were determined following the guidelines given by the standard methods (APHA, 2005). The free ammonia nitrogen (FAN) concentration was calculated based on TAN concentration and experimental conditions including temperature and pH using the following Eq.(1) (Hansen et al., 1998):

$$\frac{FAN}{TAN} = \left(1 + \frac{10^{-pH}}{10^{-\left(0.09018 + \frac{2729.92}{T(k)}\right)}}\right)^{-1} \quad \text{Eq. (1)}$$

where FAN is the concentration of free ammonia nitrogen in mg/L, TAN is the total ammonia nitrogen concentration in mg/L, pH is the pH value determined in the reactor, and T(k) is the temperature (Kelvin).

2.4 Mass flow calculation

In order to investigate the effects of OLR on the mass flow and construction scale for a commercial biogas plant, a hypothesized process of a pig farm with 5000 pigs annually on hand in Beijing of China and the annual manure treatment capacity of 2920t (collection coefficient of 80%) was used for calculation. Typically, a commercial biogas plant consists of different subsystems. The pre-treatment tank, anaerobic reactor and digestate storage tank are the principal facilities, and their volumes can be calculated based on the targeted OLR, HRT and storage time. Based on the lab-scale data, the mass flow and total solid flow of three major components (pre-treatment, anaerobic digestion, by-products utilization) of biogas plant under different OLRs were calculated.

2.5 Heating requirement and supply scenarios

The optimal and economical temperature for biogas production is around 35 ^{o}C, thereby, the biogas plant requires external heating to stabilize at the mesophilic range in moderate and cold climates. Besides that, the heating requirement also depends on the OLR and reactor volume. There are two heat demands in the farm-scale biogas plant, one is for preheating the influent in the pre-treatment tank and the other is for maintaining normal digestion temperature in the AD reactor. Heat is lost through the biogas, the reactor and evaporation, which may increase the heat consumptions. The total heating requirements were illustrated in Fig.1 and the heating requirement in winter was calculated using Eq.(2).

$$\sum Q = Q_1 + Q_2 + Q_3 + Q_4 + Q_5 \qquad \text{Eq. (2)}$$

where $\sum Q$ is total heating requirement of the farm-scale biogas plant (MJ/d); the definition of the Q_1, Q_2, Q_3, Q_4, Q_5 has been stated in Fig.1.

$$Q_1 = cm_1(T_1 - T_a) \qquad \text{Eq. (3)}$$

where c is the average specific heat of the influent (4.17kJ/(kg·^{o}C)); m_1 is the weight of the daily feeding influent (kg); T_1 is the heating temperature (considering the heat loss in the heating process, T_1(38 ^{o}C) should be higher than the desired temperature(35^{o}C)); T_a is the ambient temperature of Beijing in winter (0 ^{o}C).

Q_2 can be calculated as Eq.(4) to keep the temperature fluctuation of 1 ^{o}C.

$$Q_2 = c\rho V \Delta t \qquad \text{Eq.(4)}$$

where ρ is the density of the digestate in the anaerobic reactor (1.04 t/m^3); V is the effective volume of the anaerobic reactor (m^3); Δt is the temperature fluctuation of 1 ^{o}C.

Q_3 can be calculated by Eq. (5) (Guo et al., 2013b).

$$Q_3 = (1676 + 1772 \cdot \frac{1-f}{f})(V \cdot \gamma)(T - T_a) \quad \text{Eq. (5)}$$

where f is the methane content (%); γ is the volumetric methane production ($m^3CH_4/m^3/d$); T is the fermentation temperature (35°C).

Q_4 can be calculated as Eq. (6) (Guo et al., 2013b).

$$Q_4 = W_\omega \cdot [H_V + C_V \cdot (T - T_a)] \quad \text{Eq. (6)}$$

where W_ω is the mass flow rate of vapor from anaerobic reactor (kg/d); H_v is latent heat of vaporization(2415kJ/kg at 35 °C); C_v is the specific heat of vapor (1.886kJ/(kg·°C)). W_ω can be calculated as Eq. (7) and (8) (Guo et al., 2013b).

$$W_\omega = 0.804 \times \frac{V \cdot \gamma \cdot X_\omega}{f(1 - X_\omega)} \quad \text{Eq. (7)}$$

$$X_\omega = 1.27 \times 10^6 e^{\frac{-5520}{t+273}} \quad \text{Eq. (8)}$$

Q_5 can be calculated according to the heat transfer theory as Eq. (9).

$$Q_5 = 24 \times (T - T_a)/[\sum \frac{b_i}{\lambda_i \cdot S_i} + \frac{1}{h \cdot S_0}] \quad \text{Eq. (9)}$$

where the b_i is the thickness of the reactor base, enameled steel and insulation material (Extruded polystyrene) about 0.5m, 0.08m and 0.1m, respectively. The thermal conductivity (λ_i) of the reinforced concrete, insulation material and enameled steel is 1.3, 0.033 and 2.05W/m·°C, respectively (Pu et al., 2010). S_i is the area of the reactor bottom, wall and top (m^2), and S_0 is the surface area of the reactor (m^2). The height and the diameter of the anaerobic reactor should be assumed according to the lab experiment

results to calculate the S_i and S_0. The heat exchange coefficient of the environment is 23W/(m^{2}·°C) (Liu et al., 2012).

Based on the total heating requirements in winter, three scenarios of typical digester-warming technologies including biogas boiler, solar energy, combinedd heat and power (CHP) were analyzed to evaluate their energy efficiencies and economic cost.

3 Results and discussions

3.1 Process performance of the lab-scale experiment

3.1.1 Methane production

The results of the present study demonstrated that OLR had a great effect on methane production (Table 2). As the OLR increased, methane yields were varied (324.25-438.38 mL/gVS) corresponding to more than 73% of the theoretical maximum and not linear within the tested range. The maximum methane yield was achieved at the OLR of 1.89 gVS/L·d (5%TS), which was 11.81% and 35.20% higher than that with the lower OLR (1.13 gVS/L·d, 3%TS) and higher OLR (3.03gVS/L·d, 8%TS), respectively. As the OLR increased from 1.89 to 3.03 gVS/L·d, a decrease in methane production was observed, this was expected due to the incomplete degradation with a high level of feeding OLR_S. Consequently, the optimal OLR of PM was found to be 1.89 gVS/L·d (5%TS) under mesophilic conditions with HRT of 22 days.

The maximum methane yield, 438.38 mL/gVS, is relatively higher compared to that normally can be achieved, i.e.220-350 mL/gVS from PM (Ahn et al., 2009; Bonmati et

al., 2001) and comparable to the results of Chae et al. (2008) about 437 mL/gVS at 35 ^{o}C with the feeding load of 5%TS. The varying methane yields are due to the different chemical compositions of the PM and the AD operation process.

3.1.2 Organic matter reduction

As the OLR increased, the TCOD of the feeding, as well as the TS and VS content of the digestate increased obviously (Table 2 and Fig. 2A). The values of TS and VS removal at the steady stage were in the range of 53.25-57.43% and 59.61-66.22%, respectively. Similarly, SCOD concentrations of digestate increased with the increasing OLR. Thus, the incomplete degradation and overload inhibited the activity of methanogenic bacteria, which might occur in the reactors with higher OLRs (R5 and R6). The Values for TCOD reduction were in the range of 67.40-76.18%, and the SCOD removal was all over 80% (Fig.2B). Compared with higher or lower OLR_S, the reactors with moderate OLR_S had a more stable methane production and organic matter reduction, and the highest removal of TS (57.43%), VS (66.22%), TCOD (76.18%) and SCOD (89.99%) was obtained under the OLR of 1.89 gVS/(L·d) (5%TS).

All the reactors obtained a BOD_5 removal with a range of 74%-86% (Table 2). These results indicate all of the reactors had a high conversion capacity of organic waste and operating efficiency.

3.1.3 pH and ammonia

The initial pH value of the six reactors was 7.21 and the good digestion environment gives a buffering capacity for the feeding substrate. Thus, a stable pH for R1-R6 was

7.19±0.14, 7.27±0.05, 7.32±0.06, 7.39±0.05, 7.42±0.05 and 7.45±0.08, respectively. Liu et al. (2008) showed that the most favorable range of pH to attain maximal biogas yield is 6.5-7.5. As the OLR increased, the pH values always showed an upward trend, particularly for R5 and R6. This was because of the phenomena that the higher OLR resulted in the higher TAN and FAN concentration (Fig.3). Therefore, careful consideration was required when increasing the OLR for the purpose of enhancing the methane yield due to the simultaneous increase of the risk of ammonia inhibition.

PM is protein-rich material, thus, TAN and FAN coming from proteins breakdown are the foremost inhibitors to the AD process if they are available at high concentrations (Yenigün and Demirel, 2013). Ammonia inhibition occurred in the range of 1500-3000 mg/L TAN with pH above 7.4, and if the TAN concentration was over 3 g/L, ammonia was claimed to be toxic irrespective of pH (Abouelenien et al., 2010; Calli et al., 2005). In this study, the TAN concentration of R1-R3 was always below 1500mg/L (Fig.3A). However, with the increase of OLRs, the TAN concentration increased to nearly 2500mg/L in R6 with an average pH value of 7.45(Fig.3A and Table 2), which was within the inhibitory range. In addition, FAN concentrations ranging from 80 to 150 mg/L have shown an inhibition of the AD process (Yenigün and Demirel, 2013). The increased OLR resulted in the rising FAN concentration, especially the FAN concentration more than 90mg/L at OLR of 3.03gVS/(L·d) (8%TS)(Fig. 3B). Thus, the lower methane yields obtained in higher OLRs may have been a consequence of the high TAN and FAN concentration. It was concluded that the reactors were successfully

operated without inhibition and showed stable and high-efficiency operation under the feeding TS concentration of 5%.

3.2 Simulation study of how OLR affect system performance in large-scale

3.2.1 Mass flow and construction scale

The feeding amount and construction scale of biogas plant could be significantly affected by the varying OLRs (Fig 4). Specifically, 8.0 t/d of fresh PM and a certain amount of additional water were preheated and mixed in the pre-treatment tank. With the increasing OLR, water consumption amount reduced by 28.58%, 45.73%, 57.16%, 65.33% and 71.46% from 3% TS to 4-8%TS, respectively. After fermentation and solid-liquid separation, about 97.37%, 95.40%, 93.50%, 92.00%, 89.87% and 88.01% of the total mass made their way to the liquid digestate of 3%TS to 8%TS, respectively, 1.66-3.84% became biogas and 0.97-8.15% turned into solid digestate. In addition, about 53-57% of total TS (1.02-1.10t/d) transferred into biogas, 11-36% and 11-33% turned into solid and liquid digestate, respectively.

With respect to the construction scale, the pre-treatment tank was designed for storing at least two-day feeding. The effective volume of anaerobic digester was determined as sufficient with a 15% safe vacant according to the HRT, feeding loads and physical characteristics of PM. Considering the digestate amount and seasonal agricultural application, digestate storage tank should have enough space for storing digestate over three months. Compared with OLR of 3%TS, the volume of pre-treatment tank, CSTR reactor and digestate storage tank decreased by nearly 28%, 42%, 52%, 59% and 64%,

while OLR increased to 4%TS- 8%TS, respectively. The lower OLR, the larger amount of water consumption and liquid diegstate, as well as larger reactor volume was observed, which implied the higher initial investment and larger covering space. Conversely, only from the perspective of construction investment, higher OLR poses huge advantages. However, considering the long-term running, the OLR affected the feeding composition and degradation efficiency (Igoni et al., 2008), the higher OLR (like 8%) had great risk of unstable process and ammonia inhibition found in our study (Fig.2) and other previous studies (Guo et al., 2013a; Li et al., 2015; Li et al., 2018). Some attempts have been made to solve the problem, like co-digestion, using additives (Tada et al., 2005) or separating the pig slurry into different concentration fractions (Deng et al., 2014). Nevertheless, the extra operating cost during this process needed to be considered for a long-term run.

3.2.2 Effects of OLR on the heating requirement and supply scenarios

Based on the volume of the anaerobic reactor, the height and diameter of the reactor were assumed according to its volume (Table 3). The OLR was considered to be a key parameter that influenced the self-heating requirement. Specifically, the total energy consumption of 3%TS was 1.33, 1.66, 2.34, 2.81 and 3.31 times of that with OLR of 4%TS to 8%TS (Table 3). And the biogas needed to afford the heating energy decreased from 712.27 m^3 to 215.18 m^3 while the OLR increasing from 3%TS to 8%TS (no heat loss is considered). When the OLR was lower than 5%TS, over 50% of the produced biogas will be used for self-heating. However, irrespective of different OLRs, the

energy consumption for heating the feeding and maintaining digestion temperature accounted for a large proportion of over 97%, and heat loss only took up less than 3%. In addition, OLR also could influence the proportion of different heating requirements. While the OLR lower than 5%TS, the Q1 value was higher than 60%, otherwise, the value was lower than 50% at high OLR. For the low OLR, the large reactor volume and feeding amount resulted in a large amount of self-heating energy and operation cost. In fact, it is difficult to increase the digestion temperature for the reactor running at low OLR (Deng et al., 2017). Thus, it is adaptable to run in tropical regions with the main target of solving the problems of PM disposal and utilizing biogas digestate as fertilizer, instead of producing biogas. Conversely, for the high OLR, the balance between the investment cost and the stable operation cost for a long-term run should be estimated. Thereby, considering the running process and operational cost, the OLR of about 5%TS was more advanced in the following aspects: high methane production and limited digestate production, stable process, moderate construction scale and cost, low operation cost and moderate self-heating energy compulsion.

According to the daily biogas production and the daily energy demand, three scenarios (solar energy, biogas boiler and CHP) of reactor-heating technology was analyzed under 3%, 5% and 8%TS. (Fig.5)

Scenario 1: solar energy. The maximum and minimum monthly average solar radiation of Beijing district was occurred in May and December respectively, with its average solar radiation of 21.35MJ/(m^{2}·d) and 7.45 MJ/(m^{2}·d) (Zhou et al., 2005). As we

can see from Fig.5A, because of the low solar radiation in winter of Beijing, an enormous solar thermal collector (nearly 2000m^2 with the heat loss of 10%) was needed at low OLR of 3%TS. When the OLR increased to 5%TS and 8%TS, the area of the solar collector decreased by nearly 40% and 70%, respectively. Although, there was no need to consumer biogas while solar energy was used, the solar heating method was not suitable for districts with low solar radiation in winter, such as Beijing.

Scenario 2: biogas boiler. Owing to its concept of no pollution and high efficiency, biogas boiler had been widely applied in China. When biogas boiler was applied, there was no need to input additional energy in the coldest month, however, 95.92%, 52.69% and 34.05% of the produced biogas were consumed for self-heating under OLR of 3%TS, 5%TS and 8%TS, respectively (Fig.5B). In practice, the heat load can be used for selection of the boiler equipment. One set of 0.2-ton(8%TS), 0.3-ton (5%TS)and 0.5-ton(3%TS) boiler with heat efficiency of 80% can be used in this case and the working time is 16 hours per day.

Scenario 3: CHP. The CHP, as a promising and cost-effective technology, had been widely developed in European countries. However, it had not been well promoted in China, because of the small capacity and scattered distribution of electricity generation, biogas plants were difficult to create a market-oriented product. However, with the increasing number of large and super-large biogas plant, the installed electricity capacity increased. By 2014, only for the biogas plant treating agricultural waste, the installed electricity capacity is of 177.80MW and annual electricity production is of 426.8

million kWh (Deng et al., 2017). Thus, the situation will be gradually improved.

As can be seen from Fig.5C, for a domestic generator, the amount of power generation and surplus heat recovery by $1m^3$ biogas was about 1.8 kWh and 2.4kWh, respectively. The annual electricity production was 6.00×10^5 (3%TS), 6.71×10^5 (5%TS) and 5.19×10^5 kWh(8%TS), resulting in a profit of \$$5.19\times10^4$-$6.71\times10^4$(US dollars) at an electricity price of \$ 0.10 kWh (Zhang et al., 2013). There is also 8.00×10^5(3%TS), 8.94×10^5(5%TS) and 6.92×10^5 kWh (8%TS) of heat produced annually. The installed power generating capacity of the biogas internal-combustion generator was about 120 kWh at OLR of 3%-5%TS and 100 kW·h at OLR of 8%TS with a working time of 16 hours per day. The surplus heat that was used for heating the digester with its utilization rate of 70%, so it would not meet the energy demand in winter and supplemental heating energy of 10860.48, 3723.02 and 171.13MJ should be used under OLR of 3%TS, 5%TS and 8%TS, respectively.

Sustainability is a vital factor while selecting the heat supply scenarios, which mainly include economic sustainability and environmental sustainability (Lynch, 2017). In terms of the economic cost, the investment of the biogas boiler, solar collector and electricity generator under 5%TS was about \$$0.63\times10^4$, 1.88×10^5 and 3.14×10^5, respectively. Environmental sustainability has been widely investigated using different assessment methods, such as life cycle assessment, energy, exergy (Rosen, 2018). Specifically in energy system, energy production and utilization should avoid energy-environment conflict (Ali et al., 2018). Considering the environmental

sustainability, the three scenarios are all have less environmental impact in comparison with heating using fossil energy. CHP has great conversion efficiency, thus, can achieve maximum greenhouse gases emission saving. However, the environmental costs caused by electricity generation using fossil fuel were often ignored, and the environmental benefits of electricity generation using biogas were also covered by its economic profit. Therefore the environmental value of electricity generation using biogas is not fully reflected and the technology competitiveness is not fully realized in China. Thereby, considering the energy efficiency and cost, biogas boiler was the best option for the biogas plant with the given breeding scale under moderate OLR. And the rest of the biogas can be used as domestic fuel for pig farms and neighboring households. However, for the CHP, because of the high efficiency and high profit from electricity, so in the long run, it may be a good choice for the biogas plant under high OLR. The solar collector can be used in tropical areas to enhance the heat availability and reduce the energy requirements, simultaneously reduce the collector areas and cost.

4. Conclusions

In the tested range of lab-scale experiment, the methane yield did not show a linear increase with the increasing of OLR_S. The OLR of 1.89 gVS/L·d (5%TS) was optimum for PM anaerobic digestion with HRT of 22 days. In this condition, a rapid start-up, high methane yield and organic matter removal, and stable operation process can be obtained. The OLR also have a significant influence on the mass flow, biogas plant scale, energy requirement and supply for self-heating in the simulated large-scale operation.

Regardless of a lab experiment or biogas plant design, OLR was the important parameter and should be seriously considered.

Conflicts of interest

The authors declare no conflict of interest.

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Figure
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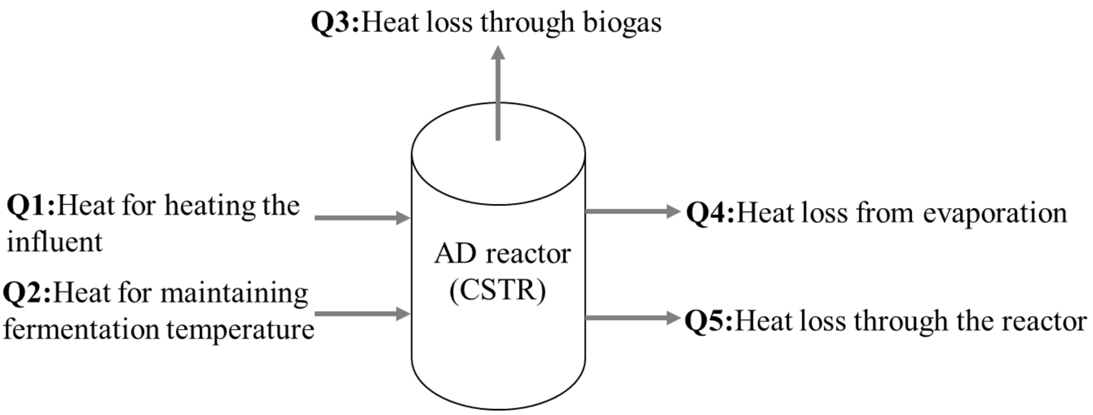

Fig.1 The heat demand of the farm-scale biogas plant

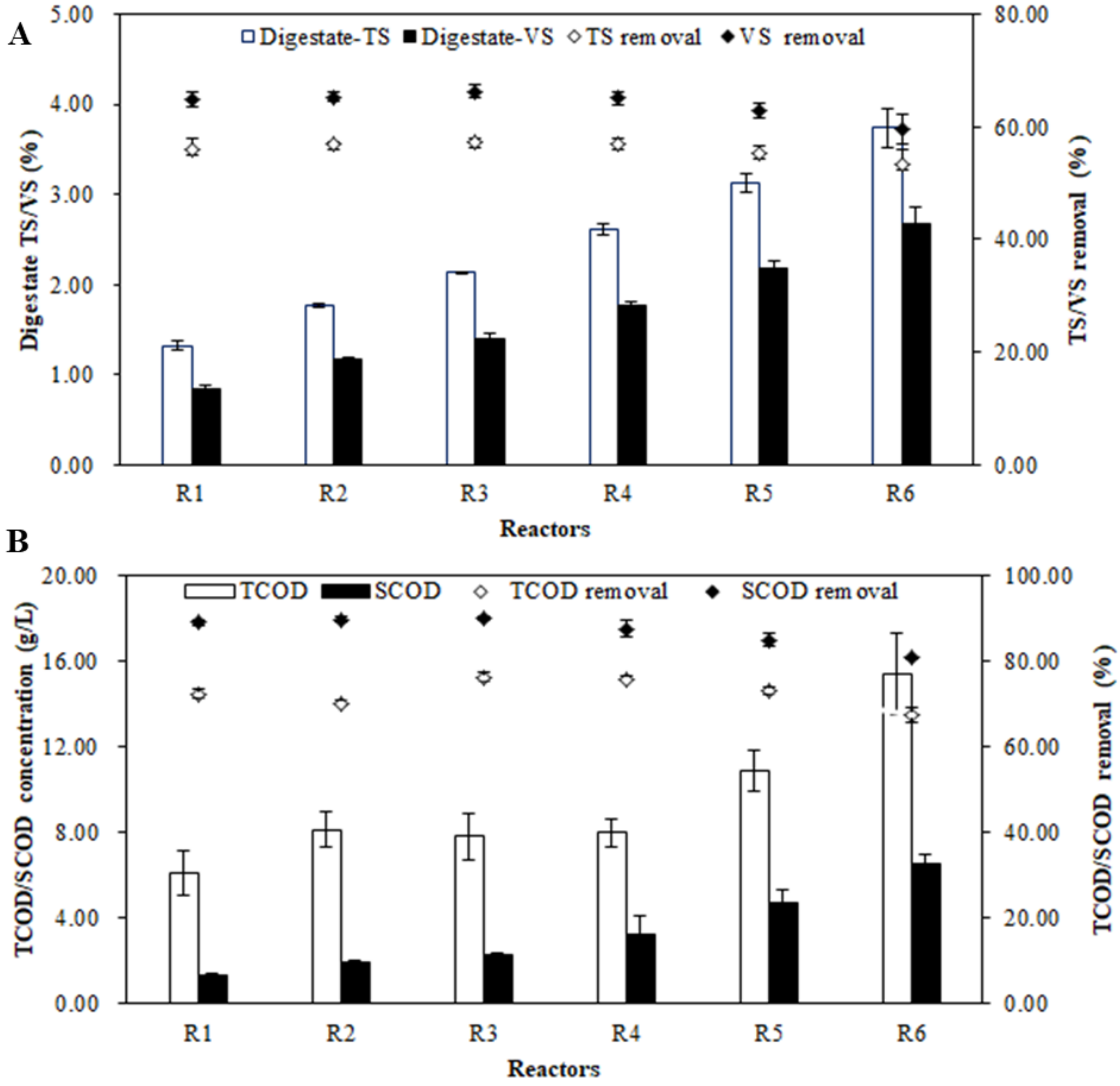

Fig.2 Degradation states under different OLRs (A: TS and VS concentration in digestate and removal, B: TCOD and SCOD concentration in digestate and removal)

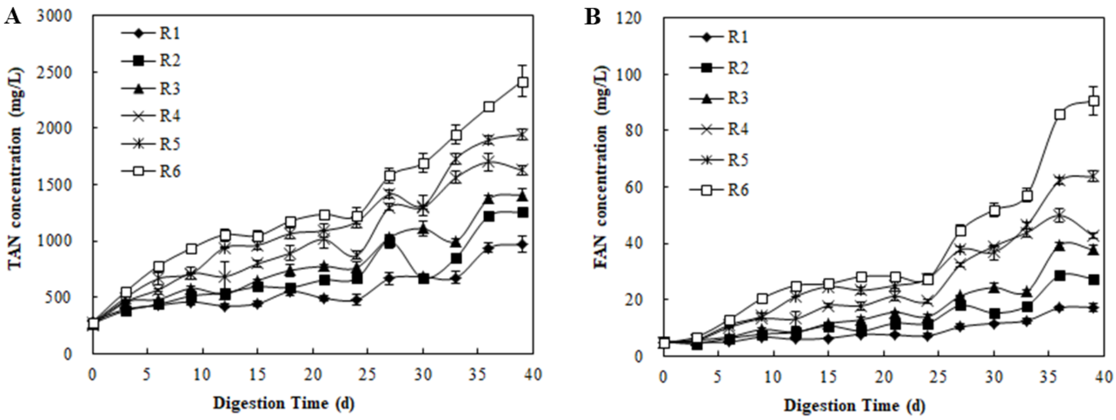

Fig.3 TAN(A) and FAN(B) concentration of digestate with different OLRs

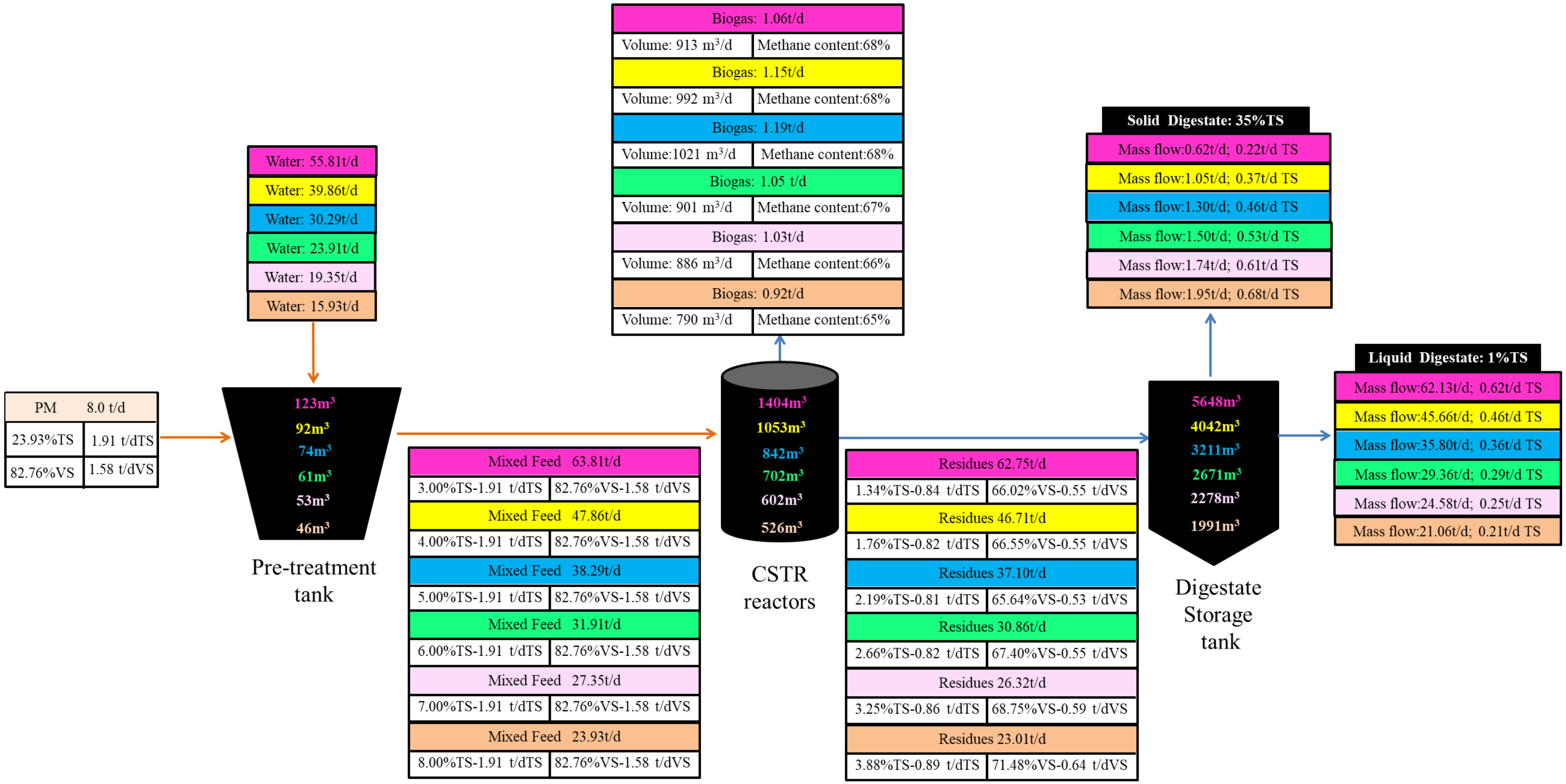

Fig.4 Mass flow and total solid matter flow in the farm-scale biogas plant

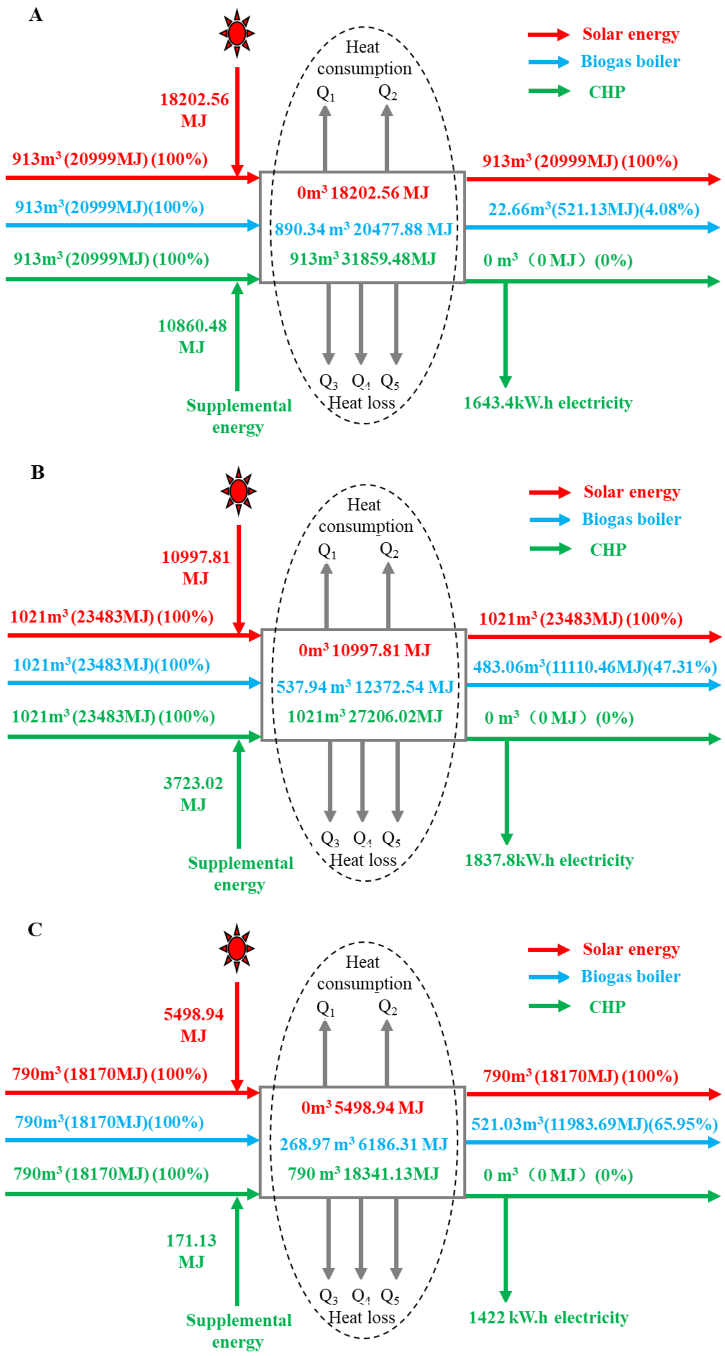

Fig.5 Energy stream diagram with different reactor-heating technology (A-3%TS; B-5%TS; C-8%TS)

Table
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Table 1

Properties of PM and inoculum in the study.

Parameter	PM	Inoculum
TS(%)	23.93±1.07	0.85±0.05
VS (of TS%)	82.76±1.14	70.59±4.10
Crude protein (dry basis %)	11.38±0.63	-
Crude fibers (dry basis%)	9.25±0.33	-
Crude lipids (dry basis%)	11.66±0.72	-
Carbohydrates(dry basis%)	59.72	-
TKN (g/kg)	4.36±1.85	-
TAN (mg/L)	-	272.73±12.30
C/N	10.64	-
pH	-	7.21±0.02

Table 2

Operation parameters for reactors (R1-R6) in lab condition

Parameter	R1	R2	R3	R4	R5	R6
TS (%)	3	4	5	6	7	8
OLR gVS/L·d)	1.13	1.51	1.89	2.27	2.65	3.03
Feeding COD_t(g/L)	21.94±1.82	23.74±0.88	32.85±2.65	33.14±0.67	40.65±1.82	47.3±2.02
Feeding BOD_5(g/L)	14.4	20.2	20.2	27.6	37.0	37.0
CH_4 (%)	63.45-73.44	63.43-72.42	64.13-72.24	61.63-71.39	61.74-70.64	58.75-70.34
Specific methane yield (mL/gVS)	392.06±6.41	425.71±7.49	438.38±12.81	381.01±17.45	368.94±7.20	324.25±4.60
Digestate BOD_5(g/L)	2.40	2.80	3.20	5.20	6.40	9.60
Digestate pH	7.19±0.14	7.27±0.05	7.32±0.06	7.39±0.05	7.42±0.05	7.45±0.08

Table 3

Heat requirement of the farm-scale biogas plant with different OLRs

OLR	Q1 (MJ/d)	Q2 (MJ/d)	Q3 (MJ/d)	Q4 (MJ/d)	Q5 (MJ/d)	Total heat demand ∑Q(MJ/d)	Equivalent Biogas (m^3)	Biogas proportion (%)
3%TS	10111.33	6088.87	46.25	32.97	102.88	16382.30	712.27	78.01
4%TS	7583.90	4566.65	50.22	35.80	96.25	12332.82	536.21	54.05
5%TS	6067.43	3651.59	51.72	36.87	90.43	9898.03	430.35	42.15
6%TS	3788.78	3044.43	46.36	33.01	85.28	6997.87	304.26	33.77
7%TS	3066.20	2610.75	44.90	31.97	78.37	5832.19	253.57	28.62
8%TS	2524.27	2281.16	40.09	28.53	75.01	4949.05	215.18	27.24

Note: the height and diameter of reactor under each TS concentration listed as follows: 12m and 13.37m (3%TS); 9m and 13.37m (4%TS); 7.2m and 13.37m (5%TS); 6m and 13.37m (6%TS); 5.4m and 12.99m (7%TS); 4.8m and 12.99m (8%TS)