Anaerobic Co-Digestion of Food Waste and Thermal Pretreated Waste Activated Sludge: Synergetic Effect and Energy Assessment

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Abstract
Thermal pretreatment was an effective method to improve the anaerobic digestion of waste activated sludge. However, its application in China was still hindered by the high energy demand. In order to balance the energy consumption of sludge thermal pretreatment integrated with anaerobic digestion, food waste was introduced as co-substrate to achieve an energy self-sustainable sludge treatment system. Anaerobic biodegradability test was performed using thermal pretreated sludge and food waste in order to clarify the kinetics and mechanism of co-digestion, especially the synergetic effect on specific methane yield. The prominent synergetic effect was an initial acceleration of cumulative methane production by 20.7-23.8% observed during the first 15 days, and the cumulative methane production of feedstock can be calculated proportionately from its composition. Between the evaluated models, modified Gompertz model presented a better agreement of the experimental results and it was able to describe the synergetic effect, assessed by the relative deviation between theoretical estimation and the experimental results of co-digestion tests. This feature made modified Gompertz model a suitable tool for methane production prediction of mono- and co-digestion. Energy assessment shown that co-digestion with food waste was a sustainable solution to maintain the integration of thermal pretreatment and anaerobic digestion energy neutral or even positive. Besides, the performance of sludge dewatering was a crucial factor for the energy balance.

Highlights
- Co-digestion led to rapid methane production at the beginning
- Relative deviation was used to quantify the synergistic impact of co-substrate
- Model validity was evaluated by its ability to describe synergetic effect
- Modified Gompertz model was superior to First-order model in production forecast
- Energy self-sustain was feasible by using food waste in co-digestion

Introduction
Nowadays, waste activated sludge is globally recognized as a potential resource for renewable energy and nutrient recovery rather than an environmental liability (Cieślak et al. 2015). Anaerobic digestion has proven to be a reliable and economical technology of waste activated sludge management, and is considered as an essential part of modern municipal wastewater treatment plants (WWTPs), to reduce the amount of waste activated sludge and to recovery bioenergy (Appels et al. 2008, Kelessidis & Stasinakis 2012).

Hydrolysis of organic particulate known as the rate-limiting step in sludge anaerobic digestion, is limited by cell walls rupture and extracellular polymeric substances (EPS) degradation (Eastman & Ferguson 1981). To sidestep the bottleneck, various disintegration methods have been introduced prior to anaerobic digestion to release the readily biodegradable substrate contained in the cell (Camacho et al. 2002, Carrère et al. 2010). Thermal pretreatment (TP) is a temperature-dependent reaction, and the maximum methane production is reached at the reaction temperature ranging from 150°C to 175°C (Bougrier et al. 2008, Haug et al. 1978, Li & Noike 1992). Several commercial technologies, such as Cambi, Exelys (Veolia) and CTH (Aqualogy), state that thermal pretreatment can integrate in WWTP with complete energy self-sufficiency (Cano et al. 2015).

With the rapid industrialization and urbanization at least 6.25 million tons of dry sludge is generated per year from over 3600 WWTPs in China, with a processing capacity of 49.43 billion m³/d, and the sludge production is expected to increase in the near future (MEP 2015, Yang et al. 2015). But the organic particulate in the sewage is removed by septic tank, and is further diluted by storm water or underground water in the pipeline. As a result, waste activated sludge in China contains only 20%-50% of organic content, much lower than that in developed countries, (70%-80%) (Yang et al.
The shortage of organics leads directly to a low energy content in the sludge, which has a critical role in the energy generation as biogas produced in anaerobic digestion. In the view of the energy balance, thermal pretreatment and anaerobic digestion of waste activated sludge will stay at its infancy in China, until higher concentration of organic matter is available in the influent of WWTPs.

To solve this problem, anaerobic co-digestion (AcoD) of the high organic feedstock such as food waste (with a methane potential of 302-716 ml CH$_4$/g VS$_{in}$) is a feasible option, to improve digestion efficiency, stability and energy benefit (Koch et al. 2015, Kondusamy & Kalamdhad 2014). Astals reported that the synergistic effect in co-digestion led to an acceleration of specific methane yield, rather than a significant change in cumulative methane production. The results showed that the benefit of anaerobic co-digestion was not only enhancing methane production, but also an acceleration of the degradation kinetics (Astals et al. 2014). The renewable energy systems (RES) in Europe have set forward a fixed goal of supplying 20% of the European energy demands from RES by 2020, and at least 25% of the bioenergy in the future can originate from biogas, produced from wet organic materials such as: animal manure, whole crop silages, wet food and feed wastes, etc. (Holm-Nielsen et al. 2009). America has also harnessed biogas from organic waste management since at least the 1920's, and feeding fats, oil and grease (FOG) to digesters presented better economic benefits (EPA 2014).

In practice, anaerobic co-digestion with food waste (FW) is a cost-effective alternative to improve the energy balance of sludge treatment system. However, few studies have evaluated the synergetic effect and the energy balance of anaerobic co-digestion integrated with sludge thermal pretreatment. Liu et al. applied thermal pretreatment to accelerate food waste solubilization, before co-digestion with waste activated sludge (Liu et al. 2015). However, Cano et al. suggested that thermal pretreatment had not showed remarkable effect on easily degradable substances rich in lipid and carbohydrate (Cano et al. 2014). Besides, Cuetos et al. reported the instability and inhibition in an mesophilic anaerobic co-digestion of thermal pretreated slaughterhouse waste (at 133°C for 20min), caused by LCFA accumulation (Cuetos et al. 2010). Thus, thermal pretreatment of the easily biodegradable substrates prior to anaerobic co-digestion seems contradictory to its original intention.

Up to now, the effects of thermal pretreatment of waste activated sludge on methane production and kinetics parameters were widely studied (Haug et al. 1978, Kepp et al. 2000, Lang et al. 2015, Mottet et al. 2009, Prorot et al. 2011), and, the synergetic effects and mechanism of co-digestion were generally discussed (Angelidaki & Ahring 1997, Elbeshbishy et al. 2012). Nonetheless, the synergetic effects of co-digestion of thermal pretreated sludge and food waste need to be evaluated, and an available tool is require to assess the energy feasibility of full-scale applications and set the basis of process control.

Therefore, the primary objective of this study was to provide better insights into the synergetic effects of thermal pretreated sludge and food waste in anaerobic digestion. Modified Gompertz model and First-Order Model were used to analyze the experimental data and to predict the performance of co-digestion. From them, an energy assessment was performed by analysis energy consumption of the five solid contents of waste activated sludge in thermal pretreatment to evaluate the feasibility to maintain an energy self-sustained anaerobic co-digestion system integrated with sludge thermal pretreatment process.

**Materials And Methods**

**Substrates and inoculum**

Press cake of wasted activated sludge from the municipal WWTP (Nanning, China), with a capacity of 4.8×10$^5$ m$^3$/d using reverse anoxic-anerobic-aerobic process, was diluted to 10% of total solid with distill water before homogenizing
(WBL25B26, Midea Co.,Ltd, China), and then the sludge slurry was stored at 4°C.

Food waste from Guangxi University canteen was used in this study. Non-biodegradable material such as bones, plastic and glass was removed manually before homogenization. Food waste slurry was packed in a plastic container and stored at 4°C. Food waste slurry was thawed to ambient temperature before feeding.

Inoculum from a pilot-scale anaerobic digester fed with pretreated sludge was sieved through a 1mm mesh to remove large particles Inoculum used in this test was degassed by incubation at 37°C until no significant methane production was observed (Elbeshbishy et al. 2012, Lang et al. 2015).

**Thermal Pretreatment**

Thermal pretreatment was conducted in an electric-heating reactor (HK-ZZ01, Hengke Instruments, China), equipped with four 1 liter hydrothermal synthesis reactors inside the chamber. Homogenized sludge slurry was treated at 165°C for 15min. Pretreated sludge was stored at 4°C for further analysis and tests.

**Anaerobic Biodegradability Test**

Anaerobic biodegradability tests were conducted to evaluate the biodegradability of the mono-substrates (WAS, TPS and FW) and three mixtures (TPS/FW= 64%/36%, 37%/63% and 17%/87%, named as AcoD-I, -II and -III) in triplicates.

All the experiments were carried out with the feeding/inoculum ratio (0.5g VS_{Feed} / g VS_{Ino}) suggested by I.angelidaki (I.angelidaki et al. 2009). The head space of the 500ml serum bottle, used as bench-scale digester, was flushed with nitrogen for 3min before sealing with rubber stoppers. The digesters were immersed in a water batch at 37±0.5°C, and manually mixed on daily basis. Daily methane yield was measured by liquid displacement method, with 2% NaOH solution (Esposito et al. 2012). All tests were performed in triplicate and methane production was normalized to the volatile solid in the substrate.

Specific methane yield and cumulative methane production were calculated in VS-base. Despite the so-called 1% criterion was adopted, the ultimate methane production obtained in this study was not equivalent to the biochemical methane potential of the substrate (VDI 2006).

**Analytical Methods**

In order to know the dilution factor of the sludge press cake and food waste, total solid (TS) and volatile solid (VS) of all the samples were determined by heating at 105°C during 24 h for total solids and 550°C during 2 h for volatile solid. Soluble fraction of the sludge were defined by centrifuging at 8000 rpm for 20 min, and the supernatant were characterized by soluble chemical oxygen demand (SCOD), ammonia, alkalinity and volatile fatty acids (VFA). VFA was analyzed according five-point titration method (Lahav &Morgan 2004). SCOD, TS, VS, pH and alkalinity were determined according to standard methods (APHA 1998). All analyses were performed in triplicate and given as mean ± standard deviation.
Table 1
Characteristics of raw sludge, thermal pretreated sludge (TPS) and food waste (FW)

|                  | Raw sludge | TPS     | FW         |
|------------------|------------|---------|------------|
| TS (%, ww)       | 21.6 ± 0.2 | 10.0 ± 0.4 | 13.6 ± 0.2 |
| VS/TS (%)        | 54.7 ± 0.3 | 52.4 ± 0.3 | 91.9 ± 0.2 |
| SCOD (mg/L)      | 2870 ± 150 | 18830 ± 530 | 10750 ± 330 |
| pH               | 8.2 ± 0.1  | 5.8 ± 0.1  | 4.8 ± 0.1  |
| VFA (mg/L)       | 330 ± 14   | 2470 ± 210 | 1850 ± 170 |
| Alkalinity (mg/L)| 645 ± 30   | 1250 ± 135 | 580 ± 23   |

Calculation

**Modeling** The objective of introducing the simplified models was to obtain kinetics parameters allowing us to compare the results and predict the methane production. Modified Gompertz model and First-order kinetics model (FO) were applied to estimate the kinetics parameter in this work, allowing reliable comparison between mono- and co-digestion. Nonlinear optimization by the least squares procedure was applied in the simulation, using Matlab® (R2014a).

Modified Gompertz model, next presented in Eq. (1), was used to describe the progression of cumulative methane production in the batch tests (Lay et al. 1997):

\[
M = M_0 \cdot \exp \left\{ - \exp \left[ \frac{R_{\text{max}} \cdot e^{-t}}{M_0} (\lambda - t) \right] + 1 \right\} \quad \text{Eq. 1}
\]

Where \( M \) is the cumulative methane production (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \)), \( M_0 \) is ultimate methane production (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \)), \( R_{\text{max}} \) is the ultimate specific methane production rate (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \cdot \text{d}\)), \( \lambda \) is lag phase time (day), \( t \) is the digestion time (day), and \( e = 2.718 \).

First-order kinetics is used to describe the hydrolysis of particulate organic matter. The progression of cumulative methane production can be described by the following equation (Veeken & Hamelers 1999):

\[
M = M_0 \times \left[ 1 - \exp \left( - k_h \cdot t \right) \right] \quad \text{Eq. 2}
\]

Where \( M \) is the cumulative methane potential (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \)) at digestion time \( t \) days, \( M_0 \) is ultimate methane potential of the substrate (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \cdot \text{d}\)), \( k_h \) is first-order hydrolysis rate constant (\( \text{d}^{-1} \)), \( t \) is the digestion time (days).

**Theoretical methane production** The term “methane production” can be referred to cumulative methane production (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \)) or specific methane yield (\( \text{mlCH}_4 / \text{gVS}_{\text{in}} \cdot \text{d}\)) in this study. If the type and the composition of the substrate are known and all the materials are converted to biogas, the theoretical methane production of the substrate can be calculated from the following equation (Angelidaki & Sanders 2004, Astals et al. 2014):

\[
M_T(t) = p \times M_{\text{TPS}} + (1 - p) \times M_{\text{FW}} \quad \text{Eq. 3}
\]
Where $M_T(t)$ is the theoretical methane production of the mixture fed in AcoD, and $M_{TPS}$ and $M_{FW}$ are the methane production of thermal pretreated sludge and food waste in mono-substrate digestion; $p$ (%) is organic fraction of pretreated sludge in the mixture.

**Relative deviation** The synergistic effect during co-digestion was expressed as the relative deviation of theoretical production, to quantify the difference between mono-substrate digestion and co-digestion, from the following equation:

$$RD = \left( \frac{M - M_T}{M_T} \right) \times 100 \% \text{ Eq. 4}$$

Where $RD$ (%) is the relative deviation of the experimental data and the theoretical estimation of cumulative methane production, $M (\text{mlCH}_4/\text{gVS}_\text{in})$ is the cumulative methane production of co-digestion, or its fitted value of the kinetics model; and $M_T(\text{mlCH}_4/\text{gVS}_\text{in})$ is its theoretical cumulative methane production calculated by Eq. 3

**Energy Assessment**

Methane production of a full-scale anaerobic digester was referred to the results of the biodegradability test. The configuration of a typical thermal pretreatment was conducted at 170°C for 30 min, fed with dewatered sludge (TS, 16%) (Kepp et al. 2000). Methane production, expressed per ton of volatile solid fed, was used in the energy assessment. For the simplest scenario of thermal pretreatment that the steam required in heating is generated in a boiler fed with biogas. Energy requirement for each ton of sludge, by wet weight, was 18.7 Nm³ natural gas for the steam boiler, equivalent to 14.6 Nm³ methane (Cano et al. 2014). A 10% loss of thermal efficiency caused by directly biogas burning without upgrading, therefore the methane consumption in pretreatment is 16.1 Nm³ methane per ton of substrate in this study.

**Results**

**Anaerobic biodegradability test**

**Cumulative methane production**

Cumulative methane production of waste activated sludge increased by 57% after pretreatment, from 197 mlCH$_4$/gVS$_\text{in}$ to 310 mlCH$_4$/gVS$_\text{in}$. It was evidenced that thermal pretreatment improved the biodegradability of waste activated sludge and unlocked its potential unable to release in anaerobic digestion without pretreatment. Cano reported similar result that methane production increased from 184 mlCH$_4$/gVS$_\text{in}$ for raw sludge to 278 mlCH$_4$/gVS$_\text{in}$ for the treated sludge (Cano et al. 2014). The maximum cumulative methane production (819 mlCH$_4$/gVS$_\text{in}$) was obtained from food waste, which was 1.64 and 3.16 times of that of raw sludge and treated sludge. Considering the organic content (92.0% for food waste and only 54.7% for RS), the actual multiples of cumulative methane production of food waste calculated on TS basis, would stretch to 7.49 and 4.95 times for the raw sludge and the treated sludge, in accordance with Mashad (El-Mashad & Zhang 2010) and Zhang (Zhang et al. 2013). This result suggested that food waste is a reasonable co-substrate to improve the energy balance of anaerobic digestion process integrated with thermal pretreatment and/or combined heat and power (CHP) system.

As shown in Fig. 2, co-digestion exerted a greater effect on the maximum specific methane yield than the cumulative methane production. Theoretical cumulative methane production of co-digestion calculated from Eq. 3 agreed with the experimental results (485, 621 and 731 mlCH$_4$/gVS$_\text{in}$ for AcoD-I, -II and -III) at the end of the experiment. Different methane production profiles were observed, however the deviation of the theoretical calculations in all tests fell below 10% after 15 day, the suggested solid retention time (SRT) for completely mixed mesophilic digesters (Bolzonella et al. 2014).
In the end, the relative deviation was down to 1.5%, indicating that the ultimate cumulative methane production of the substrates was conserved and determined by the substrate composition. Astals also reported that the synergistic effect in co-digestion led to an acceleration of specific methane yield, rather than a significant change in cumulative methane production (Astals et al. 2014).

**Specific Methane Yield**

Specific methane yield of mono-substrates (FW, TPS and raw sludge) demonstrated the different degradation kinetics in anaerobic digestion (Fig. 1). Pretreated sludge started with a rapid spike of 46 mlCH$_4$/gVS$_{in}$ • d at day 4, following by a moderate decrease since day 8, whereas that of raw sludge remained fairly constant below 15mlCH$_4$/gVS$_{in}$ • d through the experiment. The rapid methane production observed in the early phase was attributed to the liberation of the intra-cellular content from waste activated sludge, thus providing more accessible soluble and micro-particle organics for the anaerobic microorganism. This assumption was supported by the remarkable enhancement of SCOD and VFA due to the cell breakage and intercellular substrate leakage, in accordance with Mottet et al (Mottet et al. 2009). Beside the solubilization, the deflocculation of macro-flocs structure in pretreatment provided extra surface area for microorganism (Prorot et al. 2011).

For food waste, the maximum and the average specific methane yield were 108mlCH$_4$/gVS$_{in}$ • d and 32 mlCH$_4$/gVS$_{in}$ • d, equivalent to 2.34 and 3.20 times of the figures of TPS. The specific methane yield of food waste fitted a sawtooth profile with three major peaks of 57, 45, 56mlCH$_4$/gVS$_{in}$ • d, at day 2, 7, and 12, suggesting that, as a mixture of multi-substrate, food waste would show a complex degradation behavior, as the result of the combined effect of particle size distribution and chemical composition (Pavlostathis & Giraldo-Gomez 1991, Prorot et al. 2011).

Specific methane yield in co-digestions (solid line, black) along with their theoretical estimations (dot line, red) calculated according to Eq. 3, are given in Fig. 1. By summing the specific methane yield of each substrate, the superposition of the signature curve shape was obtained. As expected, the signature three-peak curve of food waste observed in mono-substrate digestion reappeared in AcoD-II and AcoD-III, which had higher weight of food waste in the mixture.

However, the prominent feature of the synergistic effect of co-digestion was the acceleration of specific methane yield observed in the first 10 day for all co-digestion tests. This difference between experimental data and theoretical prediction might be associated with the inhibitory compounds dilution. For example, LCFAs, the intermediates of lipid degradation, were known as an inhibitor for Gram-positive bacteria even at low concentration. The toxicity of LCFAs was caused by the surface adsorption on cell wall or cell membrane, which resulted in the malfunction of mass transfer and/or cell protection (Chen et al. 2008). In the test, the addition of TPS diluted the LCFAs concentration in the digester, thus reduce the probability of inhibition and improved the digestion efficiency (Astals et al. 2014). Due to the conservation of methane potential in co-digestion, the specific methane yield recorded in the experiment inevitably felt below the its theoretical estimation after the initial quick methane production, indicating the rapid depletion of organic material.

**Modeling**

The results obtained in anaerobic biodegradability tests with the modified Gompertz model and First-order kinetics fine-tuning for food waste, raw sludge, pretreated sludge and the mixture are enclosed in Table.2 and the cumulative methane yield curves are plotted in Fig. 1. Parameters obtained by both models with a degree of accuracy ($R^2$ over 0.974), enable a quantification of these kinetics improvements (Tab.2).
Modified Gompertz model uses three parameters to describe the cumulative methane production: ultimate methane production ($M_0$), ultimate specific methane yield ($R_{\text{max}}$) and lag phase time ($\lambda$). The ultimate methane potential ($M_0$) estimated by modified Gompertz model generally agreed with the experimental results with a negative deviation of 1.5%-6.0%. $R_{\text{max}}$ indicates the initial slope of the curve, described the maximum daily methane production. Waste activated sludge suffered an increase of $R_{\text{max}}$ by 204% after pretreatment, from 9.03 to 27.43 ml CH$_4$/gVS$_{\text{in}}$·d, pointed to the deflocculation and the solubilization of sludge flocs (Donoso-Bravo et al. 2011). The $R_{\text{max}}$ of AcoD-II and AcoD-III were higher than that of food waste which has the highest biochemical methane potential in the test. This result could be explained by two synergetic effects as the result of the TPS addition: (1) supplying extra buffer capacity of VFA formed in acidogenesis and acetogenesis (Angelidaki & Ahring 1997); (2) the dilution of LCFAs generated in lipid degradation, to reduce the risk of inoculum deactivation caused by surface absorption (Palatsi et al. 2009).

The negligible lag time ($\lambda$) of raw sludge and TPS indicated that no significant incubation time was needed for the inoculum to start the digestion process. Food waste, characterized as a readily biodegradable substrate with high methane potential, is worth to concern its potential inhibition caused by the metabolites of protein, grease and lipid (Alves et al. 2001). As shown in Tab.2, a high proportion of food waste in the feed would exert a negative impact on lag time, which could be explained by the depression of inoculum bioactivity caused by LCFA, even in low concentration (Chen et al. 2008).

For First-order Model, the apparent hydrolysis constant ($k_h$) is the reciprocal of time when half of ultimate methane production was achieved. In the case of the thermal pretreatment effect, the initial kinetics acceleration of waste activated sludge ($k_h$ increased from 0.04d$^{-1}$ to 0.14d$^{-1}$) demonstrated that the different degradation kinetics of the particulate organic matter and the soluble organic matter, which coincided with the increment of SCOD and VFA. The hydrolysis constant did not show a clear correlation with the substrate composition like it did with cumulative methane production. However, the comparison between the hydrolysis constant of food waste and AcoD-III (17%TPS-83%FW) highlighted the synergetic effect of AcoD that the degradation rate of food waste was apparently enhanced by the addition of pretreated sludge (Table-2). As previously discussed, the high proportion of food waste fed in the co-digestion would cause temporary inhibitory effects by LCFA absorption and/or VFA accumulation, thus suppressed the bioactivity of inoculum and slowed down the specific methane yield.

According to the square deviation (Table.2), both models seem to give a better estimation of cumulative methane production in most cases, similar results was obtained by Donoso-Bravo (Donoso-Bravo et al. 2010); however, the cumulative methane production of raw sludge and food waste were obviously distorted by First-order kinetics in this study. Referred to the lag phase in modified Gompertz model, the both substrates were slowly biodegradable, which means that they required a longer digestion period to obtain a satisfactory estimation by First-order kinetics. Besides, modified Gompertz model was superior to First-order model in describing the curve shape of cumulative methane production, as it can be seen in Fig. 1 (solid, line). Up to this point, the comparison between the models was still inconclusive, and more quantitative evidences were necessary to perform further assessment.
Table 2
Experimental data and the kinetic parameters obtained with the evaluated models

| Substrate          | Experimental | Modified Gompertz model | First-Order kinetics |
|--------------------|--------------|-------------------------|----------------------|
|                    | M           | R<sub>max</sub>         | M<sub>0</sub> | R<sub>max</sub> | λ | R<sup>2</sup> | M<sub>0</sub> | k<sub>h</sub> | R<sup>2</sup> |
| Raw Sludge         | 197         | 13.6                    | 194              | 9.03          | 0.01 | 0.985     | 259           | 0.04          | 0.990       |
| TPS                | 310         | 45.8                    | 293              | 27.43         | 0.00 | 0.974     | 305           | 0.14          | 0.996       |
| 64%TPS-36%FW       | 485         | 59.9                    | 463              | 51.97         | 0.64 | 0.988     | 479           | 0.16          | 0.995       |
| 37%TPS-64%FW       | 621         | 64.8                    | 601              | 56.94         | 1.13 | 0.995     | 619           | 0.14          | 0.995       |
| 17%TPS-83%FW       | 731         | 69.2                    | 711              | 61.12         | 1.24 | 0.997     | 742           | 0.12          | 0.997       |
| FW                 | 819         | 56.7                    | 846              | 50.69         | 1.86 | 0.999     | 1195          | 0.04          | 0.974       |

Synergetic Effects Assessment

Supposing there was no interaction between thermal pretreated sludge and food waste, the RD value shall be null in co-digestion tests. However, before the materials were fully converted, the deviation on the prediction of cumulative methane production reflects the interaction between pretreated sludge and food waste in co-digestion. Therefore, the relative deviation of the theoretical estimation was used to quantify the synergetic effect between the co-substrates. According to the experimental results, the synergic effects was 10.7%, 16.0% and 31.2% of improvements for AcoD-I, -II, -III at the very beginning (Fig. 3, solid line). During the first 15 days, the AcoD tests obtained a remarkable increment of cumulative methane production from 20.7%-23.8%. However, with the depletion of the substrate the synergetic effect faded away as the co-digestion proceeding, declining from approximately 10% at day 15 to below 1.5% by the end of the experiment. It was interesting to highlight that a small amount of thermal pretreat sludge in AcoD-III would remarkably improve the performance of food waste digestion, that might relate to the dilution of inhabitant previously discussed. In addition, no antagonistic effect was detected in trails of all tested blending ratios.

For a full-scale biogas plant, it is essential to predict the cumulative methane production, based on the amount and the composition of the substrates. Theoretically, this estimation could be calculated from Eq. 3, based on the kinetics model of each substrate. In this case, the accuracy of the estimation is depending on the kinetic models. Owning to the fact that First-order overestimated the cumulative methane production of food waste by 46%, its model-based predictions generated non-negligible relative deviations of 29%, 39% and 43% for AcoD-I, -II, -III. On contrary, the relative deviation of the prediction of modified Gompertz model was below 3%. That was to say that modified Gompertz model was superior to First-order model in predicting the cumulative methane production in co-digestion.

On the other hand, the progress of substrate degradation shall be predicted to reckon the response of the digester to the feeding regime, including the amount and composition of feedstock. Thus, a reasonable model would not merely to give the final result of the process, but also to depict the progress of the process. According to this criterion, a comparison was conducted as shown in Fig. 3 to quantify the ability of the models on the prediction of AcoD progress. The relative deviation of modified Gompertz model (dot line) was in line with the curve of the experimental data (solid line), sketching
out the rapid growth and the gradual decline of the relative deviation. Whereas, First-order model (dash line) can note describe the progress of the synergetic effect in co-digestion. Thus, modified Gompertz model is superior to First-order model to express the interaction of the substrates in co-digestion process.

**Energy Assessment**

Energy requirement for thermal pretreatment expressed per ton of substrate is converted to pre ton of VS in this assessment. Table 3 shows the main results for raw sludge with different solid content. First, it was remarkable that the performance of sludge dewatering greatly played a crucial role in reducing the overall energy cost, and the net benefit of thermal pretreatment in methane production was a constant for a certain substrate. From the findings presented, it was obvious that incorporating thermal pretreatment incurs a loss of net energy output, which was an undesirable result for the waste management enterprise relied on the energy revenues. However, thermal pretreatment could only balance its energy cost with a solid content of 26%, beyond the range in any full-scale system (Abu-Orf & Goss 2012). This conclusion indicated that other feedstock shall be introduced in this system to improve its energy balance. Therefore, food waste was introduced as a co-substrate to enhance the energy production and economic performances of the waste activated sludge management system, and the results were optimistic that a small fraction of food waste in feedstock would neutralize the extra energy demand of thermal pretreatment.

The extrapolation from the batch tests and the theoretical balance describes the prospect of a sustainable organic solid waste management system by upgrading the existing sludge digestion facilities or the new designed plants integrated with thermal pretreatment process.

| Process              | Parameter      | Unit            | Total solid of feeding sludge, % |
|----------------------|----------------|-----------------|----------------------------------|
|                      |                |                 | 10%    | 12%    | 14%    | 16%    | 18%    | 20%    | 26%    |
| Thermal pretreatment | Energy demand  | Nm³ / t VS<sub>Feed</sub> | 294.3  | 245.3  | 210.2  | 184.0  | 163.5  | 147.2  | 113.0  |
| Anaerobic digestion  | Net benefit*   | Nm³ / t VS<sub>Feed</sub> | 113    | 113    | 113    | 113    | 113    | 113    | 113    |
|                      | Energy gap     | Nm³ / t VS<sub>Feed</sub> | -181.3 | -132.3 | -97.2  | -71.0  | -50.5  | -34.2  | 0.0    |
| Anaerobic co-digestion| Food waste     | Nm³ / t VS<sub>Feed</sub> | 819    | 819    | 819    | 819    | 819    | 819    | 819    |
|                      | Blend ratio    | tVS<sub>Food waste</sub>/tVS<sub>Sludge</sub> | 0.22   | 0.16   | 0.12   | 0.09   | 0.06   | 0.04   | -      |

* Growth of methane production after thermal pretreatment, based on the experimental data in Table 2.

**Conclusions**

Thermal pretreatment led to an increase of cumulative methane production by 57%, equivalent to a net energy benefit of 113 Nm³ / t VS<sub>Feed</sub> and kinetics acceleration. The dominant synergetic effect of co-digestion was the acceleration of methane production observed at the early phase, measured with relative deviation by up to 20.7%-23.8%.

Cumulative methane production was satisfactorily predicted by both evaluated models in most cases. Methane production of anaerobic co-digestion was predictable according to the composition of the feedstock, and modified
Gompertz model was able to predict not only the amount but also the progress of cumulative methane production. However, First-order kinetics was unsuitable to the synergetic effect thus failed to predict the performance of anaerobic co-digestion.

The energy balance of the integration of thermal pretreatment and anaerobic digestion was dependent on the methane production of the substrates and the performance of sludge dewatering. Food waste was highlighted in the energy assessment as an appropriate cosubstrate in co-digestion to improve energy benefit, especially to neutralize the energy demand in thermal pretreatment. The conclusions in this study shall be verified in continuous experiment before used as a reference in full scale application, to identify the long term effects of food waste on digestion stability.

Declarations

-Funding Information
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-Ethical Approval
NOTapplicable

-Consent to Participate
NOTapplicable

-Consent to Publish
All the authors confirm that:
-the work described has not been published before,
-it is not under consideration for publication elsewhere,
-its publication has been approved by all co-authors,
-its publication has been approved by the responsible authorities at the institution where the work is carried out.

-Author Contributions
Zhang Jian, Liu Yang and Xie Tian contributed to the conception of the study; Gan Peng, Zhang Jian, and Wang Ru-yi performed the experiment; Liu Yang, Gan Peng and Zong Yi-feng contributed significantly to analysis and manuscript preparation; Zhang Jian, Gan Peng and Liu Yang performed the data analyses and wrote the manuscript; Xie Tian and Wei Yan-ling helped perform the analysis with constructive discussions.

-Competing Interests
NOTapplicable

-Availability of data and materials
This manuscript is based on our original research that has not been published previously.
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**Figures**

**Figure 1**

Cumulative methane production in the anaerobic biodegradability tests using mono- and co-substrate. Experimental data (dot), model fit (solid line) and model-base estimation (dash line)

**Figure 2**

Experimental data (black, solid line) and theoretical estimation (red, dot line) of specific methane yield in anaerobic biodegradability tests using raw sludge (without pretreatment), thermal pretreated sludge (TPS) and food waste (FW) as substrates.

**Figure 3**

Relative deviation of theoretical methane production obtained with experimental data (solid line), Reaction curve model (dot line) and the First-order kinetics (dash line).