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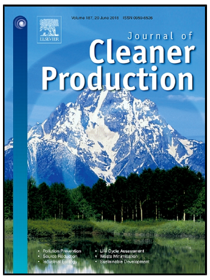

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PII: S0959-6526(18)31286-1

DOI: 10.1016/j.jclepro.2018.04.244

Reference: JCLP 12825

To appear in: *Journal of Cleaner Production*

Received Date: 23 January 2018

Revised Date: 26 April 2018

Accepted Date: 26 April 2018

Please cite this article as: Min Ye, Jianyong Liu, Chaonan Ma, Yu-You Li, Lianpei Zou, Guangren Qian, Zhi Ping Xu, Improving the stability and efficiency of anaerobic digestion of food waste using additives: A critical review, *Journal of Cleaner Production* (2018), doi: 10.1016/j.jclepro.2018.04.244

Word count resultant: 11191 words.

Improving the stability and efficiency of anaerobic digestion of food waste using additives: A critical review

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ABSTRACT

Anaerobic digestion is an effective technology to treat food waste, with methane production as renewable bioenergy. However, there are two key problems in the practical application, i.e., poor system stability and low reactor efficiency. In this paper, additives used in anaerobic digestion of food waste were systematically reviewed in view of system stability and reactor efficiency. Enzymes showed excellent property in food waste pre-hydrolysis stage with almost all macromolecular matters being rapidly resolved. Fungi fermentation process to produce hydrolytic enzymes, can be regarded as a promising and low-cost way to realize rate-limiting step elimination. It can be also concluded that adding neutralizers, buffer chemicals and some other materials is effective to maintain the pH level for practical application. Trace metals in food waste are not enough but needed for methanogens activation in long term and high loading rate operation. In addition, direct interspecies electron transfer could be much helpful for intermediate refractory organic acids degradation and methanogenesis promotion with additives of conductive materials, which is also discussed and should be studied further in anaerobic digestion of food waste. Based on literature review, a new concept is proposed for further study, suggesting that after being well liquefied with enzyme pre-hydrolysis, food waste could be co-digested with landfill leachate in a high-rate anaerobic reactor stably, resulting in a high bioenergy recovery efficiency.

Keywords: food waste; additives; enzymes; trace metals; co-digestion; direct interspecies electron transfer

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Abbreviation

FW	Food waste
AD	Anaerobic digestion
DIET	Direct interspecies electron transfer
TMs	Trace metals
VFAs	Volatile fatty acids
OLRs	Organic loading rates
TS	Total solid
VS	Volatile solid
VSS	Volatile suspended solid
SSF	Solid-state fermentation
FANs	Free amino acids
LCFAs	Long chain fatty acids
UASB	Upflow anaerobic sludge blanket
EGSB	Expanded granular sludge bed
IC	Inner circulation
ZVI	Zero-valent iron
C/N	Carbon/nitrogen
GAC	Granular activated carbon
COD	Chemical oxygen demand
$\Delta G^{\circ\prime}$	Standard free energy change
LL	Landfill leachate

1. Introduction

It is well known that food waste (FW) is a high moisture content and easily biodegradable biomass, and bioconversion process is a mainstream method to minimize waste and realize bioenergy recovery simultaneously (Chen et al., 2017; Karmee, 2016; Kuruti et al., 2017). In 2012, over 1.6 billion tons of FW was generated worldwide (Ma et al., 2017a). The amount of FW is growing by 44% from 2005 to 2025 due to rapid urban development (Capson-Tojo et al., 2016; Karmee, 2016). In China, FW production could be as high as approximately 1.4×10^8 tons per year in 2020, which is equivalent to 10 million tons of coal based on energy conversion by electricity production (Pham et al., 2015; Zhang et al., 2016). Thus, it is urgent to find the right way to manage the increasing FW properly and improve the energy recovery efficiency, from the cleaner production view (Han et al., 2016a).

Anaerobic digestion (AD) is a popular technology applied all over the world to produce bioenergy (Thi et al., 2016; Uçkun Kiran et al., 2014; Yan et al., 2016), but there are two key problems that limit its practical application in FW treatment. One is the poor system stability due to the accumulation of volatile fatty acids (VFAs); the other is the low reactor efficiency, that is, low organic loading rates (OLRs) (Braguglia et al., 2018; De Clercq et al., 2016; Zhang et al., 2014). This is mainly because of the high content of easily biodegradable suspended solids in FW, which is very different from other wastes and wastewaters. Numerous studies were performed on how to improve the stability and efficiency of FW anaerobic digestion, including additives, pre-treatments, co-digestion with other wastes, innovative digesters, and exploration of different operational conditions (e.g. temperature, retention time, and recirculation) (Li et al., 2017; Xiao et al., 2015; Zamanzadeh et al., 2016).

A few recently published review papers on enhancement of methane production from FW focused on pre-treatments, co-digestion, inhibitory factors (e.g. carbon/nitrogen (C/N) ratio, VFAs, ammonia, and environmental conditions), anaerobic reactors, microbial characteristics (Ren et al., 2018; Wang et al., 2018; Zhang et al., 2017). These review papers gave important specific information of research progress on anaerobic digestion of FW. However, this paper focuses on solving the two key problems of FW anaerobic digestion using additives to improve the system stability and reactor efficiency, based on literature review and our previous study. Though the concept of additives to general AD system had been already putted forward by Romero-Güiza et al. (2016), but it is very different in AD system treating FW (high solid content, easily degradable organic, acids accumulation, lack of nutrients, low energy conversion efficiency) from general systems (Li et al., 2018a).

Firstly, this paper is to summarize the use of additives to enhance methane production from FW based on: (i) promoting hydrolysis; (ii) adding neutralizer/bicarbonate/buffer materials to maintain a stable pH; (iii) adding trace metals (TMs) and novel additive materials to support microbial metabolism and promote microbe colonization. Based on the review and discussion, co-digestion of FW (pre-hydrolysed with enzyme) and landfill leachate in high-rate reactor is proposed as a promising way to improve the system stability and reactor efficiency of FW anaerobic digestion.

2. Methods

The literature used in this review, was collected from the online data bases of Science Direct and Web of Science via keyword research. Various keyword groups were comprised of several words including food waste, kitchen waste, anaerobic digestion, fermentation, biogas, and biomethane. Based on the analysis of the obtained papers, it was summarized that the characteristics of FW always cause poor system stability and low reactor efficiency of FW anaerobic digestion. In particular, among literatures, adding some exogenous substances including inorganic, organic, and biological matters has been widely studied to solve the above-mentioned problems. Fig. 1 exhibits the function of some mainly used additives during different AD stages (i.e. hydrolysis, acidogenesis, methanogenesis). The detail information of using additives to improve methane production from FW is to be discussed on the following sections.

3. Additives to promote rate-limiting hydrolysis

Food waste is rich in carbohydrates, proteins, and lipids, with a biochemical methane potential of approximately 460 ml CH_4/g VS (Browne and Murphy, 2013; Capson-Tojo et al., 2016). However, most of the organic contents in FW are suspended solids, which cause inefficient (low) methane production (Zhang et al., 2014). Hydrolysis is considered to be the rate-limiting step. Numerous pre-treatment methods, including thermal/hydrothermal (Ding et al., 2017), ultrasonic (Elbeshbishy and Nakhla, 2011), alkali or acid (Zhao et al., 2011), autoclaving (Tampio et al., 2014), microwaving (Shahriari et al., 2013), freezing/thawing (Stabnikova et al., 2008b), micro aeration (Rafieenia et al., 2017), and high voltage pulse discharge (Zou et al., 2016), were studied to promote FW hydrolysis and thus enhance methane production. Nevertheless, the application of these methods is restricted because of factors including by-products (e.g. furfural) generated during the pre-treatment process and impractical additional costs. Compared with physical or chemical pre-treatment methods, the use of bio-additives is relatively harmless, clean, and efficient. Bio-additives play biological role similar to enhanced fermentative bacteria in hydrolysis. Presently, studies mainly focus on enzymes and fungal mash.

3.1 Enzymes

Enzyme, as a kind of exoenzyme, can help convert macromolecule solids to soluble micro molecule matter (Han et al., 2015). In fact, enzyme additives have been applied successfully in lactic acid and alcohol fermentation using FW (Tashiro et al., 2013; Yan et al., 2011). Protease and amylase additives were also used to enhance the solubilisation of waste activated sludge by 39.7% and 54.2%, respectively (Yang et al., 2010). It can be concluded that enzyme additives are more effective for FW than for waste activated sludge, because it is more difficult to break bacterial cell walls (Parawira, 2012).

3.1.1. Specific role of enzymes for different FW components

Different FW components, such as starches, proteins, and lipids, can be disintegrated into glucose, free amino acids (FANs), and long chain fatty acids (LCFAs) by the corresponding enzymes (Yan et al., 2011).

About 55-65% of the total FW organic solid is starch (Ma et al., 2017a). Starch can be converted to glucose firstly and then to methane and carbon dioxides finally. Hence, starch is the most important component in FW for methane production. In the hydrolysis of starch, α-amylase/glucoamylase addition could help to break glucosidic bonds and thus improve the hydrolysis effect. For instance, Han et al. (2016c) investigated the starch conversion rate was 68.1%-96.2% with glucose production by 0.307-0.434 g glucose/g FW under enzyme pre-hydrolysis.

Macromolecule protein is another noteworthy matter in FW. Protease was proved to be effective to decompose protein structure with peptide links being broken during hydrolysis (Han et al., 2016b). The hydrolysate of proteins, which contain several FANs, could be broken down further into organic acids and ammonia via deaminase secreted by fermentative bacteria (Xiao et al., 2014). Therefore, using protease as an additive improves hydrolysis rate and enhances methane production (Moon and Song, 2011). Furthermore, proteins are the sole nitrogen source as nutrient of methanogen in anaerobic digestion of FW. In fact, much of the nitrogen from proteins is converted to ammonia, which is particularly important for pH self-balance of AD system treating FW (Ariunbaatar et al., 2015; Qiang et al., 2012). Therefore, using protease as an additive could accelerate the release of ammonia and timely answer the acidification of FW.

Lipids have a high theoretical methane potential and could consequently increase biogas production (Parawira, 2012), but they have been identified as the main contributor to lag for a low hydrolysis rate. A previous study showed that the lag phase of wastes rich in lipids was about 20.2–48.7 d. This was much longer than that of other wastes (about 14.9–19.9 d) (Lou et al., 2012). A study by Meng et al. (2015) showed that FW in China (rich in lipids such as floatable grease from animal fat and vegetable oil), could greatly benefit from lipase additive during hydrolysis, with methane production increase of 37.0–40.7% in digestion time of 10–40 d (Meng et al. (2017).

The three kinds of solid organics, i.e., starches, proteins and lipids, are always occupy most content of FW simultaneously. Therefore researchers investigated the effect of adding multiple enzymes (α-amylase/glucoamylase, protease, and lipase) on FW hydrolysis and found that it was an effective strategy to improve methane production (Kim et al., 2006). However, the optimal conditions and dosage ratio of these enzymes need to be confirmed case by case, using effect analyses methods, such as response surface analysis (Yan et al., 2011).

3.1.2. Solid organics liquification for possible methane production in high rate reactors

It is worth noting that enzyme additives could not only accelerate hydrolysis rates, but also liquify FW by eliminating the solid contents. It was reported that enzyme additives were used to reduce volatile suspended solids (VSS) by 52.1–61.0% through hydrolysis (Kim et al., 2006; Moon and Song, 2011). At the same time, FW liquification could also bring further improvements on methane production (Shin et al., 2001;

Stabnikova et al., 2008a).

The type of AD reactor, which is restricted by the high levels of suspended solids in FW (~20%), limits AD efficiency greatly with a low OLR. All reported AD processes of FW were performed in continuous stirred-tank reactors with OLRs of 1–9.2 g VS/L.d (Nagao et al., 2012; Wang et al., 2014b). Given that the addition of enzymes increase FW liquification, a breakthrough idea came into mind that high-rate anaerobic reactors (those with OLRs of 10–30 g VS/L.d), such as upflow anaerobic sludge blanket (UASB), expanded granular sludge bed (EGSB), and internal circulation (IC) reactors could be applied for FW. Till now, only FW supernatant from enzyme hydrolysis or fermentative leachate were treated for methane production in high-rate AD reactors (Browne and Murphy, 2014; Wu et al., 2016). No study has tried to introduce liquefied FW, including supernatants and residue solids, into a high rate AD reactor.

Existing research was paused at optimal pH levels, temperatures, and dosage ratios because of the exorbitant cost of commercial enzymes, which was a primary obstacle (Parawira, 2012). Enzyme additives are not currently widely used in biological processes because they are expensive. Therefore, cheap sources of enzymes need to be studied in future research on the application of enzyme additives. Furthermore, enzymatic hydrolytic reaction is efficient in time (less than 24 h) under a suitable environment. Hence, enzyme is usually added into FW before AD, and regarded as a pre-treatment process for feedstock. Whereas, additive is a more appropriate position for enzyme because the method of directly added into the anaerobic digester is convenience for practical engineering (Meng et al., 2015). Maintaining the high activity of enzyme additives in digester with dynamic condition changes, could be a further research trend.

3.2 Fungal mash

Fungal mash, which is rich in exoenzyme secretions and can be produced via the fungi solid-state fermentation (SSF) bioprocess, could be used as a crude enzyme cocktail for FW hydrolysis (Han et al., 2016c; Melikoglu et al., 2013b). Lin et al. (2013) adopted fungal mash to turn FW into a form available for microbes to use directly, which could also be valuable feedstock to produce chemicals, materials, and fuels. In terms of FW anaerobic digestion, Kiran et al. (2015) and Yin et al. (2016) found that fungal mash containing significant glucoamylase and protease could be obtained if *Aspergillus awamori* were used in an SSF process on the surface of waste cake. Kiran et al. (2015) got significant results when using fungal mash as an additive for methane production from FW, ultimately reducing VSS by 64% during hydrolysis and removing 80.4% of the total volatile solids (VS). Pleissner et al. (2014) got 80–90% of solid wastes reduction during hydrolysis using a fungal mash from the SSF of *A. awamori* and *A. oryzae* with blended FW. It can be concluded that, compared with commercial enzymes alone, fungal mash was more efficient due to its multiple enzymes composition.

The low cost of fungal mash increases its prospect of being applied broadly, such as simultaneous biogas and biofertilizer production from hydrolysate and residue solids, respectively (Ma et al., 2017b, c). Nevertheless, it is noted that the production of fungal mash by SSF increased the complexity of AD, with the reaction time taking as long as approximately 6 d (Melikoglu et al., 2013a). However, fungal mash is certain to be a good alternative for commercial enzyme, because of cheap cost, practical use, and high energy recycle value. Much attention should be paid to optimise SSF process in future study.

4. Additives used to maintain pH stability

Anaerobic digestion of FW alone is unstable and often fails, mainly because excess organic acids accumulate during the acidogenesis stage. It results in rapid pH decrease and further inhibition of methanogenic activity (Fisgativa et al., 2016). In general, pH intuitively represents how the dynamic variation of VFAs in a reactor affects AD efficiency. In addition, pH levels play a vital role in regulating the activity of microbes including acidogens (which have a large pH range of 4.0–8.5) and methanogens (with a limiting pH range of 6.5–7.2 and an optimum pH of 7.0) (Sen et al., 2016; Zhang et al., 2014). Therefore, acidogens can ‘trim the sails’, with pH variations relying on microbial adaptation. For instance, a pH level of 6.0 was optimal to produce VFAs in which concentrations of butyrate acid and acetate acid were dominant, while pH 8.0 was controlled for the production of propionic acid (Chen et al., 2013; Wang et al., 2014a).

In contrast, because methanogens are sensitive with a narrow pH range, the process of VFAs consumption could be easily ceased when pH levels drop to 6.5 or lower. However, acidogens can still produce acidic intermediates, leading to the accumulation of VFAs. Therefore, many researchers investigated the performance of multiple additives such as neutralizers, bicarbonates, and buffer material (i.e. zero-valent iron (ZVI)) to maintain the pH stability of AD system treating FW.

4.1 Sodium hydroxide as neutralizer

Adding a neutralizer (e.g. sodium hydroxide) into AD system was identified as an effective method to control the system pH directly and immediately. First, the pH of the FW substrate sometimes needs to be adjusted to neutral when it is around 4.3–4.5 due to the background production of lactic acid and VFAs (Chen et al., 2014c; Kim et al., 2016). After that, there is remarkable acidification in AD system in the first 2 days, especially when the reactor is operated with a high OLR. Wang et al. (2016) reported an innovative pH adjustment program to achieve high VS removal rates using low levels of neutralizer. During the first 2 days, the pH was adjusted once every 16 h, and then once per day at pH 7, with the final VS removal rate of 54.0%. Yang et al. (2015) proved the feasibility of controlling the pH at 8 within the first 5 days to avoid acidification in a thermophilic AD system using FW with a high content of suspended solids, and got 7.57 times increase of total methane production. Adding neutralizers to recover stable pH for anaerobic digestion of FW is a usually inevitable strategy in practical engineering.

4.2 Bicarbonate as buffer

Bicarbonates like $NaHCO_3$ are often recommended to cushion organic acids and maintain appropriate pH levels during AD processes. Compared to neutralizers, adding bicarbonates can achieve equivalent function with only one-time addition, and thus is widely used as a conventional pH control strategy. Gao et al. (2015) found that 1,000 mg/L of $NaHCO_3$ addition enhanced the specific methane production by 48.5% treating residue solid kitchen waste. Nonetheless, the effect of $NaHCO_3$ could only be highlighted with a low inoculum to substrate ratio. In fact, bicarbonates could not only be used as an alternative emergency strategy, like a neutralizer, but also increase system alkalinity and thus promote the self-balancing of pH levels.

Ammonia nitrogen, generated from the protein component of FW along with anaerobic fermentation, was found to act as a good buffer and help the system pH self-balance greatly. Therefore, the potential for pH self-balance based on ammonia nitrogen release in later stage (i.e., the rational C/N ratio in initial feedstock), is crucial to determine parameters such as the required neutralizer or buffer dosage and the frequency of addition. Apparently, the pH self-balance of AD system depending on FW characters greatly, need to be considered at first.

4.3 Zero-valent iron

Zero-valent iron (ZVI) is a novel additive to AD systems and could restore excessive acidification and alleviate low pH through the following pathways: (i) consuming H^+ by ZVI reducibility, as shown in (Eq. (1)) (Daniels et al., 1987); (ii) stimulating performance of microbial metabolism by iron (Hao et al., 2017); and (iii) causing a low oxidation-reduction potential (E_0 = -440 mV) that is beneficial for acetic acid production and butyric, propionic acid conversion (Xiao et al., 2013).

To eliminate H^+, Kong et al. (2016) investigated two types of ZVI (powder and scrap metal); both restrained excessive acidification. With the addition of 0.4 g/g $VS_{FWadded}$ of ZVI to an AD system, the pH of the effluent was 7.8–8.2; without the additive, the pH was close to 5.3. Notably, a delayed recovery period occurred, corresponding to the ZVI dosage, instead of the rapid additive response expected. Yu et al. (2015) found that adding Fe^{3+} to an AD 72 h after start-up could avoid excess acidification. Furthermore, propionic, butyric acid excessively accumulation is a factor causes low pH condition, and also could be alleviated by ZVI addition which function on oxidation-reduction potential change (Feng et al., 2014; Kong et al., 2016). In addition, Kong et al. (2018) proved that ZVI addition was beneficial for dominant microbial species conversion from *Methanosaeta* to *Methanofollis* and *Methanosarcina*, which relieved the accumulation of non-acetic VFAs.

$$\mathrm{ZVI} + 2H^+ + CO_2 \rightarrow Fe^{2+} + CH_4 + H_2O \qquad (1)$$

In general, sudden pH decreasing is a common phenomenon and is difficult to recover for both laboratory experiment and practical engineering. Neutralizers and bicarbonates are both useful for immediately transform of excess acidification. However, the negative effects of using neutralizer and buffer are: (i) the agents are not cheap, suitable dosage and feeding model are still uncertain, and excess heat will release during adding step and may cause activity inhibition of microbes; (ii) the recyclable disposal of biogas residue and slurry will be influenced by additives with further salinity enhancement. In contrast, adding ZVI to avoid acidification outburst could be a green, clean and economic way for anaerobic digestion of FW. For instance, waste iron scraps from industrial residue have been studied as potentially facilitating VFAs generation/translation and methane production. Furthermore, utilizing waste iron in AD system is beneficial for the value of waste recycling accomplish and both biogas residue used as soil amendment.

5. Trace metals as supplement micronutrients

Micronutrients are important to maintain microbe activity and the smooth operation of metabolic pathways (Chen et al., 2008). Consequently, the threshold, stimulation, or limitation of microbe micronutrient concentrations is also important. However, the accurate scope of different nutrients are still not fully realized (Choong et al., 2016). In numerous cases, a common unstable phase appeared in long term anaerobic digestion of FW, so that methanogenesis declined and VFAs gradually accumulated. The crisis can be solved efficiently with the addition of specific TMs, and the methane production can be recovered or even increased (Menon et al., 2017; Zhang and Jahng, 2012; Zhang et al., 2015a). The mechanism and function of TMs in AD were wildly studied, and the method of addition usage was also exploited, which is crucial for actual application.

5.1 The required trace metals

Specific TMs (Fe, Co, Ni, Se, and Mo) are basic metalloenzyme elements that control the processes of acetogenesis and methanogenesis. Hydrogenase (containing Fe and/or Ni) and formate dehydrogenase (containing Fe, Se, and Mo) are two typical enzymes that release electron from H_2 and HCOOH respectively (Banks et al., 2012; Choong et al., 2016). Iron is an important component of ferredoxin, which participates in electron transport, for example in coenzyme F_{420} (Menon et al., 2017). Cobalt has an impact on the activity of methyl transferase, which is a part of methyl transport (Schattauer et al., 2011). Moreover, nickel not only forms carbon monoxide dehydrogenase to take part in aceticlastic and acetogenic reactions, but also serves as a core element for coenzyme F_{430}, which plays an important role in autotrophic methanogenesis (Choong et al., 2016; Takashima et al., 1990; Zhang et al., 2015c).

Generally speaking, conventional elements (e.g. K, Ca, and Mg) are abundant in FW, but several specific elements (i.e. Fe, Co, Ni, Se, and Mo) are generally not enough. As summarized in Table 1, the Fe content in FW (7.17–230.7 mg/kg TS) is higher than that of Co (0.05–0.66 mg/kg TS), Ni (0.42–9.12 mg/kg TS), Se (0.07–0.6 mg/kg TS), and Mo (0.057–1 mg/kg TS). The requirement of TMs to be present in a glucose medium has been investigated by Takashima et al. (2011). More TMs were needed under thermophilic conditions than under mesophilic conditions, i.e., 0.45 vs 0.2 mg/g chemical oxygen demand (COD) removed for Fe, 0.054 vs 0.017 mg/g COD removed for Co and 0.049 vs 0.0063 mg/g COD removed for Ni, respectively. The homologous conversion index of the background TMs level in FW is lower than the threshold. Usually, the inoculum of sludge from a municipal wastewater treatment plant or laboratory-scale AD reactor contains abundant concentrations of needed TMs (Table 1), and guarantees the early stabilization of the AD reactor. The lack of TMs in substrate will appear in a long-term process, with methane production subdued. Hence, an extra supplement of TMs for anaerobic digestion of FW makes a noticeable attention.

In addition, the process of precipitation and dissolution of TMs in complex system of AD, cannot be ignored which is closely connected for bioavailability. TMs present as free ions could easily bond with carbonate, phosphate, and sulphide to form precipitates, while soluble microbial products are likely to restrict freed TMs. For example, ferrous ion will combine with acetate into $Fe(CH_3COO)_2$ and $Fe(OH)(CH_3COO)$ (Thanh et al., 2016; Yu et al., 2015). Whether such transformations are beneficial for TMs bioavailability and storage function, which permits TMs to be dissolved out, is not clear and needs further investigation. In practice, chelating agents like ethylenediaminetetraacetic acid and nitrilotriacetic acid are used to enhance TMs bioavailability in AD system (Hu et al., 2008; Pinto et al., 2014; Vintiloiu et al., 2013). On account of the bioavailability of additives and the environmental health risk, Zhang et al. (2015c) utilized the green chelating agent ethylenediamine-N,N'-disuccinic acid in batch and semi-continuous AD system experiments with FW. When 20 mg/L dose of chelating agent mixed into multi-TMs additives, the TMs dosage decreased by 50% of the optimum (Fe: 100 mg/L, Co: 1 mg/L, Mo: 5 mg/L, Ni: 5 mg/L) and resulted in a 35.5% higher methane production compared to control. The rule of TMs utilization in anaerobic digestion of FW needs further study as effectively decreasing the cost of TMs agent for engineering application.

5.2 Abundant metalloenzyme to improve system stability with high organic load rates

In order to get good effect, low OLRs (1–2 g VS/L.d) are always used at the very beginning to make AD system of FW stable. Higher OLRs (2–6.6 g VS/L.d) can be carried out step by step, till the system deteriorated or biogas production was shut off. Then, TMs can be added as additives to the multiplication of abundant metalloenzymes, which would enhance methanogenesis.

Zhang and Jahng (2012) successfully increased OLRs from 2.2 to 6.6 g VS/L.d during single-phase AD by holding the TMs concentration constant (Fe: 100 mg/L, Co: 2 mg/L, Ni: 10 mg/L, Mo: 5 mg/L); nevertheless, the methane yield decreased from 450 to 352 mL CH_4/g VS_{added}. Here, the lack of critical elements like Se and Mo is considered. According to Facchin et al. (2013), methane production potential was enhanced by 30–40% under a Mo content of 3–12 mg/kg dry matter and an Se content of 10 mg/kg dry matter in batch tests. Similarly, Zhang et al. (2015a) found that despite a supplement of multiple TMs (Fe: 5 mg/L, Co: 1 mg/L, Ni: 1 mg/L), OLRs still rose at 4.0 g VS/L.d as VFAs (30,000 mg/L) accumulated. The addition of an extra 0.2 mg/L of Se improved methane production to 465.4 mL CH_4/g VS_{added} at an OLR of 5.0 g VS/L.d. Furthermore, Banks et al. (2012) managed an AD reactor with a high OLR and discovered that a low background level of Se and Co in FW underlined the significance of their role in oxidizing propionate using the syntrophic interspecies hydrogen transfer pathway with a high ammonia concentration. Fe is also a key factor in maintaining AD system stability. Wei et al. (2014) found that sole-Fe additives could also improve conditions in which a reactor with an OLR of 4.5 g VS/L.d and regular doses of multiple TMs additives were used. In addition, the application of TMs additives in a two-phase AD system with high an OLRs was also studied; however, the contribution of the former to hydrolysis acidification, apart from their role in precipitating S^{2-} in favour of terminal biogas purification, is still not fully understood (Menon et al., 2017; Voelklein et al., 2017).

Overall, the addition of proper amounts of deficient TMs to AD reactors, based on background levels of TMs in FW, could improve the metalloenzyme system and result in good AD performances with high OLRs. Nevertheless, precise dosages could not be determined because of variations in feedstock sources, operating conditions, and reactor structures. Despite this, the relationship between COD and quantity of TMs was surveyed. In thermophilic and mesophilic AD reactors with high OLRs, the value of Fe/COD, Co/COD, Ni/COD were different; 276, 4.96, and 4.43 mg/kg COD were removed, respectively, from the former; 200, 6.0, and 5.7 mg/kg COD were removed, respectively, from the latter (Qiang et al., 2012; Qiang et al., 2013). Using this information, the simulated TM concentrations could be Fe: 5–160 mg/L, Co: 1–10 mg/L, Ni: 1–10 mg/L, Se: 0.2 mg/L, and Mo: 0.2–5 mg/L. These levels are lower than concentration limits.

5.3 Co-digestion with other wastes

Co-digestion of FW with other wastes which are rich in TMs, could be an advantageous alternative than using chemical agents. Anaerobic co-digestion of FW and sewage sludge is an accessible way. Full-scale AD of this co-substrate was early tested in the Europe, and its design and operation (e.g. mixture ratios, temperature, and OLRs etc.) was complex, unclear and more bench researches were proposed (Nghiem et al., 2017). Koch et al. (2015) studied suitable mixture ratio in batch trials and obtained the highest methane yield and production rate at volatile solid-based mixture of FW to raw sludge ratio of 35%. However, Ratanatamskul et al. (2015) found the optimal value of FW to sewage sludge ratio was 7:1 in two-stage AD with total VS removal of 74% and total COD removal of 89%.

Adding landfill leachate (LL) to FW for anaerobic co-digestion could be another pregnant way to enhance the system stability and efficiency for bioenergy recycle. LL contains abundant TMs which could compensate the defect of FW and stimulate microbial activity. Liao et al. (2014) proved that co-digestion of FW and LL was better than FW mono-digestion for high stability with methane yield of 369–466 mL/g VS and FW degradation. In addition, Zhang et al. (2015b) found that co-digestion of FW with fresh leachate compensated for the deficiency of specific TMs (Fe, Co, Ni, and Mo), and acquired a stable methane yield (452.2–506.3 mL/g VS_{added}) at OLRs of 8.1–8.3 g VS/L.d.

6. Functional materials as additives to improve methanogenesis

Recently, numerous studies in the literature concentrated on the role of functional materials to increase AD efficiency via enhanced methanogenesis. The stimulation was comprehensively researched and used mainly the following two aspects: (i) colonization of various functional microbes to decrease lag time, and (ii) change of the finite interspecies electron transfer approach to direct interspecies electron transfer (DIET).

6.1 Carrier function to decrease lag time

Microbes colonized on carrier surfaces with biofilm promotion could be an excellent way to enrich microbes (Luo et al., 2015). Compared with traditional carriers (e.g. zeolite, clay, ceramic, and plastic materials), new materials (e.g. activated carbon and biochar) possess specific surface area, ample pores, and are widely researched as functional carriers for additives in AD processes (Bertin et al., 2010).

Xu et al. (2015) proved that different particle sizes (i.e., granular activated carbon (GAC) and powdered activated carbon) produced similar effects during AD of synthetic brewery wastewater: a shorter start-up time and accommodation of increased OLRs shock in UASB systems. Luo et al. (2015) explored the community distribution of those microbes in solution, tightly or loosely bound around biochar as a dynamic variation of OLRs change and biochar particle size, with the maximum methane production rate raise of 86.6%. Hence, it is clear that carbon-based carrier is benefit for microbe's colonization and functional microbe's enrichment to effectively response the acid-crisis conditions and promote methanogenesis (Wang et al., 2017).

Sunyoto et al. (2016) studied biochar addition to a two-phase AD system for FW and found that lag time for H_2 and CH_4 production decreased by 21.4–35.7% and 41-45%, respectively, as both VFAs degradation and methane production potential were enhanced. Cai et al. (2016) also demonstrated that adding biochar shortened the lag phase by 10.9%-20.0%, 43.3%–54.4%, and 36.3%–54.0%, at inoculum/substrate rate of 2, 1, and 0.8, respectively. Actually, biogas production lag time decrease was not the key role of functional additives; on the contrary, domesticating sludge inoculum is efficient and could be an alternative. However, the effect of carrier additives like biochar on microbial metabolism is important and but less clear, which constrains the additives popularized in the anaerobic digestion of FW. The possibility that carriers are involved in stimulatory effects in addition to participating in methanogenesis, and the mechanism under which this could conceivably occur, will be shown in the following section.

6.2 Conductive function to promote direct interspecies electron transfer

Direct interspecies electron transfer, a new concept for electron transfer approach during these years, which is superior from the conventional H_2 leaded electron transfer pathway that controlled via gas diffusion (Summers et al., 2010). Multiple lines of evidence suggested that conductive function is widely hypothesized to trigger metabolic approach evolution from finite interspecies electron transfer to DIET via conductive pili and c-type cytochrome (Stams and Plugge, 2009; Thauer et al., 2008). In AD systems, sludge aggregates are conductive owing to plentiful pili, which acted as a biological interspecies electric bridge in DIET for syntrophic microbes contact (Rotaru et al., 2014a; Rotaru et al., 2014b). Interestingly, adding diverse conductive materials (e.g. GAC, carbon-cloth, biochar, carbon felt, graphite, and magnetite) could not only supply sites for the sudden microbe colony, as previously mentioned, but also act as excellent electrical conduits to promote robust DIET (Chen et al., 2014a; Chen et al., 2014b; Dang et al., 2016). There is a proposition that a boost in DIET performance would be beneficial for the stability and efficiency improvement of anaerobic digestion of FW. Current research summarized in Table 2 speculated that the potential mechanism includes two aspects: (i) resisting acidic shock from excess propionate and butyrate accumulation, and (ii) shifting electron transfer pathway to enhance methanogenesis.

6.2.1 Acidic shock mitigation

Generally, propionate and butyrate accumulation are widespread and make methanogenesis of FW in AD reactors restricted. In a syntrophic metabolism system, the course in which propionate and butyrate are oxidized into acetate is prevented because of energetically adverse thermodynamics with standard free energy change ($\Delta G^{\circ\prime}$) of +76.0 kJ/mol, and $\Delta G^{\circ\prime}$ of +48.3 kJ/mol respectively under standard condition (i.e. substance at 1 mol/L, pH 7, and 25 °C) (Muller et al., 2010). However, DIET enhancement with conductive materials could resist acidic shock. For instance, carbon cloth supplement gave rise to faster butyrate utilization rate in AD of artificial wastewater (1-butanol) (Zhao et al., 2017b). Furthermore, Dang et al. (2016) discovered that enriched *Sporanaerobacter*, *Enterococcus*, and *Methanosarcina* species on the surface of carbon cloths resulted in faster system recovery when sour appeared during AD of FW surrogate. In the same way, Dang et al. (2017) investigated GAC, carbon cloth additives permitted normal operation of AD of kitchen waste, when VFAs reached extremely high concentration (~500 mM).

6.2.2 Methane production improvement

Inefficient OLRs limits on the anaerobic digestion of FW were discussed in the previous section. Nonetheless, the enhancement of DIET could be a new concept that inferred the permission of high OLRs reactors. Zhao et al. (2015) found that the AD behaviour of ethanol was stable with a supply of conductive materials, and that OLRs increased from 4.1 to 12.3 kg COD/m^3.d in the UASB. In addition, the increased electron transfer efficiency is beneficial for methanogenesis because the electron could take part in CO_2 reduction in direct pathway instead of by relatively long term H_2 shift. For example, methane production was enhanced 12.9%–17.4% when carbon felt and GAC were added to the AD of sludge (Yang et al., 2017; Zhao et al., 2016). Li et al. (2018b) found that thermophilic co-digestion of FW and waste activated sludge could be facilitated via biochar addition which accompanied by the relative abundance of *Syntrophothermus*, *Methanosaeta*, and *Methanosarcina* increased from 3.6% to 4.7%, 30.0% to 43.9%, and 11.1% to15.8%, respectively. Therefore, it is assumed that adding conductive materials to AD systems for FW could permit high OLRs and increase the efficiency of methane production.

To be sure, the specific impact on anaerobic digestion of FW using enhanced DIET with conductive materials was less researched. First of all, the analysis of *Geobacter* and *Methanosaeta* species (Zhao et al., 2017b), and *Sporanaerobacter*, *Enterococcus*, and *Methanosarcina* species (Dang et al., 2016; Dang et al., 2017) were confirmed for syntrophic metabolism in the previous experiments. However, the underlying combination of microbial synergy warrants further detection in comprehensive systems designed especially for the anaerobic digestion of FW. Secondly, among of the different conductive materials, similar excellent performances were shown by GAC, carbon cloth, carbon felt, followed by graphite (Dang et al., 2017). The methanogenesis performance of AD systems of FW needs to be confirmed under additive materials adding with different dosages and physical sizes. Furthermore, the conductive materials like graphite, GAC, and carbon cloth are always too expensive to use as practical additives; in contrast, the biochar, which from thermal treated biomass waste, could be an ideal choose and need in-deep research.

7. Summary and perspectives

7.1 Summary

Additives applied in anaerobic digestion of FW play different roles to resolve inevitable obstacles or to optimize pathways so that the AD system remains stable and efficient with good energy conversion rate. This improvement can be achieved by: (i) promoting rate-limiting hydrolysis; (ii) maintaining a stable pH; (iii) supplying TMs to support microbial metabolism; and (iv) decreasing the lag time and strengthening the DIET pathway. According to literatures, the effect of additives was beneficial to the stability and efficiency of AD systems. However, the relationship between the input cost of additives and the output benefit of energy was not clear from different studies under various conditions. It is possible to conclude that every additive has a less than perfect implementation process and needs further exploration before the economic benefit becomes the focus.

7.2 Perspectives

7.2.1 Trace metals from other wastes to improve reactor stability

Additive TMs play an essential role in anaerobic digestion of FW. On one hand, the accurate demand level of multiple TMs needs to be determined via analysis of changes in bacterial community dynamics under different temperatures, OLRs, and reactor types. On the other hand, the morphology of TMs in complex AD systems needs to be exploited, and improving their bioavailability could effectively decrease the required dosage. Based on the research done, co-digestion could compensate the TMs lack of FW; solve the cost problem of TMs additives; and synergistically treat TMs rich waste with simultaneous bioenergy recover, needs further study as a promising and sustainable way to be applied on FW digestion.

7.2.2 Using high-rate reactor to improve anaerobic digestion efficiency

Food waste is not only a kind of organic solid waste, but also an easily biodegradable feedstock, which supports the feasibility of using high-rate reactors (i.e. UASB, EGSB, IC) with high OLRs. The most limiting factor is the high level of suspended solids in FW. Based on the above review, enzyme additives allow liquification of suspended solids, making it possible to convert FW to bioenergy efficiently. The great advantages of this process include: (i) fast liquification, or breakthroughs in changing the universal rate-limiting hydrolysis which occurred in the

AD of organic solid waste; (ii) eliminating most of the suspended solids organics and avoiding the traditional problem of unstable biogas residue; and (iii) supporting high OLRs operation and improving AD efficiency. The challenges are still highlighted for engineering applications. The amount of multiple enzymes is dynamic change based on unstable FW components. In addition, it is not economically feasible to use commercial enzyme additives. Fungal mash is a great alternative, but fungal bioengineering technologies (e.g. genomics, transcriptomics, proteomics, and interactomics) should be studied further. For example, excellent fungi breeds should be screened and the specific gene function for FW degradation need be strengthened.

7.2.3 Enhancing DIET to improve the stability and efficiency of FW anaerobic system

Direct interspecies electron transfer is an efficient electron transfer pathway compared with interspecies hydrogen transfer, but is little applied in AD systems treating complex organic matter. Its effect on FW, in particularly, is unpredictable and needs specific study to confirm its efficiency and necessity. Biochar could be a preferred functional material, as it can be obtained from biogas residue without extra expenditure. The functional groups in biochar could act as electron shuttles and are as important as its electrical conductivity (for pili), needing further research (Xu et al., 2016).

8. Conclusions

Food waste is becoming a more and more serious problem all over the world, especially for large cities. Anaerobic digestion of FW can not only reduce the solid waste amount, but also convert it to bioenergy. The value of this sustainable and clean way for waste minimization and energy recover, however, always be limited by instability and low efficiency of AD system induced by FW characteristic. Among studies, additives exhibit abundant features to compensate the defect of FW during AD process and wherefore, improve the stability and/or methane yield efficiency. Additives used in anaerobic digestion of FW were systematically reviewed in view of system stability and reactor efficiency. Liquification of organic solids could be greatly improved by adding enzymes, which not only enhance hydrolysis efficiency, but also support possible innovations in the reactor. Fungal mash would be an alternative for expensive commercial enzyme and has a superior effect (80-90% solid waste reduction) for FW hydrolysis, but its catalytic ability and optimization of fungal fermentation process need further study. The gusty pH decreasing is a major obstacle for the stable operation of AD process in practical engineering. Temporarily adding buffer chemicals like neutralizers and bicarbonates into AD reactor is an effective method, with self pH balance help of ammonia nitrogen generated from digestion of proteins in FW. In contrast, ZVI could be an available material from industrial waste, and be more suitable for practical engineering, with further study needed to confirm the specific mechanism of ZVI in acidification regulation. TMs are necessary and always not enough for anaerobic digestion of FW. Rough dosages were calculated based on present studies focusing on long-term reactor operations with high OLRs. In addition, the bioavailability of TMs need to be further studied which greatly influences the efficiency of additives. Using conductive materials to promote DIET is a good idea to increase biogas production and avoid intermediate organic acid accumulation, but further study is needed to examine its application in AD systems of FW. Biochar, recycle from thermal treated biogas residue, could be an ideal choose and need in-deep research between material feature and microbial community. At last, a new concept was proposed for further study, i.e., well liquified FW with pre-hydrolysis by multiple enzymes could be co-digested with landfill leachate (rich in TMs) in high-rate reactors with good stability and high efficiency (OLRs of 10–30 g VS/L.d).

Acknowledgements

This study was financially supported by the National Natural Science Foundation of China (51578329, 51778352), the Science and Technology Commission of Shanghai Municipality (16010500200, 18230710900) and Program for Innovative Research Team in University (IRT13078).

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Figure caption

Fig. 1. The mechanisms of different additives for anaerobic digestion of FW.

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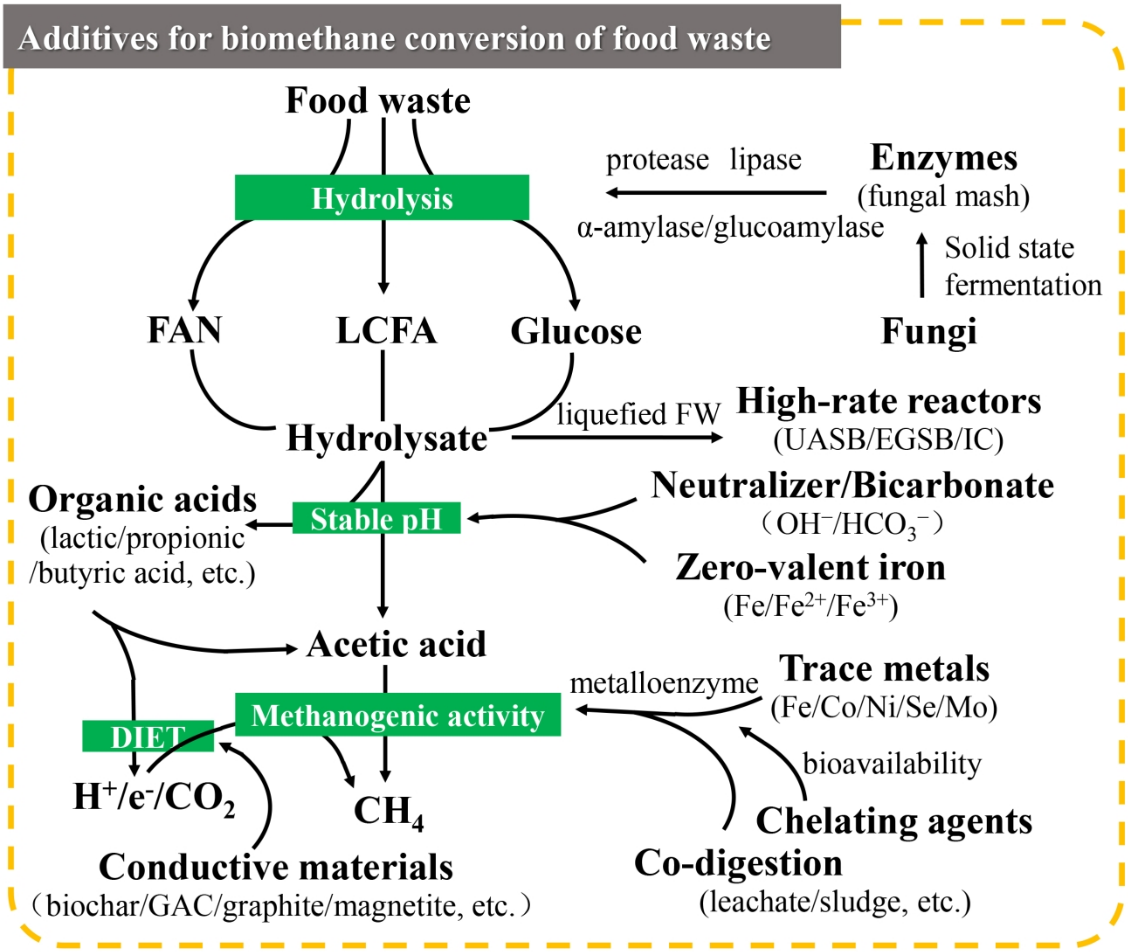

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Table captions

Table 1 Characteristic of specific TMs contents in different FWs and inoculum sludge.

Table 2 Comparison of different conductive additives for DIET enhancement.

Table 1

TMs	**Unit**	**FW**							**Inoculum**	
		China (Zhang et al., 2015a; Zhang et al., 2015c)	**Japan** (Qiang et al., 2012)	**UK** (Banks et al., 2012)	**Belgium** (De Vrieze et al., 2013)	**Italy** (Facchin et al., 2013)	**Ireland** (Voelklein et al., 2017)	**Korea** (Zhang et al., 2011)	**MWTP**[a] (Zhang et al., 2015a)	**FW-AD**[b] (Zhang et al., 2015c)
Iron (Fe)	mg/kg TS	230.7/97.4	286	54	37.3	ND[c]	31.5	7.17	2056	5133.2
Cobalt (Co)	mg/kg TS	0.38/0.14	0.66	<0.06	0.05	<2	ND[c]	ND[c]	3	160.8
Nickel (Ni)	mg/kg TS	6.72/9.12	ND[c]	1.7	0.99	9.6	0.42	0.43	63.7	163.2
Selenium (Se)	mg/kg TS	0.6/0.13	ND[c]	<0.07	ND[c]	<1	ND[c]	ND[c]	ND[c]	4.5
Molybdenum (Mo)	mg/kg TS	ND[c]/1	ND[c]	0.11	0.39	<2	ND[c]	0.057	ND[c]	1186

[a] Inoculum of sludge from municipal wastewater treatment plant.

[b] Inoculum of sludge from laboratory-scale AD treating FW.

[c] Not detected or lower than limit.

Table 2

Substrates	Additives	Reactors	Microbial community	Results	Reference
Artificial wastewater (ethanol)	Graphite, biochar, carbon cloth	UASB	-	Carbon cloth with better performance, Syntrophic metabolism for high-OLRs	Zhao et al. (2015)
Artificial wastewater (1-butanol)	Carbon cloth	Semi-continuous	*G. daltonii, metallireducens, uraniireducens* and *Methanosaeta* species	DIET substituted interspecies hydrogen transfer, 350 mL CH_4/g COD removal, resist acidic impact	Zhao et al. (2017b)
Waste activated sludge	Carbon felt	Microbial electrolysis cell	*Geobacter* and *Methanosaeta* species	Increase 12.9% of methane production	Zhao et al. (2016)
Dog food (food waste surrogate)	GAC, carbon cloth, carbon felt	Semi-continuous	*Sporanaerobacter*, *Enterococcus* and *Methanosarcina* species	Higher OLRs permitted, Faster recovery of soured reactors	Dang et al. (2016)
OFMSW (kitchen waste)	GAC, carbon cloth	Batch	*Sporanaerobacter* and *Methanosarcina* species	Permit extremely high VFAs concentration (~500 mM)	Dang et al. (2017)
Artificial dairy wastewater	GAC, magnetite	Two-phase semi-continuous	*Geobacter* and *Methanosaeta* species	Magnetite promote complex organics decomposition, GAC predominate methanogenic phase with DIET	Zhao et al. (2017a)
FW/waste activated sludge	Biochar	Batch	*Syntrophothermus, Methanosaeta,* and *Methanosarcina*	Shorten lag time and enhance methane production rate at high organic loading	Li et al. (2018b)

Highlights

- Poor system stability and low reactor efficiency are two main problems of AD of FW.
- Additives for AD of FW are reviewed regarding system stability and efficiency.
- Perspectives for future study on application of economical additives are discussed.
- Co-digestion of FW and landfill leachate in high rate reactors is proposed.