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Review

Anaerobic co-digestion of agricultural wastes toward circular bioeconomy

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SUMMARY

A huge amount of agricultural wastes and waste activated-sludge are being generated every year around the world. Anaerobic co-digestion (AcD) has been considered as an alternative for the utilization of organic matters from such organic wastes by producing bioenergy and biochemicals to realize a circular bioeconomy. Despite recent advancement in AcD processes, the effect of feedstock compositions and operating conditions on the biomethane production processe has not been critically explored. In this paper, we have reviewed the effects of feedstock (organic wastes) characteristics, including particle size, carbon-to-nitrogen ratio, and pretreatment options, on the performance of an anaerobic digestion process. In addition, we provided an overview of the effect of key control parameters, including retention time, temperature, pH of digestate, volatile fatty acids content, total solids content, and organic loading rate. Lastly, based on the findings from the literature, we have presented several perspectives and prospects on priority research to promote AcD to a steppingstone for a circular bioeconomy.

INTRODUCTION

Resource depletion and environmental quality degradation are the two major concerning issues pertinent to global sustainable development. Owing to the foreseeable end of the fossil fuel era, the energy crisis has been considered as a prime challenge and opportunity the world faces today. In 2015, the United Nations proposed a blueprint with a total of 17 sustainable development goals (SDGs) for realizing a better sustainable future; one of them is providing affordable and clean energy (known as SDG-7) (UN, 2015). Anaerobic fermentation of organic wastes exhibits many benefits and advantages, such as clean energy production and cost-effective waste treatment. Conventionally, most of the organic wastes, such as food and agriculture wastes, are incinerated, made to composts, or land-filled. However, these processes typically require significant amounts of energy input or large footprint to dispose of the wastes. Instead, anaerobic digestion (AD) has been considered as an alternative for the utilization of organic matters in the biowastes, as well as production of clean energy (i.e., biogas) to meet the SDGs. Namely, the AD process may convert organic components in biowastes into useful bioenergy and biofertilizers, thereby achieving a circular bioeconomy. The aims of the circular bioeconomy are to replace fossil resources with renewable biological resources, such as agricultural wastes. Duque-Acevedo et al. (2020) analyzed the literature published from 1931 to 2018 and found that countries with high income and big agricultural industries have made remarkable advances in the implementation of bioeconomic policies. This means that the deployment of AD processes exhibits indispensable importance and significance.

Byproducts from an AD process include digestate (the liquid part) and biosolid (the solid part). Digestate is abundant in nutrients like N and P and can be potentially utilized as a green fertilizer in agricultural practice. The AD digestate can serve as a green alternative to synthetic fertilizers by recycling nutrients for crop-growing to curb the use of synthetic fertilizers. Similarly, biosolids also exhibit multiple useful functions; for instance, it can be further converted into biochar which can promote the cycles of atmospheric CO_2 and nutrients (e.g., N and P) and, if used as a soil amendment, improve soil fertility. Figure 1 shows the simplified expression of a typical AD process which consists of four stages, viz. hydrolysis, acidogenesis, acetogenesis, and methanogenesis. The produced biogas is a mixture containing several gases, such as H_2, CH_4, CO_2, H_2S, siloxanes, and water vapor (H_2O) (Valijanian et al., 2018). H_2 and CH_4 produced via AD can mitigate climate-change aggravation by providing bioenergy and/or serving as essential

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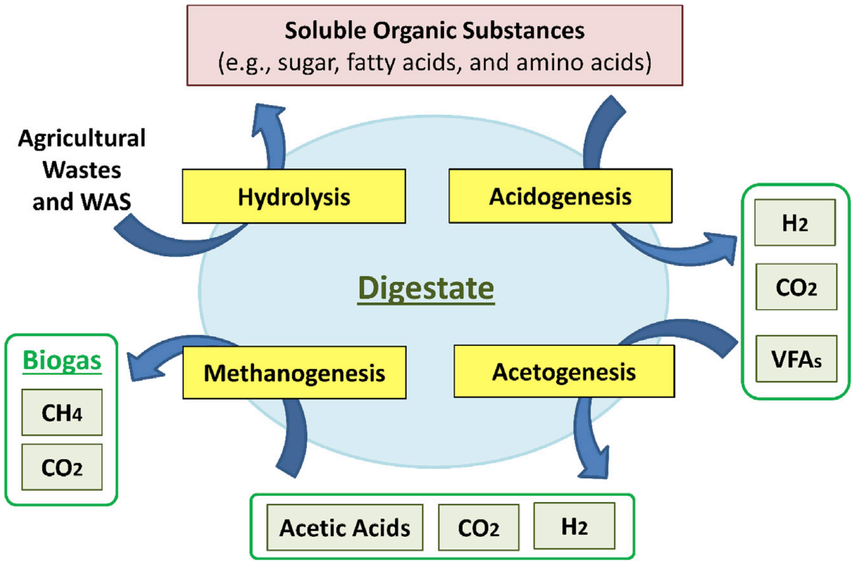

Figure 1. Process-wise stages of biogas production, including hydrolysis, acidogenesis, acetogenesis, and methanogenesis

building blocks for the production of chemicals. In general, the conversion of organic portions of biowastes can be simplified and expressed as shown in (Equation 1):

$$C_cH_hO_oN_nS_s + wH_2O \rightarrow mCH_4 + nNH_3 + sH_2S + (c-m)CO_2 \qquad \text{(Equation 1)}$$

where $m = 1/8(4c + h - 2o - 3n - 2s)$ and $w = 1/4(4c - h - 2o + 3n + 3s)$. The degradable fractions of biowastes ($C_cH_hO_oN_nS_s$) typically include carbohydrates ($C_6H_{12}O_6$), proteins ($C_{13}H_{25}O_7N_3S$), and lipids ($C_{12}H_{24}O_6$) (Mirmohamadsadeghi et al., 2019). These large organics can be degraded by microorganisms into small molecules (e.g., volatile fatty acids (VFAs), sugars, amino acids, and short-chain fatty acids) through hydrolysis. The latter is eventually transformed to CH_4, CO_2, and H_2.

Various types of organic wastes from households, agriculture and food industries, such as cabbage residues, rice straw, corn stover, sugar cane, oilseed cakes, jatropha fruit coat, and ossein industrial waste, can be anaerobically digested to generate biogas and biofertilizers (Habagil et al., 2020). The selection of biowastes for producing biogas mainly relies on their biodegradability and chemical compositions. The optimal operation parameters for dissimilar feedstock substrates in practice should depend on the local climate and forms of feedstock wastes. Kabir et al. (2013) suggested that the utilization of lignocellulosic (agricultural) materials for AD should be a promising alternative to fossil-based refinery due to their abundance, biodegradability, inexpensiveness, and renewable characteristics. According to the Food and Agriculture Organization (FAO) of the United Nations (FAO, 2011), the worldwide food-waste production is estimated at around 1.3 billion tons per year. Verma et al. (2020) explored a worldwide food waste database and found that food waste followed a linear-log relationship with consumer affluence (wealth). Their findings also implied that the FAO's estimate on food waste (FAO, 2011) might be lower than the true value by a factor of 2 or more. Also, over 60% of the primary sparse organic matter flowing into a wastewater treatment plant (WWTP) is concentrated in the form of waste activated sludge (WAS) (Garrido et al., 2013), resulting in the production of an abundant quantity of wastewater sludge around the world. In general, digestion of wastewater sludge (WAS from WWTPs) exhibits less efficient conversion of organic matter to CH_4 (i.e., 30–35%) (Zhang et al., 2014).

Food waste can be co-digested (so-called anaerobic co-digestion [AcD]) with another cellulosic substrate and/or WAS to produce biogas (i.e., 25–50% CO_2 and 50–70% CH_4). The AcD of food waste with WAS provides additional benefits, such as additional buffering capacity, adjustment of nutrient balance (e.g., C/N ratio), and dilution of toxic chemicals (e.g., Na^+). Table 1 summarizes the performance of AcD processes using different types of agricultural organic wastes (including food waste) in the literature. Parawira et al.

Table 1. The performance of AcD using food waste and other organic wastes

Feed substrates	Description of feed substrates (composition)	C/N ratio	Inoculum type	Operation (batch/ continuous)	Max CH_4 yield (mL/g-VS)	Operating pH	References
Food waste	Rice and smaller amounts of flour products, soup, vegetables, and meat	13.9	AD sludge (sewage sludge and food waste)	Continuous (4.0 g-VS/L/d)	494	7.7	(Jo et al., 2018)
Food waste	Rice, meat, tofu, vegetables, fats, and oil	17.5	Thermophilic AD sludge (municipal sludge)	Continuous	364	-	(Shi et al., 2018)
Food waste	Cooked bone: 2.6%, cooked eggshell: 1.3%, pasta/rice: 27.7%, fruit peeling: 20.9%, and cooked vegetable: 24.3%	14.4	Mesophilic AD sludge (WWTP)	Batch (1–6 d)	372	4.5	(Li et al., 2016)
Food waste	Pasta, rice, meat, fruit, and vegetable peelings	14.2	80% cattle slurry and 20% grease trap waste	Continuous	529	4.1	(Browne and Murphy, 2013)
Food waste	Fruit, vegetable matter, pasta, bread, and meat	-	AD sludge (WWTP)	Continuous	380	7.3	(Grimberg et al., 2015)
Food waste	Rice: 15%, noodles: 10%, pork: 10%, chicken: 5%, egg: 5%, cabbage: 20%, potato: 20%, carrot: 13.8%, oil: 1%, and table salt: 0.2%	-	Mesophilic AD sludge (WWTP)	Continuous	407	7.9	(Li et al., 2017)
Food waste and paper waste	Food waste (fruits: 30%, vegetables: 36%, meat/fish/ egg: 14%, and rice/noodles: 20%) with paper waste (new toilet paper, used office paper, and used newsprint at the ratio of 1:1:1)	15.2	Mesophilic AD sludge (WWTP)	Semi-continuous (40 d)	460	4.2	(Xiao et al., 2019)
Food waste and cattle manure	-	15.8	Activated sludge	Batch (18 d); Semi-continuous	388	7.5	(Zhang et al., 2013)
Organic fraction of municipal solid waste and fruit/vegetable waste (at a ratio of 1:3)	Organic fraction of municipal solid waste: egg shells, coffee powder, carrot and chayote peels, lettuce and arugula leaves, beans, rice, pasta, and bread. Fruit/vegetable waste: banana 20%, papaya 20%, apple 10%, cabbage 12%, lettuce 12.5%, onion 12.5%, and potato 12.5%.	34.7	Mesophilic AD sludge (food waste)	Batch (12–18 d)	397	7.4–8.2	(Pavi et al., 2017)

(2005) performed the AcD of solid potato waste (starch-rich) with sugar beet leaves and produced biogas with the CH_4 content of about 84%. The cumulative CH_4 production by an AcD process run at the inoculum-to-substrate ratio of 115 and with substrate containing TS of 40% was 31–62% higher than that by mono-digestion. For the properties of feedstock, the average C/N ratio of feed substrates resulting in the maximum methane production was 18.0 ± 6.9 ($p < 0.05$; $n = 7$), as shown in Table 1. The average methane production from AcD of food wastes with other organics, such as cellulosic substrates and WAS, was calculated to be 421 ± 45 L/kg-VS ($p < 0.05$; $n = 9$).

With AD processes, Duque-Acevedo et al. (2020) indicated that the fundamental aspects, such as research, innovation, and technological development, are the imperative elements in the progress of the transition to circular economy system. The performance of an AcD process should be largely related to the properties of its feedstock (e.g., the C/N ratio) and operating conditions (e.g., pH and temperature). Refining operation parameters should be carefully carried out since extreme changes would significantly cause a negative impact on the performance of an AcD process.

Despite the recent progress in the AcD technology, the effect of feedstock and operating conditions on biogas production from an AcD process has not been critically evaluated. In this review, therefore, we first investigate the effects of factors related to feedstock (organic wastes), including particle size, C/N ratio, and pretreatment options, on the AcD performance. An overview of potential control parameters, such as retention time, temperature, pH, VFAs, organic loading rate (OLR) and additives, for AcD processes is provided. We also review the recent development in model-based process control schemes and system optimization of an AcD process, as well as the microbial community analysis for elucidating AcD mechanisms and pathways. Lastly, a few perspectives and prospects on the future directions of AcD research are proposed to facilitate the movement toward a circular bioeconomy.

FACTORS RELATED TO FEEDSTOCK

Agricultural wastes can be categorized into two major types: (i) food, and (ii) non-food (inedible) portions of crops, such as rice husks, wheat (paddy) straws, groundnut shells, corn stover, corn cobs, woods, leaves, stalks, and orchard trimmings. The performance of an AcD process highly depends on the types of feedstock as different types of agricultural wastes exhibit diversified physico-chemical properties. In this section, the effect of organic feedstock (wastes) properties, including particle size and C/N ratio, on the AcD performance, was reviewed.

Particle size

Prior to the AcD process, the ensiling stage plays an important role in determining the properties of agricultural waste. Mohd-Setapar et al. (2012) investigated the effect of chopped length (particle sizes ranging from 2 to 30 mm) on the properties of agricultural waste during the ensiling process. They found that the chopped length did not notably influence the property of the produced silage. In the case of amaranth crop silage, however, smaller particle size may lead to a high dry matter loss of biomass while ensiling the crop (Haag et al., 2015). Nonetheless, the particle size of agricultural waste would significantly affect the methane production from the subsequent AD process. Extensive studies have revealed that the particle size would be directly related to the kinetics of hydrolysis of complex substrates (Silvestre et al., 2015; Weide et al., 2020). However, there is no rule-of-thumb regarding the particle sizes to follow due to a wide research gap in the physico-chemical properties of different agricultural wastes. In general, the decrease in particle size benefits the methane production as a higher available specific surface area would result in the reduction of both polymerization and cellulose crystallinity (Menardo et al., 2012).

A reduced particle size may facilitate the lactic-acid bacteria fermentation, thereby avoiding significant loss of organic matter. For instance, Herrmann et al. (2012) evaluated several crops, such as sorghum, forage rye, winter rye, maize, and triticale, and found that the methane yield could be enhanced by reducing the chopping length; an increased methane production (CH_4 content of 53–63%) could be observed when the size of particles was decreased to 7–8 mm, demonstrating that the decreased chopping length could lead to a more extensive generation of lactic acid. However, Haag et al. (2015) found that further reducing the chopping length from 8 to 1 mm would not exhibit any additional benefit. They suggested that the low CH_4 yield (in the case of a 1–mm chopped crop) may be attributed to the less lactic acid generation during ensiling. A similar result was observed in the case of corn stover; when the particle size was reduced from 0.42-0.84 to 0.18-0.42 mm, significantly lower methane and biogas yields could be observed

(Ajayi-Banji et al., 2020). In addition, a digester can be easily clogged in the case where fine particles are fed, which should be carefully considered in the design.

Chemical properties

The chemical properties of feedstock, especially the contents of volatile solids (VSs) or biodegradable organic fractions, are directly related to the biogas production in the AcD process. It is noted that the contents of dry matter (or total solids, TSs) and VS in feedstock should be distinguished. For most cases, VS is considered as an approximation of the content of organic matter. Apart from the amount of VS, a sufficient retention time should be provided for microorganisms to transform organic matters to biogas. Therefore, OLR, an operating parameter of an AD system, should be carefully determined.

In addition to VS, the C/N ratio plays a crucial role in biogas production. The C/N ratio represents the relative amount of carbon over nitrogen present in a substrate and has been considered as one of the major operating parameters in a variety of biological treatment systems. In the AcD process, the carbon source is mainly used for CH_4 production, while nitrogen is used by methanogens for their protein essentials. A high C/N ratio implies that the formation of protein in microbes would be limited by nitrogen (Zhu, 2010). Conversely, excessive nitrogen (i.e., a low C/N ratio) would lead to the accumulation of NH_4^+ via the nitrogen liberation (Khalid et al., 2011). Free ammonia (NH_3) would, if present in excess, inhibit the growth of methanogens.

The AcD process using multiple types of feedstock could overcome the process instability due to the inappropriate C/N ratio in the case of using single feedstock. Feedstock exhibiting a lower C/N ratio can be blended with that exhibiting a higher C/N ratio to create a suitable condition for microbial growth. Table 2 summarizes the C/N ratio and other physical and chemical properties of various organic wastes used in the literature. Agricultural (plant) wastes, such as sawdust and wheat straw, usually contain a large amount of carbon, thereby exhibiting a relatively high C/N ratio (>100). In contrast, animal wastes, such as cattle dung, swine manure, and human excreta, have a low C/N ratio (usually less than 25). In fact, the effect of the C/N ratio on biogas production by an AcD process has been extensively investigated. Siddiqui et al. (2011) noticed that the desirable C/N ratio would be rather wide-ranging between 9 and 30 for AD. Similarly, Panichnumsin et al. (2012) suggested that the optimum C/N ratio for the AcD should be within 20–35. Borowski et al. (2014) also found that the suitable C/N ratio for the AcD of municipal sewage sludge and swine/poultry manures should be 20–30; they achieved a maximum biogas yield of 0.4 m^3 per kg-VS. Gerardi (2003) also suggested that the C/N ratio should be designed to be > 25 to achieve stable gas production.

PRETREATMENT OPTIONS

Lignocellulosic substances, such as agricultural residues and industrial wastes (e.g., paper wastes and municipal solid wastes) exhibit a great potential for biochemicals and/or bioenergy production due to their abundance around the world. However, lignocellulosic substances are typically difficult to be decomposed and then utilized by microorganisms due to the crystalline structures of their three major building blocks, viz. lignin, cellulose, and hemicellulose. Pretreatment of the feedstock could effectively improve the biodegradability of lignocellulosic substances and augment the availability of biodegradable substrates. Consequently, it is essential to deploy an appropriate pretreatment process to break down the lignocellulosic, thereby increasing the contents of carbohydrates and thus subsequent biogas production.

Previous studies revealed that improvements in pretreatment techniques would raise the microbial availability of inexpensive feedstock, and increase biogas production (Song et al., 2012). Various pretreatment options, such as physical, chemical, physico-chemical, and biological methods, are available for treating lignocellulosic substances in the feedstock. Among them, biological pretreatment has been considered as one of the most environmentally friendly approaches due to the advantages of cost effectiveness, low energy consumption, higher specificity, and environmental welfare (Divya et al., 2014).

In most cases, agricultural residues are anaerobically co-digested with WAS to adjust the C/N ratio. However, a few studies report that sludge possesses hard cell walls and complex floc structures (e.g., extracellular polymeric substances) which causes a less efficient decomposition of organic solids and requires a long retention time (Abelleira-Pereira et al., 2015). It has been noted that hydrolyzing particulate organic matter into soluble substances should be the rate-limiting step for the AcD of WAS (Cano et al., 2015). If

Table 2. Physico-chemical properties of different types of organic wastes

		Elemental (%)							Component (wt.%) [a]				
Category	Type	C	H	O	N	S	TS (%)	VS (%)	CL	HCL	LN	C/N ratio	References
Agricultural waste	Corn stover	42.1 ± 0.4	5.88 ± 0.07	-	0.95 ± 0.07	0.64 ± 0.01	93.8 ± 0.2	86.2 ± 0.2	-	-	-	44.3	(Liu et al., 2019)
	Leaves	43.7 ± 1.2	5.82 ± 0.13	39.9 ± 0.9	1.06 ± 0.22	0.06 ± 0.01	-	-	22.3	34.3	18.4	41.2	(Intani et al., 2016)
	Corn stalk	41.9	5.72	42.0	0.50	-	-	-	29.1	26.0	15.0	83.8	(Hu et al., 2019)
	Teff straw	-	-	-	-	-	91.6 ± 0.4	84.3 ± 0.1	36.7	32.4	9.4	-	(Chufo et al., 2015)
	Sawdust	48.3	6.21	44.0	1.50	-	-	-	19.6	27.2	51.5	32.2	(Li et al., 2012)
	Wheat straw	45.0	5.70	44.6	0.44	0.37	-	-	37.5 ± 0.4	21.2 ± 0.1	21.3 ± 0.1	102.3	(Ma et al., 2018)
	Spent ground coffee	52 ± 3	7 ± 0	37 ± 0	2 ± 0	0 ± 0	95 ± 2	92 ± 2	68.9	29.4	4.2	23.8	(Atelge et al., 2021)
Food waste	Food waste	42.7	9.1	46.2	1.97	0.3	22.6 ± 0.8	21.2 ± 0.1	29.2 ± 3.8	11.2 ± 1.2	3.4 ± 0.8	21.6	(Panigrahi et al., 2020)
	Orange bagasse	41.6	-	-	1.38	-	19.2	18.3	15.2	6.61	1.35	30.1	(Santos et al., 2020)
	Food waste	50.6 ± 0.5	6.6 ± 0.3	39.0 ± 0.6	2.3 ± 0.4	-	-	-	-	-	-	22.0	(Gupta et al., 2019)
Animal waste	Swine manure	37.05 ± 0.06	5.84 ± 0.02	-	3.04 ± 0.02	-	25.87 ± 0.05	20.82 ± 0.01	28.91 ± 0.05 [b]	-	23.05 ± 0.03	12.2	(Yu et al., 2020)
	Cattle manure	32.1	-	-	1.65	-	18.8	15.6	22.3	18.9	12.9	19.5	(Şenol et al., 2020)
	Cow dung	36.2	5.10	-	1.20	-	-	83.0	-	-	-	31.6	(Prajapati et al., 2014)
	Hay and horse manure	46.6	-	-	1.40	-	19.9	17.5	-	-	-	33.3	(Böske et al., 2014)

[a]CL: Cellulose; HCL: Hemicellulose; LN: Lignin.
[b]the value includes cellulose and hemicellulose (%TS).

a pretreatment technology is properly selected for WAS and agricultural (food) waste, it can accelerate the emancipation of intracellular substances by breaking the cell wells and making them more available to microbes. Therefore, different types of pretreatments, such as chemical (Zhen et al., 2014), thermo-chemical (Wang et al., 2014), thermal (Kinnunen et al., 2015), ultrasonic (You et al., 2019), enzymatic methods (Pei et al., 2010), microwave (Yue et al., 2021), or their combinations (Ariunbaatar et al., 2014), have been applied for WAS.

OPERATING PARAMETERS RELATED TO DIGESTORS

The AcD of agricultural wastes and WAS exhibits four important benefits, compared to the AD of agricultural wastes as a sole feedstock (Esposito et al., 2012): (i) alteration of moisture content, C/N ratio, and pH of feedstock, (ii) an increase of biodegradable organics content and buffer capacity of feedstock, (iii) reduction of toxic compounds and/or potentially inhibitory effects, and (iv) an increase in system resilience as a result of more diverse microorganism species. The optimization of control parameters plays an essential role in maximizing the performance of an AcD process. In this section, we provide an overview of the effect of five important control parameters on AcD processes, including retention time, temperature, pH, VFAs, and OLR.

Retention time

The biogas production by AcD largely relies on the retention time (Gerardi, 2003), which is considered as one of the most principal operating parameters that control the transformation of VS into CH_4, as well as the microbial growth rate. For batch tests, the methane production rate generally increases during the initial stage, and then gradually decreases afterward (Santosh et al., 2004). For evaluating the residence time, two types of performance indicators are commonly used in the biological process: (i) solids retention time (SRT): a measure of the time length for microbes (or solid wastes) to stay in a bioreactor, as described by (Equation 2), and (ii) hydraulic retention time (HRT): the average time that the digestate remains in a bioreactor, as determined by (Equation 3):

$$SRT = \frac{V \times C_d}{Q_e \times C_e} \qquad \text{(Equation 2)}$$

$$HRT = \frac{V}{Q_i} \qquad \text{(Equation 3)}$$

where V is the volume of a bioreactor (digester, m^3), C_d is the VS concentration in the reactor (kg/m^3), C_e is the VS concentration in the effluent (kg/m^3), Q_e is the effluent flow rate (m^3/s), and Q_i is the influent flow rate (m^3/s).

For the AcD process, the SRT is usually equal to the HRT due to no recirculation of microbes in the effluent back to the digestor; therefore, $Q_i = Q_e$. The HRT of a digester varies from a few days to months, depending on the substrate types and process configurations. A longer retention time commonly yields a higher cumulative methane production, as well as leads to a greater reduction of the total VS. A long retention time can also help microbes adapt to noxious compositions (Gerardi, 2003). A large digester volume would be necessary for a long HRT as a short retention time may lead to the washout of microbes. In the case of a short HRT, the rate of microbe loss may exceed the rate of bacterial growth, thereby making the AcD process fail. In addition, a short HRT usually resulted in the accumulation of VFAs in the digester. Dinsdale et al. (2000) used fruit and vegetable wastes with WAS as feedstock to evaluate the performance of a two-stage AcD. They found that the contents of VFAs in the effluent were significantly reduced if the HRT was increased from 10 to 13 days. However, a decrease in retention time would reduce both the capital (decrement in the digester volume) and operation costs of a biogas plant (e.g., costs associated with gas production and purification) although complete digestion might not result.

For an engineering design, the HRT of an AcD process is usually determined by a number of factors, such as the natures of feedstock substances, types of mixing, operating temperature (usually depending on the types of inoculum), process configurations, weather (climate), and even utilization pathway of digestates. Especially, the types of feedstock for an AcD process would greatly affect the retention time of a digester. For instance, wastewater from food industry usually comprises soluble organics, such as sugar and starch-rich molecules that are easily digested in an AcD process, mainly because the hydrolysis stage can be completed in a shorter time. In contrast, the feedstock from fibers and cellulose-containing plants would require a relatively longer retention time. In addition to the feedstock, the HRT is greatly affected by the types of mixing. A number of mixing strategies such as air-mixing (Yang and Deng, 2020) and ultrasonication (Ambrose et al., 2020) have been evaluated and applied to accelerate the bioconversion and thus the biogas production. Zhang et al. (2020) found that continuous mixing followed by intermittent mixing should be an alternative operating strategy to optimize biogas production and process energy consumption.

In terms of the retention time, the retention time of 15-30 days is recommended for a single stage mesophilic AcD plant (Monnet, 2003), or 15–40 days for a two-stage mesophilic plant (Olsson, 2012). In contrast, a thermophilic AcD would require a shorter retention time (~14 days) for complete digestion of organics (Sung and Santha, 2003). Nonetheless, Mara and Horan (2003) suggested that the HRT should be maintained at > 10 days. Regarding the effect of climate conditions, the HRT may be as long as 100 days in the region of cold climates, while it can be 30–50 days in a warm climatic environment (Kigozi et al., 2014). In addition, the utilization pathway of the digestate (especially the solid portion) relies on the HRT of ADs. The retention time should be relatively longer if the final digested sludge would be landfilled, while the former can be relatively shorter if the latter would be incinerated.

Temperature

As aforementioned, the operating (ambient) temperature would affect the duration of fermentation (or retention time) and biogas production. The growth rate, metabolism, and population dynamics of microorganisms highly depend on the operating temperatures. Therefore, the operating temperature of the AcD process should be carefully designed since methanogens are quite temperature-sensitive. Appels et al. (2008) found that severe process failure would occur if the temperature fluctuation of the mixed liquor in an AD is above 1 °C per day. To avoid localized temperature variation, sufficient stirring or mixing of the digestate is needed. In general, the operating temperatures of psychrophilic, mesophilic, and thermophilic digestions are around 25, 35, and 55 °C, respectively (Ogejo et al., 2009). Several studies demonstrated that psychrophilic methanogens could grow even at 10–20°C (Wang and Wan, 2009). A small-scale AcD reactor (e.g., Imhoff tanks, septic tanks, and sludge lagoons) has been frequently applied in the range of psychrophilic temperature. It does not require extensive heating as the temperature of digestate is similar to that of the surrounding environment. However, the biogas production would virtually cease if the temperature of the digestor is lower than 10°C.

At the temperature range of 20–45 °C, mesophilic microorganisms become active and produce methane (Balasubramaniyam et al., 2008). Ehimen et al. (2011) reported that the methane content of biogas produced from the AcD of algal remnants increased from 54% to 61% when the temperature increased from 25 °C to 35 °C. In general, the temperature of a traditional mesophilic AcD process is maintained at 35–37 °C, while several recent studies suggest that the temperature for the mesophilic degradation of agricultural wastes should be 21–40 °C (Weide et al., 2020; Zhang et al., 2019b).

Compared with the mesophilic AcD process, the thermophilic one typically exhibits several advantages, including the greater capacity of biogas generation, faster reaction rate, higher degradation rate of pathogens and cell walls in feedstock (such as weed seeds), and better separation of digestate into solids and liquid phases. Ward et al. (2008) reported that degradation of fatty acids at 55 °C (thermophilic) was much faster than that at 38 °C (mesophilic). In their batch study, only 11 days were required for a thermophilic AD reactor to achieve 95% of the maximum methane yield, compared to 27 days requirement for mesophilic ADs (Ward et al., 2008). Although thermophilic AcD processes could generate a greater amount of biogas, they require higher energy input for maintaining the mixed liquor at a high temperature (Karagiannidis and Perkoulidis, 2009). A slight change in the reaction temperature would significantly influence the thermophilic AcD process (El-Mashad et al., 2003). Therefore, one of the major issues of thermophilic AcD systems is to maintain operating temperature at the optimum for achieving a good cellulose degradation.

pH of digestate

The pH of digestate markedly influences the performance of an AD process (Hagos et al., 2017), i.e., the growth and activity of microorganisms. However, after the hydrolysis stage, the pH of the digestate drops significantly due to the accumulation of VFAs (Kim et al., 2003). A similar phenomenon is also observed in the acidogenesis stage of a two-stage AD system. It has been noted that the methane-forming bacteria would start to consume the VFAs when the HRT is greater than 5 days. Nonetheless, the pH of the digester needs to be maintained at 6.8–7.2 to ensure an effective transformation of VFAs into CH_4 and CO_2. On the other hand, CO_2 is continuously produced during the AcD process, and the concentration of generated

CO_2 also influences the pH of the digestate (Gerardi, 2003). The increase of CO_2 concentration during the acidogenic phase would cause the pH to drop because excessive CO_2 can be dissolved in the digestate. At the same time, the produced VFAs also make the system pH drop below 6.0. A pH lower than 5.0 would in turn have a negative impact on the hydrolysis and acidogenesis stages. Therefore, continuous monitoring and adjustment of the digestate pH should be imperative to ensure a "healthy" AcD performance.

Automatic pH controllers have been extensively applied to monitor and maintain the pH of an AcD process (Yasin et al., 2011). For the pH adjustment, the addition of alkaline, such as KOH, is commonly employed. Aside from the chemical additives, the digestate pH can be maintained at a certain range by controlling OLR. In practice, the pH of the digestate is adjusted to a specific value once a certain period, e.g., every 24 hr (Fang et al., 2006), for obtaining the optimum operation (Tawfik and El-Qelish, 2014). In the case where an AcD process is run at low initial pH values of 4.5–5.5, buffers (e.g., $H_2CO_3/HCO_3^-/CO_3^{2-}$) and/or nutrients should be supplied to increase the alkalinity of digestate to maintain its pH at neutral range (Marone et al., 2014). Recently, several innovative approaches, such as electrochemical separation (Pan et al., 2016), have been applied as a side treatment to avoid the accumulation of VFAs in the digestate, and thus continuous pH-drop during AcD.

For AcD, it is imperative to maintain the pH of the digestate at the optimal range for each individual reaction stage (e.g., hydrolytic, acidogenic, acetogenic, and methanogenic) in order to achieve the overall system optimization. The pH of the digestate largely depends on the formation rate of intermediates during AcD. At the beginning of AcD, acidogens, and acetogens would create organic acids (e.g., VFAs) and acetate, respectively (Kumar Das et al., 2020), thereby decreasing the pH of the digestate. Parawira et al. (2005) indicated that acidogenesis and acetogenesis would accumulate the organic acids, where the pH of the digestate could eventually drop below 5. During methanogenesis, methane-forming bacteria would consume the VFAs and CO_2 to generate alkalinity and CH_4, resulting in the increases of the digestate pH (Gerardi, 2003). The control of the system pH may provide an effective approach to reduce the risk of the occurrence of AcD inhibitors in the digester. For instance, a slightly acidic environment would enhance the protonation of ammonia to form NH_4^+ as described by (Equation 4), which can accumulate in the digestate. The accumulated NH_4^+ can be reversed to NH_3, which is harmful to methanogens, if the system pH increases. It was noted that ammonia inhibition would occur if the digestate pH is above 7.4 in the case where TN of digestate is 1.5–3.0 g/L or higher (McCarty, 1964). A low C/N ratio of the feeding substrate for an AcD process could also potentially lead to the production of toxic ammonia.

$$NH_3 + H^+ \rightarrow NH_4^+ \qquad \text{(Equation 4)}$$

Excessive ammonia and/or VFAs accumulation would eventually inhibit the activity and metabolism of methanogens. Therefore, different process designs, such as the two-stage configuration (Ward et al., 2008) (the first stage: hydrolysis-acidification; the second stage: acetogenesis-methanogenesis) has been developed to overcome the process hurdle. Acid-forming bacteria would be active in the pH range of 4.0–8.5; however, the optimal pH for hydrolysis and acidogenesis is reported between 5.5 and 6.5 (Appels et al., 2008; Ogejo et al., 2009; Ward et al., 2008). Horiuchi et al. (2002) studied the digestion of glucose under the acidic and neutral environments, and they found that different types of organic acids were produced at different pHs. In contrast, methanogens are extremely susceptible to the change of pH, thereby affecting methane production (Normak et al., 2015). Most methanogenic microorganisms (majority anaerobes) work in the pH range of 6.6–7.6, with an optimum pH of approximately 7.0. Several studies showed that the growth rate and metabolism of most methanogens significantly would decrease if the pH value was lowered below 6.6 (Ogejo et al., 2009; Ward et al., 2008). Only *Methanosarcina* could survive the acidic environment of pH < 6.5. On the other hand, the methane production would also decrease if the pH would be maintained at 7.0 to 10.0.

VFAs in mixed liquor

The total content of VFAs is another critical factor, apart from the system pH, in the methane production via AcD. During the acidogenesis and acetogenesis stages, the catalytic decomposition (metabolism) of organic solids would produce the intermediates and/or final products like VFAs (Hassan et al., 2016; Rani et al., 2014). In the subsequent methanogenic stage, if the system pH is lowered below 4.0 by accumulating VFAs (or sometimes organic acids), the biogas production would be inhibited (Fang et al., 2006; Rani et al., 2014). Under such circumstance, the methanogens cannot consume VFAs and hydrogen effectively and VFAs will accumulate in the mixed liquor (digestate) and lower the system pH, as well as deplete the system buffering capacity. The concentration of VFAs has been used as a key indicator to identify the levels of the AcD destabilization and a constraint on methanogens growth (Hassan et al., 2016).

The process performance of hydrolysis and acidogenesis stages is highly related to the pH and the VFA content. However, it is still unclear if the accumulation of VFAs, or the pH decrease inhibits the hydrolysis. Several studies indicated that the accumulation of VFAs may decrease the hydrolysis of solid organic substrates, regardless of the digestate pH (Rajagopal et al., 2013). In either case, VFAs and pH can be controlled individually to avoid inhibition during the system operation. Various approaches have also been proposed to overcome the VFA issue during the AcD operation. For instance, Haider et al. (2015) found that the VFA accumulation (TS in the digester) and OLR play critical roles in biomethane production of an AcD process. Depending upon the TS content in a digester, the AD system could be classified into three types: high solids content (28–33%), medium (19–28%), and low (10–14%) (Motte et al., 2013). It is noted that the trace nutrient limitation and ammonia toxicity would be significant if the TS content of digested material is greater than 25% (i.e., high solids content). To overcome this issue, Jewell et al. (1993) controlled the C/N ratio of feeding substrate and trace nutrient content to ensure a steady AD performance. TS content of the digester also reflects the amount of VS available to microorganisms. However, there is no rule of thumb to determine the optimal TOC content in a digester, as it highly depends on the type of feedstock and inoculum and the operating parameters of the AcD, such as the HRT and operating temperature. For instance, Paramaguru et al. (2017) investigated the effect of solids content (i.e., 5%, 10%, 15%, and 20%) on biogas production, and found that the solids content of 10% exhibited the highest biogas production among all treatments. In their study, De Schamphelaire and Verstraete (2009) observed the higher biogas yield was obtained at 0.6 g-VS per liter of sludge. Ehimen et al. (2011) recommended that the most effective substrate concentration in a digester would be 5.0 g-VS/L. They also found that, regardless of the HRT and the C/N ratio of feed substrates, the VFA concentrations greater than 5 g/L was observed in the case where the substrate concentration was over 40 g L^{-1}.

In addition, the TS content of a digester deserves careful attention since it reflects the physical properties of the digestate, such as the density, viscosity, and rheology. For instance, Karim et al. (2005) reported that the extent of mixing was an important factor for a digester operated at a high solids content (same to a high OLR) while no apparent effect was observed for a digester operated at a low solids content. Similarly, the intensity of the mixing should be properly enhanced as the viscosity of the digestate increases.

Even in the case an AcD reactor is periodically supplied with organic wastes (feedstock), the biomethane is continuously produced. To evaluate organic components in the feedstock, the OLR is defined as the amount of chemical oxygen demand (COD) (see (Equation 5)) or VS (see (Equation 6)) fed into the digester, with the unit of mass per unit volume of the digester per unit of time.

$$OLR = COD_i \times \frac{Q_i}{V} = \frac{COD_i}{HRT} \quad \text{(Equation 5)}$$

$$OLR = VS_i \times \frac{Q_i}{V} = \frac{VS_i}{HRT} \quad \text{(Equation 6)}$$

where COD_i is the COD of inflow solid wastes (kg-COD per m^3), and VS_i is the VS of inflow solid wastes (kg-VS per m^3).

The biogas yield generally increases with the increasing OLR. For instance, Tanimu et al. (2014) evaluated the effect of the OLR ranging 1.0 to 6.1 kg-VS/m^3/d on the performance of a thermophilic AcD system fed with food wastes. They found that the optimum OLR was approximately 2.1 kg-VS/m^3/d. A high OLR could significantly reduce the size of a digester and thus the capital cost. However, an excessive OLR may result in digestion inhibition due to a high formation rate of C4–C5 VFAs (e.g., valeric and butyric acids) (Gerardi, 2003), by hydrolysis and acidogenesis. Therefore, a low OLR is recommended for the feedstock of a high VS content, e.g., food wastes and agricultural residues, and the OLR around 1–4 kg-VS/m^3/d is commonly used for an AcD system (Atelge et al., 2021).

Additives

During the AD process, microbial growth requires not only the macronutrients (such as carbon and nitrogen) but also a variety of trace nutrients, including metals and vitamins. These trace nutrients (micronutrients) are important substances to maintain the growth and metabolism of anaerobes. The microbial activity would be significantly inhibited if the trace elements are insufficient. Therefore, these micronutrients should be sometimes supplemented. Several reports also indicated that the supplementation of additives containing trace elements could effectively reduce the accumulation of VFAs and thus improve the microbial activity in an AD system (Moestedt et al., 2016).

Trace metals, e.g., iron (Fe), cobalt (Co), copper (Cu), molybdenum (Mo), selenium (Se), manganese (Mn), Zine (Zn), and nickel (Ni), are frequently utilized to promote the performance of an AD system. Especially, Cai et al. (2018) reported that the order of the bioavailability of trace metals would be as follows: Se > Mn > Zn > Ni > Mo > Cu > Co > Fe. However, the required amount of each metal and its function are still not well understood. In their study, Shamurad et al. (2020) found that trace metals, including Fe, Co, and Ni, were required for acetogens to produce metalloenzymes which are used for producing formate dehydrogenase and carbon monoxide dehydrogenase. They also indicated that all methanogens would need nickel for the synthesis of cofactor F_{430} to produce methane (Shamurad et al., 2020). Trace nutrients, such as vitamins (cobalamin), would also enhance the activity of enzymes in an AD system, thereby accelerating the biosynthesis of CH_4. These trace elements are part of the cofactors of enzymes or coenzymes involved in the growth of microorganisms and the synthesis of CH_4.

DEVELOPMENT OF KINETIC MODELS

As aforementioned, the AD is a process with complex biochemical and physico-chemical reactions, involving different microbial species to degrade and transform organic matters and intermediate products. During the AD process, factors related to the substrate properties and operating conditions could interact with one another to influence the performance of the AD process.

Mathematical modeling of an AD process is generally recognized as a formidable tool to optimize the operating parameters of AD systems. A number of models could be found in the literature to describe the kinetics of AD, such as first-order kinetics (Ramirez et al., 2009; Santos et al., 2020; Yu et al., 2018) and modified Gompertz models (Panigrahi et al., 2020; Şenol et al., 2020; Sun et al., 2019). Table 3 summarizes the kinetic analyses of the AD process using different models. For instance, the hydrolysis of the AD is often evaluated with a first-order kinetic model (as described by (Equation 7)), by assuming that the hydrolysis rate would not depend on the concentrations of the organic portions in the digestate or hydrolytic biomass.

$$P = P_0 \left(1 - \exp(-k_h t)\right) \qquad \text{(Equation 7)}$$

where P is the amount of biomethane production, P_0 is the cumulative biomethane production potential, k_h is the hydrolysis rate constant, and t is the digestion time. Ramirez et al. (2009) suggested that, in order to account for the slowly degradable materials and complex substrates, the first-order model should be modified. However, due to the diversified properties of organic matter, it is difficult to establish a universal model applicable for all the AD processes.

For a simplified approach, the modified Gompertz model has been extensively used to determine the production capacity and kinetics of an AD reactor in a batch mode. The governing equation of the modified Gompertz model can be described by (Equation 8):

$$P = P_0\, exp\left\{ - exp\left[\frac{\mu_m e}{P_0}(\lambda - t) + 1\right]\right\} \qquad \text{(Equation 8)}$$

where μ_m is the maximal biomethane production rate, e is the Euler's number (i.e., 2.718), and λ is the lag-phase time. Zhao et al. (2018) found that the modified Gompertz model would reasonably delineate the biomethanation reactions involving carbohydrate components rather than protein-predominated wastes.

Among reported AD models, the Anaerobic Digestion Model No. 1 (ADM1), officially launched by the International Water Association in 2002, has been considered as the most capable and complex model. The ADM1 is a structured model that uses differential algebraic equations to describe biochemical and physico-chemical processes (Li et al., 2021a, 2021b). The ADM1 has attracted widespread attention in the field of fundamental research and practical applications of the AD since it could provide theoretical guidance and technical support for the design, operation, and optimization of AD processes. Different variations of the ADM1 have been developed by modifying equations accounting for biochemical reactions in the

Table 3. Modeling of AD processes using first-order kinetics or modified Gompertz models

	Conditions				First-order kinetics (Equation 7)			Modified Gompertz model (Equation 8)				
Types of feedstock in AD	Tempearture (°C)	Duration (day)	ISR (–)[a]	TS (g)	P_0 (mL/g-TS)	k_h (1/d)	R^2	P_0 (mL-CH_4)	μ_m (mL- CH_4/d)	λ (day)	R^2	References
Starch	37	22	2.66	8	496.9 [b]	0.147	0.997	463.8 [b]	48.1 [b]	−0.16	0.993	(Yu et al., 2018)
Cellulose	37	22	2.66	8	422.1 [b]	0.122	0.998	385.8 [b]	33.9 [b]	−0.32	0.992	(Yu et al., 2018)
Protein	37	22	2.66	8	431.3 [b]	0.178	0.996	409.9 [b]	49.6 [b]	−0.18	0.988	(Yu et al., 2018)
Orange bagasse	37	60	12.4	0.4	128.6 ± 1.3 [c]	0.10 ± 0.00	0.96	123.0 ± 0.4 [c]	–	1.7 ± 0.1	0.980	(Santos et al., 2020)
Yard/food waste	-	30	0.40	-	-	-	-	456 [c]	21 [c]	3.14	0.978	(Panigrahi et al., 2020)
OFMSW	37	30	-	-	-	0.14	0.940	395 [c]	31.2 [c]	1.27	0.990	(Shamurad et al., 2020)
Beer lees	35	40	0.33	-	-	-	-	401.8 ± 7.7 [c]	-	5.88	0.990	(Sun et al., 2019)
Beer lees	55	40	0.33	-	-	-	-	456.8 ± 7.7 [c]	-	3.37	0.990	(Sun et al., 2019)
Spend coffee ground	37	50	1	-	-	-	-	317.8 [c]	22.7	9.0	0.993	(Atelge et al., 2021)
Vegetable waste	37	24	-	-	-	-	-	421.1 ± 2.9 [c]	34.1 ± 0.6 [c]	2.92 ± 0.10	0.995	(Zhao et al., 2018)
Manure, corn silage and beet pulp	39	45	-	-	-	-	-	427.4 [b]	11.3 [b]	2.78	0.990	(Şenol et al., 2020)

[a]inoculum-to-substrate ratio (ISR).
[b]mL-CH_4 per g-TS.
[c]mL-CH_4 per g-VS. OFMSW, Organic fraction of municipal solid waste.

model (Bareha et al., 2019; Li et al., 2020; Maharaj et al., 2019). For instance, Bareha et al. (2019) modified the ADM1 to evaluate the fate of organic nitrogen during AD. Similarly, Li et al. (2021a), 2021b simplified the ADM1 to determine the fates of C, N and P elements in an AD process fed with pig manure. Maharaj et al. (2019) also modified the ADM1 to predict the precipitation and dissolution of trace elements in an AD process.

MICROBIAL COMMUNITY ANALYSIS

Microbial communities in an AD reactor which consist of a variety of microbial species are the driving energy to degrade organic matters and produce biogas. However, the interactions within the communities in an AD reactor are still poorly understood due to their complex pathways. In this study, we summarized the analysis on major microbial communities in several given biomethane reactors in the literature, as presented in Table 4. Many syntrophic bacteria belonging to *Firmicutes* have been reported to generate VFAs, such as acetic and butyric acids, via hydrolysis of various organic substrates in food wastes or agricultural wastes. Acetic acid is the primary component for biomethane production through acetoclastic methanogenesis, while butyric acid (C_3H_7COOH) is used by some of the genera from *Firmicutes*. Similarly, some species belonging to *Porphyromonadaceae* families can utilize protein in food waste to generate VFAs, such as acetic, isobutyric, propionic, and isovaleric acids (Kurade et al., 2020). *Porphyromonadaceae* can be involved in the biogas production associated with methanogens. *Syntrophomonas* is one of the commonly observed predominant bacteria (*Syntrophomonadaceae* family) for rapid metabolism of long-chain fatty acids into acetates, which was further converted to methane through acetoclastic methanogens in a syntrophic relationship (Kurade et al., 2020). *Syntrophomonas* was reported to increase to ~15% of the total bacterial community if the contents of fats, oils, and greases in the feeding material were increased to 3% (Ziels et al., 2016). For the archaeal communities in an AD system, the commonly observed phylum should be *Euryarchaeota*, where the genera mainly include acetoclastic and hydrogenotrophic methanogens. As the common acetoclastic methanogens, *Methanosaeta* spp. is usually found to be dominant in a stable mesophilic methanogenic system. The hydrogenotrophic methanogens, such as *Methanolinea*, *Methanospirillum*, *Methanobacterium* and *Methanoculleus*, are also commonly found in an AD system (Zhao et al., 2018). Some species of *Methanomicrobiaceae* families, such as *Methanogenium*, are found in marine methogens and thus are tolerant to high salinity. There is a limited number of studies reporting the relatively high abundance in a high-salinity anaerobic digester.

It is noteworthy that the functions of microorganisms in an AD system cannot be identified without elucidating their metabolic pathways. Zhu et al. (2019) proposed an integrated cellulose catabolic network which was reconstructed according to gene annotations, as shown in Figure 2. They identified a novel model of glucose mineralization without acetate formation, which was asserted in a pair of syntrophs: *Clostridiaceae* sp. and *Methanoculleus thermophilus*. They also highlighted that the synergistic network of anaerobic microbes should be established on the catabolic complementarity and the equilibrium principles of electron transfer and energy conservation.

Zhao et al. (2020) also critically reviewed the detailed microbiology of AD, i.e., direct interspecies electron transfer (DIET), and then proposed strategies for re-engineering practices of digester design. They noted that, with the addition of electrically conductive materials (e.g., graphite-based suspended carrier and carbon fiber), the performance of an AD system could be effectively improved due to the DIET enhancement. However, digesters with these conductive materials should be carefully considered from an economic point of view; they are economically viable only if the conductive materials are permanently installed within the digester.

PERSPECTIVES AND PROSPECTS

To facilitate the progression toward a circular bioeconomy, we identify three important elements regarding the future directions of the AcD using agricultural wastes, i.e., (1) establishment of regional circular centers utilizing agricultural wastes, (2) development of optimization strategies for an AD system, including substrate pretreatment, and system configuration and control, and (3) maximization of economic benefits of digestate utilization. Additionally, an insightful deliberation and prospect are introduced.

Establishment of regional circular center using agricultural wastes

The AcD process has the potential to be used as the core technology to supply energy (biogas) and resource (biofertilizer) if a supply chain is established to provide enough organic wastes. In other words,

Table 4. Major microbial communities present in biogas-producing reactors

AD condition	Function	Families	Taxonomy (phylum)	Metabolic features	Metabolic product[a]	References
Mesophilic	Acetogenesis	*Syntrophomonadaceae*	*Firmicutes*	Some species utilize long-chain fatty acids. Syntrophic association with acetoclastic methanogens.	CH_3COOH	(Kurade et al., 2020; Ziels et al., 2016)
Mesophilic	Acetogenesis	*Syntrophaceae*	*Proteobacteria*	Utilize propionate and butyrate. Some species utilize long chain fatty acids. Syntrophic association with hydrogenotrophic methanogens.	H_2, CO_2, CH_3COOH	(Wang et al., 2019)
Mesophilic	Acetogenesis	*Lactobacillaceae*	*Firmicutes*	Utilize VFAs.	H_2, CO_2, CH_3COOH	(Wang et al., 2019)
Mesophilic	Acetogenesis	*Pseudomonadaceae*	*Proteobacteria*	Utilize organic substances.	H_2, CO_2, CH_3COOH	(Xiong et al., 2020)
Thermophilic	Acetogenesis	*Planococcaceae*	*Firmicutes*	Multi-functions (hydrolysis and fermentation). Some species utilize organic substances, such as cellulose.	VFA	(Li et al., 2020)
Mesophilic	Acidogenesis	*Porphyromonadaceae*	*Bacteroidetes*	Some species can hydrolyze protein into VFA and NH_3. Some species ferment carbohydrates into monosaccharides.	VFA, NH_3	(Kurade et al., 2020; Zhou et al., 2019)
Mesophilic	Acidogenesis	*Anaerolineaceae*	*Chloroflexi*	Ferment glucose	H_2, VFA	(Zhang et al., 2019a)
Mesophilic	Acidogenesis	*Candidatus*	*Cloacimonetes*	Ferment amino acids into H_2. Some species involve in propionate degradation (syntrophic)	H_2	(Zhou et al., 2019)
Thermophilic	Acidogenesis	*Thermotogaceae*	*Thermotogae*	Ferment carbohydrates and peptides	VFA	(Hao and Wang, 2015)
Mesophilic	Acidogenesis	*Porphyromonadaceae*	*Bacteroidetes*	Hydrolyze polysaccharides and proteins; ferment sugars.	VFA	(Hahnke et al., 2015)
Mesophilic	Acidogenesis	*Ruminococcaceae*	*Firmicutes*	Ferment glucose.	H_2, VFA	(Liu et al., 2015)
Mesophilic, thermophilic	Acidogenesis, Acetogenesis	*Clostridiaceae*	*Firmicutes*	Multi-functions (hydrolysis and fermentation). Ferment carbohydrates such as sucrose, glucose, xylose, hemicellulose, cellulose, and starch	VFA, CH_3COOH, H_2, CO_2	(Hao and Wang, 2015; Wang et al., 2019; Zhu et al., 2019)

(*Continued on next page*)

Table 4. *Continued*

AD condition	Function	Families	Taxonomy (phylum)	Metabolic features	Metabolic product[a]	References
Mesophilic	Acidogenesis, Acetogenesis	*Spirochaetaceae*	*Spirochetes*	Acetate oxidation. Some uncultured members have potential for VFA production.	CO_2, H_2	(Deng et al., 2018)
Mesophilic	Methanogenic	*Methanobacteriaceae*	*Euryarchaeota*	Hydrogenotrophic methanogens. Utilize hydrogen and carbon dioxide. More tenacious tolerance.	CH_4	(Yu et al., 2020; Zhao et al., 2018)
Mesophilic	Methanogenic	*Methanosaetaceae*	*Euryarchaeota*	Acetoclastic methanogens. Dominant at stable methanogenic systems.	CH_4	(Zhao et al., 2018)
Mesophilic	Methanogenic	*Methanosarcina*	*Euryarchaeota*	Mixotrophic methanogens. Some species utilize hydrogen and carbon dioxide.	CH_4	(Liu et al., 2020)
Mesophilic	Methanogenic	*Methanoregulaceae, Methanospirillaceae*	*Euryarchaeota*	Hydrogenotrophic methanogens.	CH_4	(Gao et al., 2019)
Mesophilic	Methanogenic	*Methanomicrobiaceae*	*Euryarchaeota*	Hydrogenotrophic methanogens. Tolerant to high salinity.	CH_4	(Gao et al., 2019)

[a]VFA

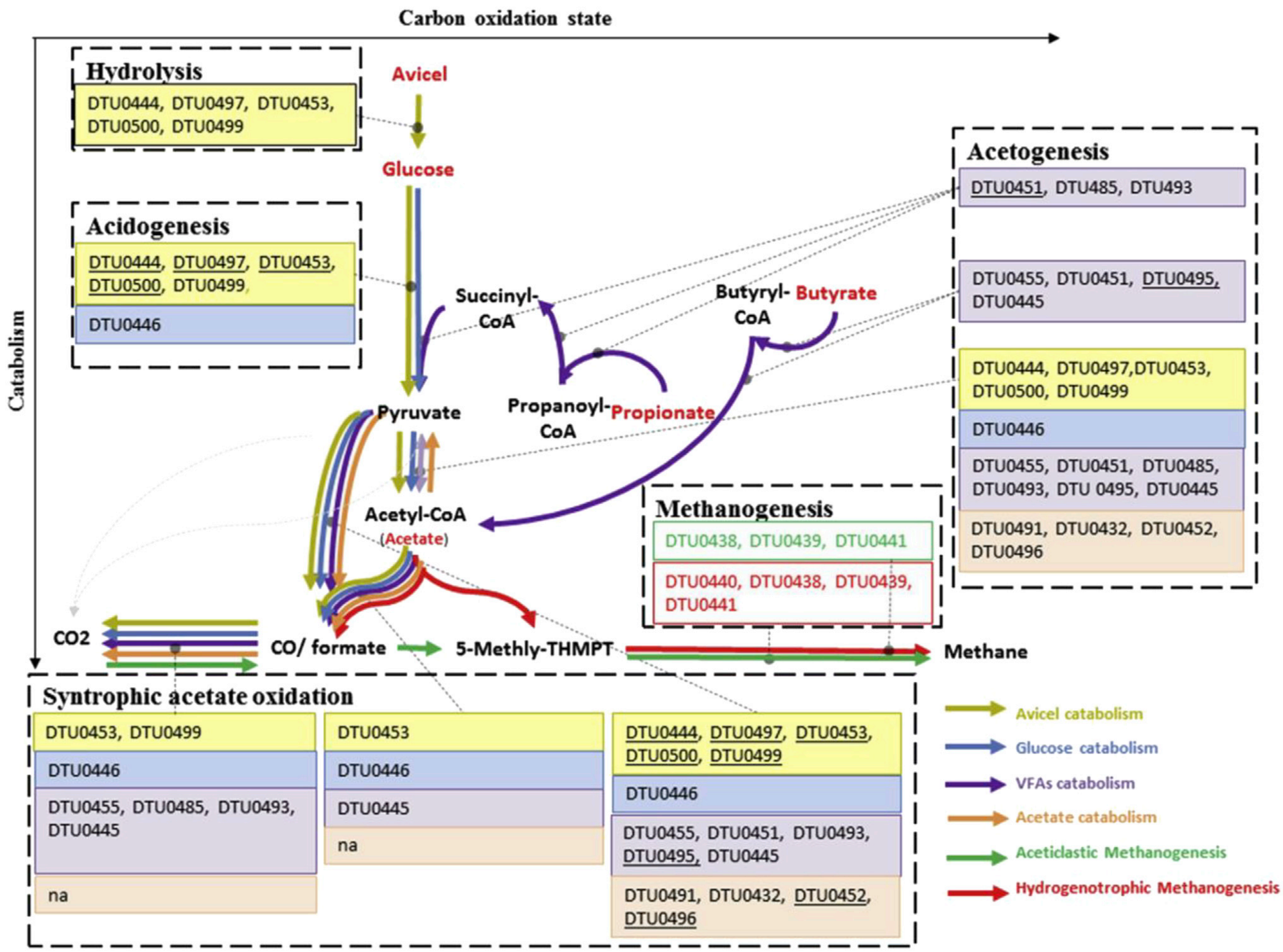

Figure 2. Integrated cellulose catabolic network reconstructed according to gene annotations

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a regional waste-to-energy and -resource supply center (or so-called circular center) based on AcD technologies can be established toward a circular bioeconomy. Figure 3 illustrates the concept of establishing a regional circular center for circular bioeconomy. It is noted that the 5R principles (i.e., reduction, reuse, recycling, recovery, and reclamation) should be taken into account in building the circular supply chains (Pan et al., 2015). Tait et al. (2021) conducted a critical evaluation of Australian agro-industries for establishing sustainable AD systems based on agricultural wastes. As a result of their study, it was suggested that the AcD using agro-industrial organic wastes should be promoted to maximize benefit and minimize cost, and the municipal WWTPs should be included in the design of AcD processes. Depending upon the physicochemical properties of organic wastes that are available in a nearby region, different unit processes for AcD (e.g., single-stage, two-stage or three-stage) could be designed and deployed. As an alternative to conventional organic waste treatments (e.g., landfill or incineration), the AcD process could efficiently convert the organic portions in the waste to alternate energy source, i.e., biogas, while the remaining solids after AcD could be used as biofertilizers or soil amendment agents. The circular center approach can realize the concept of sustainable materials management in the agro-environmental systems.

System optimization: pretreatment, configuration, and control parameters

The design of an AcD system including pretreatment processes should be designed based on the physicochemical properties of feedstock. Tait et al. (2021) showed the importance of formulating feedstock, such as the C/N ratio, in optimizing AcD systems. Appropriate pretreatment could effectively enhance the mixing and decomposition of organics and thus promote the utilization of organic substrates by

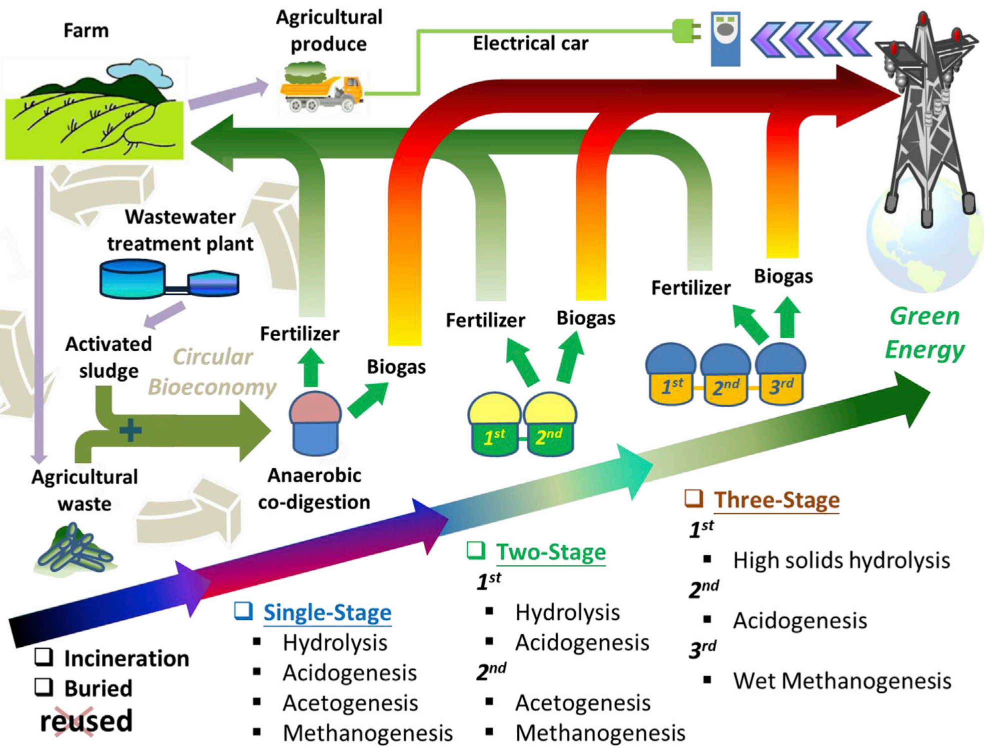

Figure 3. Concept of waste-to-energy and -resource supply center for moving toward circular bioeconomy
Different stages of unit processes for the AcD could be designed and deployed depending on the physico-chemical properties of organic wastes that are available at the nearby region.

microorganisms (Yue et al., 2021). The hydrolysis reaction is typically considered as the rate-limiting step in the AcD process. To effectively increase the biogas production, deployment of proper pretreatment and conditioning processes for bio-feedstock prior to AD is essential. Effective pretreatment of bio-feedstock can also decrease the required HRT for the subsequent AcD process. The performance of pretreatment in promoting the biodegradability of feedstock largely depends on the physico-chemical properties of feedstock, as well as the pretreatment method. In general, milling is considered as the simplest physical pretreatment technology for bio-feedstock such as agricultural residues. The mechanical pretreatment technologies (including chemical pretreatments) can reduce the particle size of bio-feedstock and wastewater sludge, while a chemical pretreatment is to increase the fractions of soluble organics. Especially for food wastes, the pretreatment should be carefully selected since food wastes contain significant quantities of biodegradable substrates that may be easily destructed by a chemical pretreatment. In some cases, pretreatments would produce inhibitory intermediates and thus decrease the biodegradability of food wastes.

An AD process can be designed with different configurations, such as those systems of a single stage or multiple stages. The CH_4 yield of a single-stage digester is usually rather limited as the optimum operating conditions for hydrolysis, acidogenesis, acetogenesis, and methanogenesis are difficult to achieve. It is simply because many control variables, such as metabolic properties, nutrition requirements, and retention time, should be considered simultaneously. The deployment of multi-stage ADs could provide a total solution to the process hurdles associated with a single-stage digester. A number of studies have reported improved biogas yields through the multi-stage configuration (Zhang et al., 2019b). Despite the lower capital cost and fewer technological failures for the single-stage system, the configuration with multiple stages leads to a greater CH_4 yield with higher system stability and capacity of generating H_2 as a byproduct, as shown in Figure 4. It is noted that each step of a multi-stage digester is easily controllable for performance optimization, thereby enhancing the overall biogas yield. The optimal operations for different steps of the multi-stage digester are different to achieve maximum CH_4 production. For instance, Nathao et al. (2013) found that the pH of the first digester of a two-stage AD system should be maintained at 5.5-6.5 for efficient acidogenesis, while the pH of the second digester should be increased to around 7.0 for the maximum

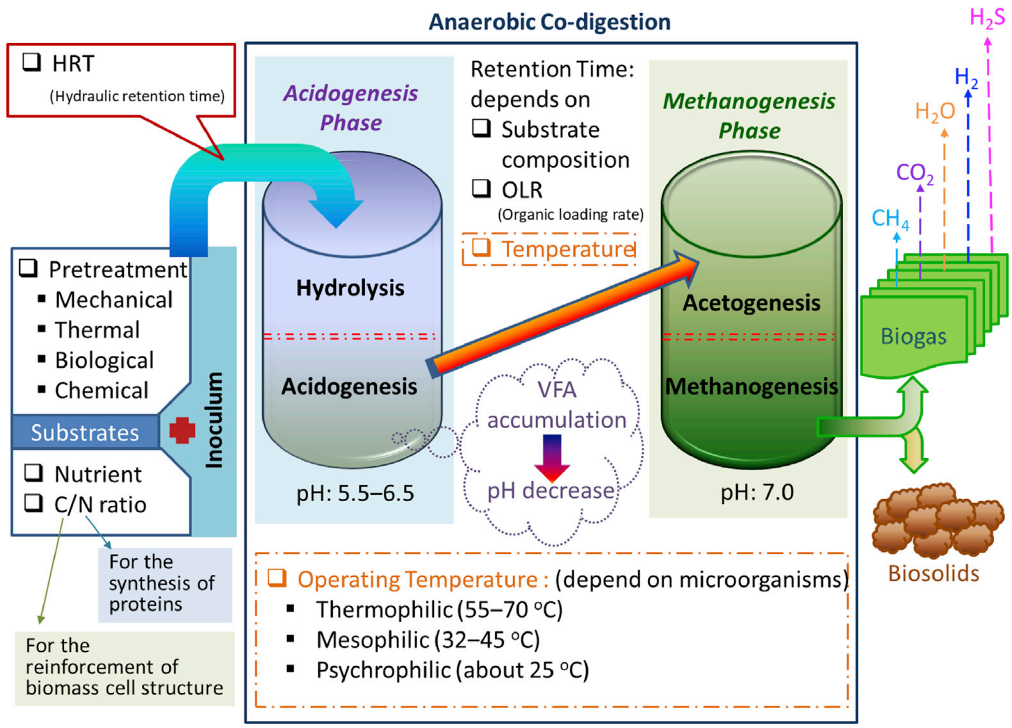

Figure 4. Two-stage configuration of AcD with key operation parameters

In general, the AcD contains four steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis.

methanogenesis. Aside from the pH control, a number of essential factors, such as digestion temperature, should be considered as well.

Maximization of economic benefits of digestate utilization

Digestate management with green solutions has gained increasing popularity in the field of AD. The AcD digestate is a mixture of fibrous solids and liquid portions. The utilization of digested materials could provide a unique opportunity of recovering value-added resources (e.g., humus, organic acids, and nutrients) to augment the economic benefits of the AcD, in addition to biomethane production. This perspective has been recently addressed by Lü et al. (2021) that several selective separation processes were critically reviewed to harvest value-added products from the liquid digestate. Meanwhile, with selective separation as a side unit of AD, the performance of AD processes will be more stable as the inhibitory compounds (such as organic acids and ammonia) are continuously removed from the digestate. During the AcD process, acidogenic bacteria would produce various types of organic acids (e.g., sulfuric acid, and acetic acid), and the accumulation of VFAs would further inhibit the AcD process as mentioned above. One of the practical solutions to overcome the VFAs accumulation is to apply side-treatment processes, such as electrodeionization (Datta et al., 2013), for simultaneous separation of organic acids. The side-treatment process also can serve as a conditioning process for the hydrolysis stage and organic waste pretreatment. The separated acids, e.g., sulfuric acid, can be recycled and then used in pretreating lignocellulosic substances. With a higher purity, the separated organic acids could be further used as bio-chemicals to replace fossil-based chemicals, thereby realizing a circular bioeconomy. Bhatt et al. (2020) conducted a comprehensive analysis on the current development of wet AD systems and found that producing short-chain carboxylic acids (C2-C4) provided an economically viable alternative to conventional biomethane production. In particular, the theoretical energy yields of these acids equal and even exceed biogas, making this move toward carbon mitigation goals (Bhatt et al., 2020). Li et al. (2021a), (2021b) also indicated that both biogas and nutrients in digestate (as organic fertilizers) would largely determine the cost effectiveness of an AD process.

The solid portion in the digestate could be converted to biochar by pyrolysis, which could be further utilized as soil amendment agents or solid fuels. Biochar has gained considerable attention since it provides multiple functions, such as nutrients management and regulation of natural carbon cycle in the soil system.

As for the soil amendment, a number of studies have shown that biochar could improve soil fertility while enhancing crop growth and yields (Azeem et al., 2019). Biochar also can increase the capacity of CO_2 mineralization by soils while increasing the retention of nitrogen and phosphorus in soils, thereby decreasing non-point source pollution. Despite its environmental benefits, the use of biochar as a soil amendment agent has been in its infancy. Still, limited knowledge is available regarding the fate and transport of nutrient species in biochar in the soil environment. The knowledge gaps and research needs should be identified for advancing biochar in future environmentally sustainable applications.

Prospects for circular bioeconomy

As we are heading into a new era that highly values resource circulation and sustainability, the concept of waste management has been transformed from "proper ultimate disposal" to "zero waste by total recycling". To ensure an everlasting terrestrial ecosystem, the SDGs proposed by the UN clearly stipulate that efforts should be expended on the promotion of affordable and clean energy. In line with the proposition of the UN, the European Union (EU) announced the European Green Deal, in which the "Clean Energy" and "Eliminating Pollution" would be the two proposed strategies to facilitate the efficient use of resources by moving to a clean and sustainable circular economy. The use of clean energy instead of fossil-driven power generation is a strategic option to achieve environmental sustainability. While the UN and EU advocate the necessary actions to achieve a circular economy, responsible production and consumption become another popular concept to ensure the environmental sustainability.

The present study systematically reviewed the requirements of using an AcD process for agricultural waste treatment, including the composition of the feedstock, pretreatment, operating parameters, kinetics/modeling, and microbial population and abundance. Anaerobic co-digestion may be regarded as the total solution to agricultural waste and WAS treatment by generating methane, liquid fertilizers, and organic-rich digestate that can be used for power generation, crops growing and soil amendment, respectively. Also, the possible separation of organic acids from digestate may add the industrial attractiveness and economical value to the application of the AcD for organic waste treatment. By recognizing the benefits and disadvantages of using AcD from various perspectives, the agricultural organic waste can be properly treated, and the valuable contents in the organic waste can be recovered or regenerated. In other words, potential environmental impacts can be eliminated through deploying AcD of agricultural wastes and WAS, thereby realizing the sustainable production and consumption. Additionally, three possible devotions of the establishment of the regional circular center, system optimization of the AcD process, and maximization of economic benefits of digestate utilization were proposed in this study to practice the suggested strategies.

Anyhow, efficient resource utilization is regarded as the keystone of the realization of a circular economy. The required capital expenses for land acquisition, facilities construction, and system operation & management should also be considered when putting the concept of sustainable development into practice. Other supplementary measures such as the collection of the feedstock, logistics for recovered resources for transdisciplinary applications, and pollution reduction and prevention are important to enable the projected circular economy to be realized. Lastly, the degree of social acceptance should not be ignored since public participation could be a significant support to facilitate the progress toward the sustainable circular bioeconomy.

CONCLUSIONS

As a part of the prospect of bioeconomy, the AcD of agricultural wastes (including food waste) with WAS is an effective approach to creating bioenergy and producing biochemicals in a sustainable manner. In this paper, we critically reviewed the significance of the factors related to feedstock (organic wastes) and important control parameters affecting the performance of AcD processes. The improvement of biomass processing (pretreatment) and conditioning technologies would effectively increase the efficiency of organics conversion and the subsequent bio-gas production. We analyzed the operating conditions and key performance indicators for the AcD reported in the recent literature and found that the average C/N ratio of feed substrates at the condition of the maximum methane production would be 18.0 ± 6.9 ($p < 0.05$; $n = 7$). The average methane production from the AcD of food wastes with other organics, such as agricultural wastes and WAS, was found to be 421 ± 45 L/kg-VS ($p < 0.05$; $n = 9$). Lastly, we highlighted perspectives and prospects on future priority research directions, such as (i) establishment of the regional circular center utilizing agricultural wastes, (ii) development of optimization strategies for an AD system, including substrate pretreatment, and system configuration and control, and (iii) maximization of economic benefits of digestate utilization. We also proposed an insightful prospect for the role of the AcD in the area of waste recycling and resource circulation, thereby realizing a circular bioeconomy.

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AUTHOR CONTRIBUTIONS

Conceptualization, S.-Y.P., H.K and C.F.; Methodology, C.-Y.T., C.-W.L., and S.-W.W.; Investigation, S.-Y.P., C.-Y.T., and H.K.; Writing – Original Draft, S.-Y.P. and C.-Y.T.; Writing – Review & Editing, S.-Y.P., H.K. and C.F.; Funding Acquisition, S.-Y.P. and C.-W.L.; Resources, C.-W.L., S.-W.W., and C.F.; Supervision, H.K and C.F.

DECLARATION OF INTERESTS

The authors declare no competing interests.

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