A review of anaerobic digestion systems for biodegradable waste: Configurations, operating parameters, and current trends

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ABSTRACT

With benefits to the human health, environment, economy, and energy, anaerobic digestion (AD) systems have attracted remarkable attention within the scientific community. Anaerobic digestion system is created from (bio)reactors to perform a series of bi-metabolism steps including hydrolysis/acidogenesis, acetogenesis, and methanogenesis. By considering the physical separation of the digestion steps above, AD systems can be classified into single-stage (all digestion steps in one reactor) and multi-stage (digestion steps in various reactors). Operation of the AD systems does not only depend on the type of digestion system but also relies on the interaction among growth factors (temperature, pH, and nutrients), the type of reactor, and operating parameters (retention time, organic loading rate). However, these interactions were often reviewed inadequately for the single-stage digestion systems. Therefore, this paper aims to provide a comprehensive review of both single-stage and multi-stage systems as well as the influence of the growth factors, operating conditions, and the type of reactor on them. From those points, the advantages, disadvantages, and application range of each system are well understood.

Keywords: Anaerobic digester, Anaerobic digestion systems, Single-stage, Solid waste treatment, Three-stage, Two-stage

1. Introduction

Solid waste generation is an inevitable consequence of human activities and its rapid increase in recent years has caused significant problems that humankind has to deal with. In 2010, nearly 1.3 billion metric tons of municipal solid waste (MSW) were produced worldwide, and the annual generation is estimated to increase up to 2.2 billion metric tons by 2025 [1, 2]. The organic fraction of MSW (OFMSW) was often reported around 50-60% of total MSW, which had been cast off in landfills for many years [2-5]. According to Hoornweg and Bhada-Tata [1], an annual amount of 250.05 million tons MSW was dumped into landfills in high-income countries; that of low-income and middle-income countries were 2.67 and 157.1 million tons, respectively. Besides OFMSW, major sources of biodegradable waste also originate from agriculture (animal manures, energy crops, algal biomass, harvest remains), food industry (food/beverage processing, dairy, starch,

sugar, pulp/paper, etc.), and sewage sludge [6, 7]. Landfilling biodegradable waste leads to various issues that threaten the environment and public health [1, 8]. Also, the fact that the world population continues to grow together with the progress of human civilization leads to the increase of global energy demand. The traditional energy sources such as fossil fuel are exhausting, and nuclear energy is a potential risk to the environment and human health [9]. Thus, the increasing energy demand is becoming a global challenge, and this has promoted the interest in search of alternative energy sources. Meanwhile, biodegradable waste can be converted to methane gas (renewable energy source) by using anaerobic digestion (AD) systems. Therefore, with benefits to the human health, environment, economy, and energy conservation, the AD systems have attracted remarkable attention within the scientific community [10, 11].

The AD systems are constituted from reactors to perform a series of bi-metabolism steps including hydrolysis/acidogenesis,

acetogenesis, and methanogenesis [11-13]. By considering the physical separation of the digestion steps in various reactors, the AD digestion systems include single-stage, two-stage, and three-stage. In which, reactors are the place where growth factors (temperature (T), pH, nutrients) and operating parameters (retention time (RT) and organic loading rate (OLR)) are controlled to allow one or several digestion steps occur [14, 15]. The growth factors affect the living conditions of anaerobes and determine the success or failure of the reactors [11]. Meanwhile, the RT determines the contact time between microorganism and substrate so that it is long enough to complete the transformation. A too long RT will lead to such a big reactor that will increase investment and operating costs. The OLR determines a measure of the amount of daily organic matter treated by a certain volume of the reactor. Both RT and OLR rely on the growth factors and the type of reactor. So the performance of the AD systems is determined by the complex relationships among the growth factors, the operating parameters, system type, type of reactor, and also coordination of reactors in the system. These relationships were often reviewed inadequately in the single-stage digestion system [6, 11, 12]. Meanwhile, for the multi-stage digestion systems, the information is still limited. Demirel and Yenigün [16] are two of few authors who did a review on the operation of the two-stage digestion system. However, they only investigated one configuration of the continuous stirred tank reactor (CSTR) for hydrolysis/acidogenesis and the upflow anaerobic sludge blanket (UASB) reactor for the methanogenesis. Moreover, the three-stage digestion system has limited information. So far there have been no studies providing a systematic and full review of the AD systems, including the coordination between the functional reactors in a system as well as the operation of each reactor.

This paper aims to provide a comprehensive review of the AD systems for biodegradable waste including the single-stage, two-stage, and three-stage systems. Each type of them is discussed about classification, configuration, and operation. Moreover, advantages, disadvantages, and application ranges for each system are also evaluated. The factors influencing the AD systems are identified and discussed based on the existing literature. Therefore, this study is as a guideline for the AD system design. This paper is organized into five following sections. Section 1 is an introduction. Section 2 presents mechanism of AD processes and anaerobic reactors which are responsible for the AD process. In section 3, the operation of the AD systems is systematized according to the biological phase separation, and the newest achievements are reviewed within the literature. Section 4 presents the current applications. Finally, section 5 is conclusions and recommendations.
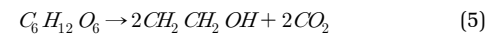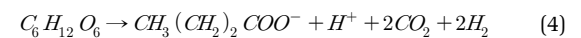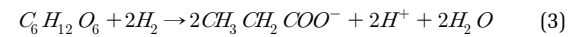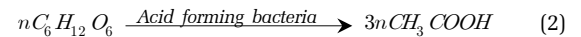
2. Basics of Anaerobic Digestion

2.1. Biochemical Mechanism of Biogas Production

The anaerobic digestion is a series of bio-metabolism steps including hydrolysis, acidogenesis, acetogenesis, and methanogenesis, respectively [11-13, 17-20] (see Fig. S1). In which, every digestion step has different growth characteristics as presented below.
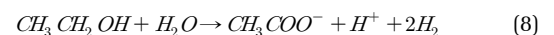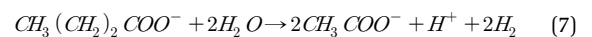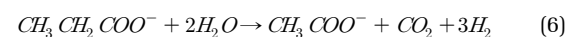
The first step (hydrolysis, also called solubilisation) breaks down high molecular weight constituents (e.g., lipids, carbohydrate, and protein) into smaller soluble organic matter (e.g., fatty acid, glucose, and amino acid) by exo-enzymes, and is represented by reaction (1) [15, 21]. The hydrolytic microorganism has ability to strongly resist the environmental fluctuations and the toxins which may be present in the feedstock [22]. They can work in a wide range of pH (4-11) [23]. However, pH-values in the range of 6-8 were often reported to provide an optimum working condition for hydrolysis [3, 24, 25]. The hydrolysis of lignocellulosic materials is a relatively slow process. Thus, hydrolysis is considered as a rate-limiting process in the digestion of lignocellulose [13, 18, 26]. Fortunately, this issue can be accelerated significantly by using pre-treatment processes including physical, chemical, and biological methods. The details of these methods are presented by Ariunbaatar et al. [27].

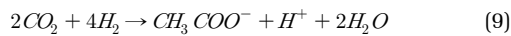$$(C_6 H_{10} O_5)_n + n H_2 O \xrightarrow{\text{Hydrolysis}} n C_6 H_{12} O_6 \qquad (1)$$
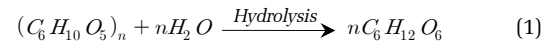
The second step (acidogenesis) transforms the products of the hydrolytic process into volatile fatty acids (VFAs) such as propionic acid, butyric acid, acetic acid and ethanol by the action of the acidogenic bacteria [28-30]. They have characteristics of strong and fast growth with a minimum doubling time of 30 min [31]. Acidogenesis can be described by Eq. (2)-(5) [15, 18, 21, 26]. The pH-conditions significantly influence VFA products. By stepwise shifting pH from 4 to 8, the main products changed from butyric and acetic acids to acetic and propionic acids [32, 33]. Moreover, the VFAs formation was strongly inhibited with pH below 4.0 [34]. The pH in the range of 5.5-6.5 was often reported as the optimal range [11].

$$n C_6 H_{12} O_6 \xrightarrow{\text{Acid forming bacteria}} 3n CH_3 COOH \qquad (2)$$

$$C_6 H_{12} O_6 + 2H_2 \rightarrow 2CH_3 CH_2 COO^- + 2H^+ + 2H_2 O \qquad (3)$$

$$C_6 H_{12} O_6 \rightarrow CH_3 (CH_2)_2 COO^- + H^+ + 2CO_2 + 2H_2 \qquad (4)$$

$$C_6 H_{12} O_6 \rightarrow 2CH_2 CH_2 OH + 2CO_2 \qquad (5)$$

The third step (acetogenesis) transforms most products of acidogenesis into acetic acid (CH_3COOH), hydrogen (H_2), and carbon dioxide (CO_2) as shown in Eq. (6)-(9) [14, 15, 18, 35]. The growth kinetic of acetogenesis is slower than which of acidogenesis, with a minimal doubling time in the range of 1.5-4 d [31]. Acetogens are strict anaerobes, the present of oxidants like oxygen or nitrate is toxic [26], and they work better in acid-weak environment (pH from 6.0 to 6.2) [31]. Remarkably, high partial pressure of hydrogen product ($\geq 10^4$ atm) inhibits Eq. (6) and (7), hence hydrogen product should be released [34, 36].

$$CH_3 CH_2 COO^- + 2H_2 O \rightarrow CH_3 COO^- + CO_2 + 3H_2 \qquad (6)$$

$$CH_3 (CH_2)_2 COO^- + 2H_2 O \rightarrow 2CH_3 COO^- + H^+ + 2H_2 \qquad (7)$$

$$CH_3 CH_2 OH + H_2 O \rightarrow CH_3 COO^- + H^+ + 2H_2 \qquad (8)$$

$$2CO_2 + 4H_2 \rightarrow CH_3COO^- + H^+ + 2H_2O \qquad (9)$$

The fourth step (methanogenesis) plays the most important role in generating methane gas by methanogens. There are two basic mechanisms for methane generation including acetoclastic and hydrogenotrophic methanogenesis. The first way, acetotrophic bacteria ferment acetic acid to CH_4 and CO_2 as Eq. (10) [26, 37]. The second one, hydrogenotrophic methanogens use CO_2 and H_2 as a food source, as given in Eq. (11) [20, 26, 37]. While the minimum doubling time of hydrogenotrophic bacteria is in the range of 4-12 h, acetotrophic bacteria have a much lower maximum growth rate with doubling times of 2-3 d [31, 37]. Generally, methanogens are extremely sensitive to pH condition, the presence of oxygen, and other factors such as free ammonia (FAN), H_2S, and VFAs [15]. They cannot work at pH condition lower than 6.2, even die in the pH condition under 6.0 [15, 18], and can be inhibited by releasing free ammonia (FAN) when pH-value is over > 7.8 [31]. The optimal pH-value was often reported at neutral environment (7.0-7.2) [11, 18, 38, 39]. Methanogenic process lost stable stage when FAN concentration reached 0.6-0.69 g/L [40, 41], reduced 50% performance when FAN concentration was 1.45 g/L [42], and failed by FAN concentration of 1.7-1.8 g/L [43]. The optimum concentrations of sulfur for the growth of methanogens were reported in the literature to vary from 16 to 160 mg-S/L [44]. The methanogenesis was often stable with a low VFA concentration (< 200 mg/L), and inhibition occurred when acetate concentration exceeded 3 g/L [36]. The oscillation of temperature in the reactor should be as small as possible, that is, < 1°C/d for thermophiles and 2-3°C/d for mesophiles [15].

$$CH_3COOH \xrightarrow{\text{acetoclastic bacteria}} CH_4 + CO_2 \qquad (10)$$

$$CO_2 + 4H_2 \xrightarrow{\text{hydrogenotrophic bacteria}} CH_4 + 2H_2O \qquad (11)$$
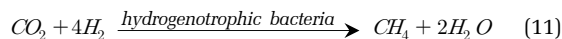
2.2. Anaerobic Reactors

Anaerobic reactor (digester) is considered as the heart of digestion systems, which encourages anaerobic microorganisms to thrive inside for responding to the digestion steps. While the success of the digester depends very much on pH condition as described in section 2.1, its digestion rate is significantly dependent on temperature condition. Temperature is not only one of the most important in the selection of microbial group inside reactors, but it also influences the state of substrates such as the solubility, metabolic rate, and ionization equilibria. According to temperature conditions, there are three microorganism groups including psychrophiles, mesophiles and thermophiles, respectively living in the temperature range of 4-15°C, 20-40°C and 45-70°C [45]. The influence of temperature on the growth rate of each group is shown in Fig. S2. In general, the increase in temperature has benefits of enhancing reaction rates significantly [17]. However, the mesophiles are less sensitive to environmental changes than the thermophiles, hence operation reactor at thermophilic temperature requires higher technic than mesophilic condition [14, 46].

The anaerobic digesters can be categorized in several different ways, but the main classification can be set between wet and dry types relied on total solid (TS) contents because solid concentration has a great impact on the cost, performance, and technique of the AD process [47, 48]. Categorization of anaerobic reactors is summarized in Fig. 1(a). The dry digesters have been designed to serve the feedstock having TS ≥ 20%, hence solid concentration within the reactor is also high (TS ≥ 15%) [3, 47, 49]. In the dry digester, the substrate particles play the roles of nutrient source and also supporting media. Especially, the microorganisms not only attach to the surface but also penetrate through substrate particles [50]. The dry type can be classified into three groups including the horizontal plug-flow, vertical plug-flow, and non-flow (batch type). The brief descriptions of these digesters are shown in Table S1, and the discussion about them is presented in the next section. The wet digesters are defined to serve the feedstock having TS ≤ 15% and well known in the wastewater treatment field [5, 51]. They can often be classified based on the growth of bacteria inside including suspended growth and attached growth [52]. In the suspended growth digester, the microorganisms are maintained in suspension within the liquid. Meanwhile, they attach and grow on the surface of supporting media in the attached growth reactor. Detail descriptions of their representatives are shown in Table S2.

The design and operation of the anaerobic reactor are characterized by two parameters including RT and OLR. In which, the RT is defined as the average time that the substrate maintained in a reactor and quantified by the equation: RT = reactor volume/daily flow. The RT has to be long enough to ensure the completion of one or several digestion steps that the reactor is in charge. For the wet reactor, the substrate materials are often separated into liquid and solid materials thus the RT is divided into hydraulic RT (HRT) and solid RT (SRT). Meanwhile, the OLR gives a measure of the amount of daily organic matter treated by a certain volume of the reactor and calculated indirectly via RT by equation OLR = Organic-concentration/RT. Both RT and OLR depend on the process parameters that influence the anaerobic bacterial growth such as temperature, pH, and waste characteristics.

3. Anaerobic Digestion Systems

The simplest anaerobic digestion system contains one reactor which is responsible for doing all four digestion steps. It is called the single-stage digestion system. However, along with the development of science and biotechnology, scientists have found that each digestion step has different optimal thriving conditions [3, 11, 31]. Therefore, the idea of the physical separation of the digestion steps (multi-stage digestion) has been given the aim to optimize each of them to reach the highest performance of transformation. Currently, the multi-stage digestion systems only include two-stage and three-stage systems. The two-stage system performs hydrolysis and methanogenesis in two different reactors. More complex, the three-stage system performs the hydrolysis, acidogenesis/acetogenesis, and methanogenesis in various reactors. This paper classifies the subcategories of the AD systems based on type (wet or dry) of reactor employed as shown in Fig. 1(b).
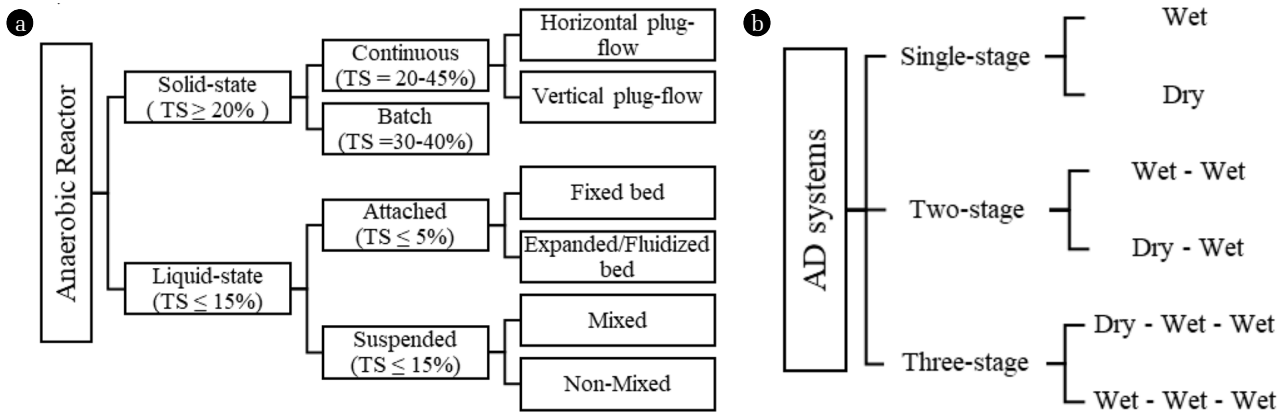
Fig. 1. a) Classification of digesters; b) classification of AD systems.

3.1. The Single-Stage Systems

In principle, the environmental conditions are not necessarily optimal for any digestion step, but they satisfy all four steps [18]. However as shown in section 2.1, the range of growth conditions for the methanogen is covered by the other process steps. Moreover, methanogens are the most sensitive to the environment and also have the slowest growth among microorganism consortium. Therefore, the environmental conditions in the single-stage digestion systems should be optimized for methanogenesis. Commonly, the single-stage system operates with some requirements: Carbon to nitrogen (C/N) of the substrate in the range of 15-30 [6, 11, 13]; pH ranging between 6.8 and 7.4 [6, 11]; HRT 30 d at mesophilic temperature, 20 d at thermophilic, and 50 d at psychrophilic.

3.1.1. The wet single-stage systems

They are the single-stage systems employing the wet reactor, and they can be classified into two groups including low-rate (OLR = 0.5-1.6 kg-VS/m^3.d^{-1}) and high-rate (OLR = 1.6-4.8 kg-VS/m^3.d^{-1}) [31]. A number of studies have shown that they are sensitive to inhibitor such as FAN. According to the study of Duan et al. [41], FAN level > 0.6 mg/L was the key factor influencing system stability. Nakakubo et al. [42] showed that methane yield was reduced 50% when FAN concentration was up to 1.45 g/L. And Yen and Brune [53] reported that the system could fail when FAN increased within to the range of 1.7-1.8 g/L.

3.1.1.1. The wet low-rate system

The wet low-rate system is the oldest and simplest type, has its key characteristics such as long RT (30-60 d), poor mixing process, and non-heating process [39, 54]. It is a very simple operation, but very low performance; hence the low-rate systems have only been used within household-scale and farm-scale in developing countries such as China, India, and Vietnam [21]. The diagram principle of these systems is shown in Table S2(a). It doesn't matter if it is small or large-scale, there is always a stratification inside reactors with four zones: (i) a scum layer, (ii) a liquid layer (or supernatant), (iii) a layer of digesting solids, and (iv) a layer of digested solids [17, 39]. Among these systems, the household-scale one (Table S2(b)) is also known as floating-dome digester or fixed dome digester. Although it has capacity in the range

of 1-8 m^3, the 1-3 m^3 one is the most commonly used. In this system, the daily feedstock is about 1/40-1/50 of the reactor volume [21]. The farm-scale one (lagoon digester or anaerobic pond, see Table S2(c) is operated with an HRT in the range of 30-60 d, SRT of 50-100 d, and TS of the feedstock of 0.5 to 5% [52].

3.1.1.2. The wet high-rate systems

The wet high-rate systems have been developed from the low-rate digestion for improving performance. The feedstock can be heated and mixed to make a uniform environment, which leads to the fact that the reactor is less volume, more stable and more efficient [54]. The first generation of the wet high-rate systems uses the wet plug flow (WPF) reactor (see Table S2(d)), which is supported by a heat source. This system has been applied the most for manure with solid content in the range of 11-14% and HRT of 15-20 d [21, 55]. A more complicated reactor called completely mixed reactor (CMR) or continuous stirred tank reactor (CSTR) requires internal mixing activities, see Table S2(e). This system often operates at mesophilic temperature with OLR in the range of 1.5-5 kg-VS/m^3.d^{-1} and RT from 15-20 d [21]. In the CSTR, the bacteria get washed away together with the effluent out of reactor [21]. Meanwhile, to reach higher OLR, the reactor need a higher concentration of bacteria maintained inside. Therefore, the biomass of the effluent is separated in a settling tank and pumped back to the reactor where the biomass concentration is maintained in the range of 5-10% VS [21, 34, 36]. It is called anaerobic contact (AC) system as shown in Table S2(f) Waasa, a famous brand name of AC system, is used for treating OFMSW. In Waasa system, fresh materials are mixed with effluent water to attain 10-15% TS. The OLR could reach 4-8 kg-VS/m^3.d^{-1} with the efficiency of 100-150 m^3-biogas/ton-waste and TS reduction of 50-60% [5].

When the substrate of the feedstock is mainly in the soluble state (or TS < 3-5%), the wet reactors with high biomass concentrations such as upflow anaerobic sludge blanket (UASB), expanded granular sludge bed (ESGB), expanded bed (EB), fluidized bed (FB), internal circulation (IC), and anaerobic fixed bed (AFB) can be employed. The principles and application range of these reactors are presented in Table. S2. They are well known in the industrial wastewater treatment sector. In the United States, application allocation of anaerobic technologies in the industrial wastewater treatment

for period 2002-2007 included 34% UASB, 33% IC, 22% EGSB, 1% AFB, and 10% others [37].

3.1.2. The dry single-stage systems

The high solid waste can be treated by the wet digestion systems, but it needs to be added a large amount of water. Unfortunately, dilution of the waste stream not only demands higher costs of water and energy consumption but also requires a reactor with higher volume. Moreover, dilution of waste can drag on reducing biogas yield and make more wastewater [47]. Therefore, the dry digesters have been developed to deal with high solid waste (see Table. S1). The main issues with the operation of dry systems compared to which of wet systems are the mixing and pumping of the substrate with high viscosity [51]. Along with the evolution of technologies, these issues gradually reduced. Nowadays Dranco, Valorga, Kompogas, and Biocell are the most commonly applied systems of pilot scale plans in Europe [5, 51]. Generally, the dry systems are stronger than the wet ones because of stability even with ammonia concentration up to 2.5-3 g/L and VFAs in the range of 23-24 g/L [3, 56].

The Dranco system is such a vertical plug-flow digester (see Table. S1(a)). In this system, a part of the digestate is taken back to the mixing pump where it is blended with fresh materials (separated sources, particle size < 4 cm) for inoculation with a ratio of 6-8:1. After that, the mixture is introduced into the top of the reactor and moves downward to the conical bottom where a screw conveyor removes the digestate. The rest of digestate is dewatered before being composted. There is no active mixing within the reactor, hence 20-30% of biogas can be lost because of the incomplete digestion [3]. The system has been proved successful to treat solid wastes with TS ranging from 20 to 50% [47]. When OFMSW (TS of 30-45%) is processed in the mesophilic condition with HRT 20 d, VS decreases 55% and 5-8 kg-VS/m^3.d^{-1} OLR is produced [3, 7]. In the thermophilic temperature with RT 14 d, the amount of OLR could be up to 15 kg-VS/m^3.d^{-1} and 65% of VS could be destroyed [5]. For digestion of OFMSW, alkali additive is often unnecessary, and pH inside the reactor is nearly 8. Each ton of waste can produce 120-170 m^3-biogas (equivalent to 200 kWh) and the plant consumes 30-40% of it and exports the rest [3].

The Kompogas system employs a horizontal plug-flow digester with internal rotors to assist in degassing and homogenizing waste (see Table. S1(b)). The incoming waste stream (particle size < 50 mm) is mixed with the liquid of the digestate to attain TS in the range of 23-28% for the system to flow properly. At higher TS values, the mixture is hard to flow because of too high viscosity while lower TS values lead to accumulating sand and glass inside the reactor [5, 47]. The system often operates in the thermophilic condition (55-60°C) with HRT of 14-20 d. The digestate is dewatered by using a screw press then the solid part is composted by aeration within 2-3 weeks [3, 5]. For one ton of waste, the system can produce 130-150 m^3-biogas, 500 kg-compost, and 300 L liquid fertilizer.

The Valorga system uses a vertical plug-flow reactor (see Table. S1(a)). The feedstock is the mixture of the incoming waste stream (size < 30 mm) and the digestate with TS adjusted in the range of 25-30%. If TS concentration is less than 20%, the grit particles will quickly settle down to the bottom and clog the recirculated-gas pores [5]. The system produces about 140 m^3-biogas/ton-waste with VS removal rate of 50% [7]. For the substrate of OFMSW, the pH inside is often in the range of 7.8-8, hence alkali additive is not required. When pH drops, the feeding pump should stop until process regulates itself [3]. A part of the biogas product is injected back at the bottom of the reactor with high pressure (5 bar) for mixing purpose and keeping materials suspended [3, 21]. The system is often operated in the mesophilic temperature with RT of 18-23 d and produces biogas yield of 220-270 L/kg-VS. After digestion, the digestate is composted within 2-3 weeks [5].

The dry-batch digestion system (see Table S1(c)): This system originates to mimic the landfill process. However, unlike natural biodegradation in the landfill, the reaction in the dry-batch system is accelerated by two basic factors. The first one is the continuous recirculation of the leachate. It permits formation of a high humidity environment and diffuseness of the inoculum and the substrates. The second one is that temperature in the systems is controlled at the optimum range. This resulted in many fold higher biogas production rates and lower RT than observed in landfills [49]. The digester is loaded with fresh materials (TS of 30-40%), discharged, and then filled with a new batch. The digestion time finishes when biogas production ceases. After more than 20 d digesting at the mesophilic temperature, the digestate is aerated for 1-3 weeks for composting [5, 51]. For each ton of waste, the system produces 90 kg of biogas (58% CH$_4$), 455 kg wastewater, and 310 kg of compost [39]. Despite saving 40% equipment cost compared to continuous-dry systems, the batch-dry systems have much lower applied ratio because they need much more space for construction [5].

3.1.3. Assessments

The single-stage digestion systems have a wide application range and can cover the most type of biodegradable waste. The evaluation of the systems based on some technical targets is shown in Table 1; it also includes their advantages and disadvantages. In the single-stage digestion systems, the strong group bacteria (facultative microorganisms) can easily repulse the weak groups (methanogens) when living in the same reactor. Thus, the fluctuations of the load, pH, and solid concentration of the feedstock could harm the stability of the system. If the rate of acid formation is more than the rate of methane formation, it means there is an accumulation of VFAs, the system must be stopped and waited for turning stable status. Therefore, the two-stage system can be considered as an optimal solution to deal with the mentioned issues above.

3.2. The Two-Stage Systems

3.2.1. Configuration of two-stage systems

The growth characteristics of the hydrolysis/acidogenesis and the methanogenesis are very different. Therefore, the idea for two-stage digestion is given to optimizing every digestion step. In the present, there are two different viewpoints about separating digestion-steps into two reactors. The first perspective supposes that the first tank allows the hydrolysis/acidogenesis to occur, and the second tank optimizes the acetogenesis/methanogenesis [49, 76]. Meanwhile, the second perspective proposes that the acetogenesis

Table 1. Comparison between Wet and Dry Single-State Digestion Systems [5, 21, 55].

Criteria	Wet				Dry	
	Low rate	High rate contact process	High rate complete mixed	High rate Others	Continuous	Batch
TS (%)	0.5-5	< 5-10	10-15	< 3-5	20-35	30-40
HRT/SRT (d)	30-60/50-100	>15/30	30	< 20/30-50	14-20	20
OLR	0.5-1.6 (kg-VS.d^{-1}/m^3)	1.6-4 (kg-VS.d^{-1}/m^3)	4-8 (kg-VS.d^{-1}/m^3)	Up to 50 kg-COD d^{-1}/m^3	5-15 (kg-VS.d^{-1}/m^3)	< 10 (kg-VS.d^{-1}/m^3)
Solids characteristics	Fine	Fine	Fine	Fine	Coarse (4-5 cm)	Coarse (4-5cm)
Capacity range (tons/y)	1-5	-	1,000-230,000	Up to 8,760,000	3,000-270,000	3,000-150,000
Heating	None	√	√	Sometime yes	√	√
Mixing	None	√	√	None	None	None
Level of technology	Low	High	High	High	High	Medium
Water demand	Very high	High	High	High	None	Low
Construction cost	Low	Very high	Very high	High	Medium	High
Maintenance cost	Very low	High	High	High	High	Low
Equipment invest	Low	High	High	High	Very high	Medium
Advantages	The simplest operation; The cheapest investment cost;	Inspired by the wastewater treatment field; easily pumping and piping;	It can be used to treat the high solid contents, but the water needs to be added;	Very high OLR; High efficiency;	High OLR; the simplest pre-treatment; low water usage, less heat requirement, and smaller reactors.	Less pre-sorting and treatment; low cost for equipment invest and operation.
Disadvantages	Very low biogas production rate; only be applied on a small scale.	The complicated pre-treatment; need a settling tank; high energy cost for heating, mixing and recycling concentrated sludge;	Incomplete degradation; sensitive to the environmental changes; need a good pre-treatment;	The substrate is almost in the soluble state; Need complex pre-treatment process.	Need an inoculation loop; cannot be applied for low solid materials; the incomplete degradation.	Recirculation of leachate; Highly variability of gas production; cannot be applied for low solid materials; the incomplete degradation; too large reactor.
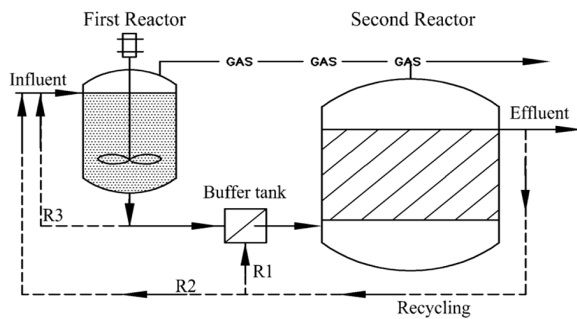
Fig. 2. The configuration of the two-stage digestion system.

should be in the first reactor and the second reactor only optimizes methanogenesis [18, 54]. The second point of view might like to maximize hydrogen product from acetogenesis. However, there are several reasons why this idea is not reasonable. Firstly, hydrogen production from AD is currently not economically viable because of the high cost required to enrich the hydrogen gas to meet the commercial quality standards [38]. Secondly, as described in section 2.1, methanogens (hydrogen consumer) and acetogens (hydrogen producer) should work in close cooperation. Thirdly, acetogens are also sensitive and strict anaerobes; they should not work together with facultative bacteria (hydrolytic/acidogenic microorganism). In fact, the true separation of the digestion-steps is very difficult to achieve [21], hence the acetogenesis can occur in both reactors but mostly in the second one. Diagram of two-stage systems is shown in Fig. 2.

Because of the different environmental conditions between two reactors, a buffer tank is often set between two reactors as shown in Fig. 2 for many purposes such as removing non-hydrolysable materials, controlling pH, and even controlling the organic concentration [38, 77]. The two-stage system can use one, two, or all three water recirculation loops (R1, R2, and R3, shown in Fig. 2) in case of need. Using these recirculation loops brings many advantages, such as further controlling of pH (reduction of acidity due to using the high alkalinity effluent from the second reactor); mixing/diluting of the high solid feedstock; and improving activities of bacteria [38, 76]. However, if the inhibitors are accumulated inside reactors and cause the unstable condition of the system due to long time being used, the incoming substrate should be diluted by fresh water. After hydrolysis, the solid content of feedstock decreases significantly. Therefore, the two-stage system may be a dry-wet configuration (high solid in the first digester and low solid in the second digester) or wet-wet configuration (both stages are low solid) [5, 49]. The hydrolysis/acidogenesis usually employs a CSTR [37]. Meanwhile, the methanogenesis uses the wet high-rate reactors including CSTR, UASB, AFB, FB, EB, IC, and EGSB [14, 36].

3.2.2. The hydrolytic/acidogenic reactor

The mission of this reactor is to convert the substrate to the VFAs

as fast as possible while avoiding being inhibited and overloaded. Therefore, a series of operating conditions including pH, temperature, acid concentration, nutrients, and substrate concentration (TS) must be controlled. For the high-solid waste such as OFMSW, the waste stream should be shredded to reduce particle size (< 15 mm) before being fed to the reactor [3]. The hydrolysis/acetogenesis generally proceeds sufficiently fast in the mixing tanks, hence no further reactor has been developed for the hydrolysis/acetogenesis [37]. Among mixing tanks, the CSTR is used the most, the wet plug-flow reactor sometimes [3, 49].

Total solids: Hydrolyzing the high-solids feedstock allows the reactor to operate larger capacity, requires less energy for heating and less water consumption. However, a too high solid content can cause high viscosity of the mixtures, which leads to the fact that insufficient mixing or mixing can be too energy-consuming [18, 78, 79]. Moreover, the increase of solid content (in the range of 5-40%) causes the increase of inhibitors and insoluble solids content, which leads to the decrease of the hydrolytic conversion rate [79]. Also, a high solid concentration (TS > 15%) of feedstock requires much longer RT (10-15 d) than usual [18]. Thus, TS of 15-20% in the feedstock is often considered as the upper limit for hydrolysis/acidogenesis when using the mixed reactor [78].

pH value: Moestedt et al. [80] reported that the pH below 4.5 led to requiring HRT up to 15 d in the hydrolytic/acidogenic reactor. Yu and Fang [81] lifted up pH from 4 to 5.5, which resulted solubility and acidification of substrates increased significantly [81]. Whereas under alkaline conditions, Zhang et al. [25] reported that VFAs formation was significantly decreased when pH increased from 7 to 11. Deublein and Steinhauser [35] showed that pH > 10 caused an irreversible loss of the activity of the microorganisms. The pH values between 5.5 and 6.5 were often reported as an optimal range and the best pH values was 6.0 [54, 82-84]. These results are reasonable for each condition of hydrolysis and acidogenesis as presented in section 2.1.

Temperature: Most studies have agreed that the hydrolytic/acidogenic rate is proportional to the increase of temperature [23, 85, 86]. In addition, compared to the mesophilic conditions, the thermophilic regimes have been increased destruction of pathogens which might have a severe impact on reactor and environment [87]. Therefore, thermophilic temperature seems better than mesophilic one. However, Kozuchowska and Evison [88] reported that the mesophilic operations provided a more stable condition than the thermophilic operation in acidification of coffee waste. Komemoto et al. [89] even notified that the solubilization rate of food waste was significantly higher in mesophilic conditions than others. Thus, regarding energy and efficiency, the mesophilic temperature in the range of 35-37°C is still preferred to use for the hydrolysis/acidogenesis of organic waste, see Table 3.

Retention time: RT of this reactor depends on not only the environmental conditions but also waste characteristics. For hydrolysis/acidogenesis of the source-sorted OFMSW (TS 8.2%), Pavan et al. [90] recommended that RT would be safe in the range of 2-3 d at the mesophilic temperature. At the same temperature above but for food waste, Paudel et al. [91] optimized RTs and reported that one d was the best. Meanwhile, for agro-industrial wastewater, Dareioti and Kornaros [92] even got the highest efficiency of acidogenesis achieved at lower RT (0.75 d). In general,

Table 2. Illustrative Examples of the Applications of the Single-Stage Anaerobic Digestion

Type of waste	Reactor	Operation mode	TS (%)	T (℃)	pH	HRT(day)	OLR	Biogas yield	Reduction(VS %)	References
OFMSW	CSTR	Continuous	1.75	55	7.94	25	1.35 g-VS.d⁻¹/L	404 mL-CH$_4$/g-VS	68.4	[57]
OFMSW	CSTR	Batch	11.0	30	7.0-7.4	19	-	314 mL-Biogas/g-VS	41.8	[58]
FW	CSTR	Continuous	2.57	35	7.8	60	2.4	448 mL-CH$_4$/g-VS	74.1	[59]
FW	CSTR	Semi-continuous	6.8	35	6.8-7.3	4.6	9 g-VS.d⁻¹/L	540 mL-Biogas/g-VS	-	[60]
FW	CSTR	Continuous	7.2	37	7-8	16	9.2 g-VS.d⁻¹/L	455 mL-Biogas/g-VS	92.2	[61]
FW and fresh leachate	CSTR	Continuous	8.28	37	7.2-7.8	10	8.3 g-VS.d⁻¹/L	506 mL-Biogas/g-VS	71.6	[4]
FW Rice husk, and cow dung	CSTR	Batch	23	37	7.7	45	5 g-VS.d⁻¹/L	584 mL-Biogas/g-VS	78.0	[62]
FW and LW	Horizontal plug-flow reactor	Continuous	40	30-40	--	40	7.5 g-VS.d⁻¹/L	250 mL-Biogas/g-VS	80.0	[63]
FW and Piggery waste	CSTR	Continuous	17.22	37	7.4	20	6.35 g-COD.d⁻¹/L	396 mL-Biogas/g-VS	75.6	[64]
FW and WAS	CSTR	Sequencing batch	24.6	35	7.3	13	4.27 g-VS.d⁻¹/L	346 mL-Biogas/g-VS	67.4	[65]
Sewage Sludge	CSTR	Continuous	4.0-5.2	35	-	15	2.1-2.5 g-VS.d⁻¹/L	260-306 mL-CH$_4$/g-VS	40.9	[66]
Sewage Sludge	CSTR	Continuous	2.5	35	7.7	20	1.43 g-VS.d⁻¹/L	451 mL-CH$_4$/g-VS	43.5	[67]
Vegetable and FW	CSTR	Batch	3.0	37	6.7-7.2	72	-	393.7 NmL- CH$_4$/g-VS	-	[68]
Sewage	UASB	Continuous	0.035 0.043	18-25	7.0	23-27 h	1.4-1.6 g-VS.d⁻¹/L	249-439 mL-CH$_4$/g-COD	62-70	[69]
Stillage	CSTR	Batch	3.44	37	7.2-7.6	28	1.1 g-VS.d⁻¹/L	260 mL-CH$_4$/g-COD	80.0	[70]
Cattle Manure	CSTR	Batch	5.6	55	7.3-7.5	15	2.98 g-VS.d⁻¹/L	236 mL-CH$_4$/g-VS	42.0	[71]
Horse dung	CSTR	Batch	2.5	37	--	45	0.45 g-VS.d⁻¹/L	291 mL-Biogas/g-VS	-	[72]
Grass	CSTR	Continuous	2.3	35	7.0	20	2.5 g-VS.d⁻¹/L	310 mL-CH$_4$/g-VS	63.9	[73]
FW and dairy manure	CSTR	Semi-continuous	14.6	36	7-8	54	3 g-VS.d⁻¹/L	510 mL-Biogas/g-VS	66.6-69.6	[74]
Sludge and vegetable waste	CSTR	Batch	2.5	37	--	45	0.56 g-VS.d⁻¹/L	504 mL-Biogas/g-VS	-	[75]

Table 3. Illustrative Examples of the Applications of the Two-Stage Anaerobic Digestion

Type of waste	1st Reactor / 2nd Reactor	Operation mode	TS (%)	T1 (°C) / T2 (°C)	pH1 / pH2	HRT1 / HRT2	OLR	Biogas yield	Reduction	References
MSW	CSTR/ CSTR	Semi-continuous/ Continuous	3.9/ 1.9	37 ± 1/ 37 ± 1	5.5-6.0/ -----	5.5 d/ 15 d	9 g-VS.d⁻¹/L / 2.5 g-VS.d⁻¹/L	710 ml-biogas/g-VS	-	Li et al. [97]
MSW	CSTR/ CSTR	Semi-continuous/ Continuous	8.2/ 6.7	34.6/ 54.9	---/ ---	4.6 d/ 7.7 d	16.4 g-VS.d⁻¹/L / 7.4 g-VS.d⁻¹/L	560 ml-biogas/g-TVS	74.1% VS	Pavan et al. [90]
Swine waste and biowaste	CSTR/ CSTR	Semi-continuous/ Semi-continuous	3.9/ 3.4	55.0/ 55.9	5.52/ 7.61	3 d/ 22 d	11.2 g-VS.d⁻¹/L / 1.16 g-VS.d⁻¹/L	402 mL-CH₄/gVS	60.7% VS	Schievano et al. [57]
FW and wastewater	CSTR/ CSTR	Continuous/ Continuous	5.7/ 4.8	37 ± 1/ 37 ± 1	5.0-5.5/ 7.0-7.5	8 h/ 20 d	106 g-VS.d⁻¹/L / 1.24 g-VS.d⁻¹/L	728 mL-CH₄/g-VS	74.5% VS	Paudel et al. [91]
FW	CSTR/ AB	Semi-continuous/ Semi-continuous	11.7/ 10.8	55.0/ 35.0	5.5/ 7.3-7.7	1.3 d/ 5.0 d	38.4 g-VS.d⁻¹/L / 6.6 g-VS.d⁻¹/L	464 mL-CH₄/g-VS / 283 ml-H2/g-VS	57.6% VS	Chu et al. [82]
FW	CSTR/ CSTR	Continuous/ Continuous	3.57/ 3.06	55.0/ 35.0	5.36/ 7.59	3 d/ 12 d	14.2 g-VS.d⁻¹/L / 2.6 g-VS.d⁻¹/L	440 mL-CH₄/g-VS	80.1% VS	Wu et al. [59]
FW	CSTR/ CSTR	Continuous/ Continuous	7.91/ 7.53	55.0/ 35.0	3.61/ 7.32	6 d/ 24 d	12.0 g-VS.d⁻¹/L / 3.1 g-VS.d⁻¹/L	450 mL-CH₄/g-VS	71.6% VS	Wu et al. [59]
Kitchen waste	CSTR/ UASB	Batch/ Continuous	12.9/ 12.5	35-37/ 35-37	7.0/ 7.0	3.1 d/ 1 d	16 g-VS.d⁻¹/L / 20 g-COD.d⁻¹/L	520 mL-CH₄/g-TS	86,9% COD	Zhang et al. [25]
Pelletized grass	CSTR/ CSTR	Semi-continuous/ Semi-continuous	5.5/ 4.8	35.0/ 35.0	5.5/ 7.0	18 h/ 11.25 d	66.6 g-VS.d⁻¹/L / 4.4 g-VS.d⁻¹/L	305.3 mL-CH₄/g-VS	62.2% VS	Massanet-Nicolau et al. [73]
Pelletized grass	CSTR/ CSTR	Semi-continuous/ Semi-continuous	5.5/ 4.8	35.0/ 35.0	5.5/ 7.0	18 h/ 19.25 d	66.6 g-VS.d⁻¹/L / 2.6 g-VS.d⁻¹/L	394.4 mL-CH₄/g-VS	67.2% VS	Massanet-Nicolau et al. [73]
Stillage	CSTR/ CSTR	Batch/ Batch	3.69/ 3.53	37.0/ 37.0	5.5/ 7.18	8 h/ 16.7-17.8	---	360-380 mL-CH₄/g-COD	80% VS	Nasr et al. [70]
Cattle manure	CSTR/ CSTR	Semi-continuous/ Semi-continuous	5.48/ 4.48	68.0/ 55.0	7.3/ 7.5	3 d/ 12 d	14.9 g-VS.d⁻¹/L / 3.725 g-VS.d⁻¹/L	260 mL-CH₄/g-VS	47.1-2.08% VS	Nielsen et al. [71]
Sewage	UASB/ UASB	Continuous/ Continuous	0.038/ 0.030	26.0/ 26.0	6.88/	8-10 h/ 5-6 h	3.6-5.0 g-VS.d⁻¹/L / 2.9-4.6 g-VS.d⁻¹/	261 mL-CH₄/g-COD	65% VS	Halalsheh et al. [69]
Starfish	CSTR/ UASB	Batch/ Continuous	3.93/ 1.60	35.0/ 35.0	7.4/ 7.5	2 d/ 1 d	5.0 g-COD.d⁻¹/L / 6.0 g-COD.d⁻¹/L	296 mL-CH₄/g-COD	44% VS	Kim et al. [77]
Sludge	CSTR/ CSTR	Semi-continuous/ Semi-continuous	7.8/ 6.7	55.0/ 55.0	5.40/ 8.25	3.3 d/ 12.6 d	16 g-VS.d⁻¹/L / 4 g-VS.d⁻¹/L	640 mL-CH₄/g-VS	-	Cavinato et al. [99]
Sludge	CSTR/ CSTR	Semi-continuous/ Semi-continuous	16.8/ 13.8	55.0/ 55.0	4.3/ 7.6	6.6 d/ 12.6 d	21 g-VS.d⁻¹/L / 10 g-VS.d⁻¹/L	580 mL-CH₄/g-VS	-	Cavinato et al. [99]
Sludge	CSTR/ CSTR	Continuous/ Continuous	4.6/ 3.5	35/ 35	7.7/ 8.0	5 d/ 10 d	6.4-7.6 g-VS.d⁻¹/L / 3.2-3.8 g-VS.d⁻¹/L	302-360 mL-Gas/g-VS	53.2% VS	Salsali et al. [66]
WAS	CSTR/ CSTR	Batch/ Continuous		37/ 41	---/	6 d/ 12 d	4.3 g-VS.d⁻¹/L / 2.17 g-VS.d⁻¹/L	290 mL-CH₄/g-VS	32.3% VS	Park et al. [100]

RT in the range of 1 to 3 d is preferred to use to deal with high-solid waste in the mesophilic condition [49, 54]. In case the reactor is operated in batch mode, RT should be maintained within 7-12 d [38].

Inhibitions: Despite strong and resilient characteristics the hydrolytic/acidogenic microorganism can still be inhibited by such high concentrations of organic acids (VFAs) and free ammonia. It has been reported that VFA up to 30 g-COD/L did not inhibit the hydrolysis process at neutral pH (6-7) [93], but VFA levels of 40-50 g-COD/L at low pHs (5.0-5.5) caused stopping the hydrolysis [94]. Koster and Lettinga [95] reported that acidogenesis were severely affected by ammonia concentration in the range of 4,051-5,734 mg NH₃-N/L.

Nutrients: Deublein and Steinhauser [35] are two of the few authors who mentioned about nutrient demands for hydrolysis/acidogenesis and noted that C/N ratio should be in the range of 10-40 while trace elements are not a special requirement.

3.2.3. The methanogenic/acetogenesis reactor
This phase often employs wet high-rate reactors such as CSTR, UASB, AFB, FB, EB, IC, and EGSB [14, 36]. These reactors have to be maintained with the methanogen-rich anaerobic environment which is obligate anaerobic and sensitive to the variation of temperature, pH, and also RT [11, 15]. Therefore, operating conditions in the second reactor have to be complied strictly.

Solid concentrations: TS of the substrate coming to the methane reactor depends on the type of reactor employed. In case of using CSTR or CMR, TS of the substrate is allowed up to 10%. Meanwhile, AFB reactor requires lower solid content of the influent (TS ≤ 5%) [21, 55]. UASB, EB, FB, EGSB, and IC reactors even require lower solid content of the influent (TS ≤ 3%) with biomass concentration inside be maintained in the range of 3.5-4% [21, 36].

The pH value: As shown in Section 2.1, the optimal range of pH condition for methanogens and acetogens are very close to each other. Moreover, acetogens are stronger than methanogens. Therefore, optimizing pH condition for methanogens does not affect the growth of acetogens. In fact, pH in the range of 7.0-8.0 was often used (see Table 3). Moreover, this phase is an alkalinization process (converting acids to biogas), hence pH of the substrate to this reactor should be lower than the range above. Remarkably, when pH inside reactor drops below 6.5, the process should be stopped to adjust pH value.

Temperature: The optimal temperature for methanogenic bacteria under the mesophilic and thermophilic conditions are 35-37°C and 55°C, respectively [15, 96]. Li et al. [97] and Pavan et al. [90] performed the digestion of MSW, used the same condition in hydrolysis but the different temperatures in methanogenesis. While at a temperature of 37°C, Li et al. [97] had to operate the reactor with HRT 15 d to get 710 mL-CH₄/g-VS, at a higher temperature (55°C) Pavan et al. [90] only needed to operate the reactor with HRT 12.5 d to get the same methane yield. In general, the biogas production rates under the thermophilic condition are sharply higher than those under mesophilic condition [15, 46]. Therefore, using thermophilic condition leads to increasing OLR and saving the construction cost significantly. However, thermophilic methanogens are more sensitive to environmental change than mesophilic anaerobes [15]. Thus, thermophilic reactor requires higher

technology.

RT, OLR: Because of using the wet reactor then the term RT includes SRT and HRT. The SRT values greater than 20 d are needed for effective performance at the mesophilic condition, and it should be 7-15 d at the thermophilic temperature [36]. Meanwhile, HRT is shorter than SRT, varies from several h to nearly 20 d depending on the hydrolytic products, is often coupled with temperature and OLR values (see Table 3). Turovskiy and Mathai [54] recommended that HRT should be about 10 d at the mesophilic temperature for sludge treatment. Also at mesophilic temperature, Paudel et al. [91] optimized the methanogenic reactor with HRT 20 d, ORL of 1.24 g VS/L/d for co-digestion of FW and brown water. Rincón et al. [98] announced that 17 d was the best HRT with OLR of 9.2 g-COD/L/d when processing olive mill solid waste. To deal with MSW at mesophilic temperature, Li et al. [97] reported that OLR up to 3.8 kg-VS/m³.d⁻¹ with HRT of 15 d resulted in biogas yield of 540 mL/g-VS. Meanwhile, also for MSW but at the thermophilic condition, Pavan et al. [90] needed a shorter HRT (7.7 d) to get higher OLR (5.7 kg-VS/m³.d⁻¹) with the same biogas yield. Also, Pavan et al. [90] recommended that HRT should be in the range of 8-9 d to treat OFMSW at the thermophilic condition.

Nutrients and Inhibitions: It was often reported that the optimal C:N:P ratio in the range of 75-150:5:1 [36]. However, Lissens et al. [47] showed that methanogenic reactor run well with C/N below 10. Methanogenic inhibition occurred when acetate concentration exceeded 3 g/L, even though there was sufficient alkalinity to maintain pH above 7.0 [36]. The optimum concentrations of sulfur for the growth of methanogens were reported in the literature to vary from 16 to 160 mg-S/L [44]. Total ammonia of 3-7 g/L may be tolerated in this reactor, but threshold values of FAN toxicity were reported to be in the range of 100-250 mg/L [36].

3.2.4. Assessments
The idea of the two-stage digestion not only optimizes each of hydrolysis/acidogenesis and methanogenesis to accelerate overall digestion progress but also brings many other advantages. The methanogens grow in their optimal environment, which leads to the fact that it is more difficult for them to be invaded by other groups compared to the single-stage digestion systems. Therefore, the two-stage digestion system is much more robust than the single-stage one and can run well with the fluctuation of the waste stream. Moreover, biogas produced from the hydrolysis/acidogenesis is not economical, can be removed out of the gas collection system easily without any more process. Thus, by this way, the costs of methane enrichment could be saved significantly. The recent results in applying the two-stage digestion systems are shown in Table 3. Most studies indicated that the two-stage system was better than the single-stage one because of shorter RTs or higher values of VS destruction [59, 66, 70, 71] (more details see Table 2 and Table 3). However, with the low solid contents in the feedstock (TS < 3%), using the two-stage system does not bring more efficient than the single-stage system. For examples, Schievano et al. [57] used both single-stage and two-stage digestion systems to treat OFMSW at low TS condition (1.75-3.4%) with the same conditions (HRT of 25 d, temperature of 55°C) and reported that the methane yield and VS destruction were observed almost the same in both

systems. Halalsheh et al. [69] who experimented digestion of sewage with SS in the range of 350-430 mg/L, also confirmed that the two-stage digestion system was not better than the single-stage.

3.3. The Three-Stage Systems

3.3.1. Configuration of three-stage systems

The second reactor of the two-stage digestion systems is responsible for both steps of acetogenesis and methanogenesis. Unfortunately, acetogens and methanogens have substantially different physiological properties and nutrient requirements. The growth balance between them can be lost by the environmental changes leading to severe inhibition [101]. Therefore, the idea of three-stage digestion has come out to separate the hydrolysis/acidogenesis, acetogenesis, and methanogenesis to various reactors [46, 101, 102].

However, as the reasons discussed in the previous section, the separation of both processes is not a good idea. From another viewpoint, Zhang et al. [103] and Stewart [22] proposed the three-stage digestion model with sequential steps: incomplete hydrolysis, hydrolysis/acidogenesis, and acetogenesis/methanogenesis. This proposal would increase the hydrolytic rate of feedstock in the first reactor significantly. Therefore, the second proposal is more valuable. In fact, due to non-homogeneous metabolism in reality then the distribution of digestion steps in three-stage digestion is, respectively hydrolysis/acidogenesis, acidogenesis/acetogenesis, and acetogenesis/methanogenesis. Hydrogen can be obtained in the first and second reactors, however, as being discussed in the previous section, the collection of hydrogen is not economical. Diagram of the three-stage digestion system is shown in Fig. 3.

Also, like the two-stage systems, the waste stream coming to the three-stage system has to undergo a mechanical pretreatment (particle size < 15 mm), and water is sometimes added to the feedstock to adjust TS (max. 15%). CSTR is still often used in the hydrolysis reactor. After hydrolysis, the non-hydrolysable materials are removed in the first buffer tank. Here, pH and VFAs concentration of the substrate can also be corrected for the next step. The second reactor operates such an anaerobic suspended growth process. Therefore, the second reactor is often a CSTR or UASB reactor. The effluent of the second reactor coming to the third one has to pass through a buffer tank as shown in Fig. 3 for purposes of controlling pH, and concentration of acetic acid. Selections of the type of reactor, feeding mode, and temperature

Fig. 3. Diagram of the three-stage anaerobic digestion system.

phase for each digestion step make a complex matrix configuration of the three-stage digestion systems (see Table S3).

3.3.2. Operations

The first reactor: It is optimized for hydrolysis accompany with mechanical size reduction of the incoming feedstock. Biogas produced in this phase is mostly CO_2. This reactor allows the prevailing acidic conditions to increase the rate of particle disruption and hydrolysis [25, 29]. Thus, the buffering agent is not often required in this reactor [22]. However, sometimes pH dropping below four leads to inhibiting hydrolysis immediately [80]. In fact, whether or not using a buffering agent depends on characteristics of the waste and the ways to operate the system. High temperature accelerates solubilization of the feedstock. He et al. [85] performed hydrolysis of food waste and reported that RT was three day to maximize COD-soluble concentration (46.76 g/L) at 70ºC, but it took a longer time (5 d) at 55ºC (43.33 g/L) and 35ºC (36.25 g/L). However, mesophilic temperature (30-35℃) is still often used along with RT of 2-3 d. Operation of this reactor could be referenced to the hydrolytic/acidogenic reactor discussed in the previous section. Also, the inhibitors in the feedstock can be retained in this reactor caused by absorption into sediment, and thus they reduce adverse impact on the next reactors [22].

The second reactor: It is optimized for acidogenesis and acetogenesis. Biogas produced in this phase mainly includes CO_2 and H_2 [22]. As shown in section 2.1, acidogenesis and acetogenesis thrive optimally in the pH condition in the range of 5.5-6.5 and 6.0-6.2, respectively. Therefore, the best condition for the second reactor is around pH 6.0. The mesophilic temperature (35-37ºC) is preferred to use in these processes with HRT of 2 d to deal with OFMSW (see Table 4). The completion of the second stage is marked by the conversion of almost VFAs into acetic acid.

The third reactor: The operation of this reactor is similar to the second reactor of two-stage digestion systems, such as optimum pH between 7.0 and 7.2 [18].

3.3.3. Assessments

As reported by Salsali et al. [66], three-stage digestion system had a higher performance of VS reduction and biogas yield compared to the two-stage system in the digestion of waste activated sludge. In fact, using three-stage digestion systems brings many disadvantages such as the high cost of investment, operations, and maintenance. Therefore, it is difficult to find out the application of the three-stage anaerobic digestion system for pilot scale. Pile [104] gave little information about the Inland Empire Regional Water Recycling Plant I, California, US. Before 2001, the plant had been operated as a single-stage system but not efficiently. Therefore, since 2001, the plant has been operated in separated-phases including the first reactor with HRT 2.5-3.5 d at 32-40℃; the second reactor with HRT 18-20 d at 56-58℃; and the third reactor with HRT 13-17 d at 42-48℃. VS reduction has been improved from approximately 55% to 60-65%. However, from Tables 3 and 4, the three-stage systems did not produce more methane yield than the two-stage systems. The three-stage digestion is obviously a good idea for optimizing all digestion steps in principle, but it has not been proved persuasively to be better than the two-stage digestion system even in the lab scale.
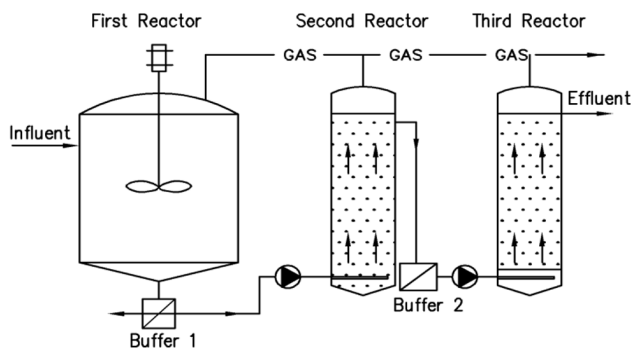
Table 4. Illustrative Examples of the Applications of the Three-Stage Anaerobic Digestion

Type of waste	1st Reactor / 2nd Reactor / 3rd Reactor	Operation mode	TS%/ VS%	T1(°C)/ T2(°C)/ T3(°C)	pH1/ pH2/ PH3	HRT1/ HRT2/ HRT2 (day)	OLR1/ OLR2/ OLR3 (g-VS/L.d)	Biogas yield	Reduction	References
FW and HM	CSTR/ CSTR/ CSTR	Semi-continuous/ Semi-continuous/ Semi-continuous	29.25/ 25.1	35.0/ 35.0/ 35.0	7.3/ 7.7/ 7.9	2.0/ 2.0/ -	12.5 (for all system)	370 mL-CH$_4$/g-VS	71% VS	[105]
FW	CSTR/ CSTR/ CSTR	Batch/ Batch/ Semi-continuous	1.18/ 0.77	35.0/ 35.0/ 35.0	5.1/ 6.0/ 7.3	2.0/ 2.0/ -	10 (for all system)	307 mL-Biogas/g-VS	83.5% VS	[103]
FW	CSTR/ UASB/ UASB	Semi-continuous/ Semi-continuous/ Semi-continuous	12.38/ 11.05	45.0/ 35.0/ 50.0	3.7-3.8/ 3.3-3.4/ 7.2-7.9	2.0/ 2.0/ 12	103.5 8.7/ 63.4/ 52.8	282 mL-CH$_4$/g-COD	90.6% COD	[102]
WAS	CSTR/ CSTR/ CSTR	Batch/ Semi-continuous/ Semi-continuous	--/ 2.6	30.0/ 37.0/ 41.0	6.7/ --/ --	3.0/ 6.0/ 12	8.7/ 4.3/ 2.17	430 mL-CH$_4$/g-VS	75% VS	[100]
WAS	CSTR/ CSTR/ CSTR	Semi-continuous/ Semi-continuous/ Semi-continuous	4.6/ 3.5	42.0/ 35.0/ 35.0	8.0/ 8.0/ 8.0	5.0/ 5.0/ 5.0	2.13-2.53 (for all system)	559-664 mL-Gas/g-VS	60.2% VS	[66]
WAS	CSTR/ CSTR/ CSTR	Semi-continuous/ Semi-continuous/ Semi-continuous	4.6/ 3.5	35.0/ 35.0/ 35.0	8.0/ 8.0/ 8.0	5.0/ 5.0/ 5.0	2.13-2.53 (for all system)	559-664 mL-Gas/g-VS	55.6% VS	[66]
FW	CSTR/ UASB/ UASB	Batch/ Semi-continuous/ Semi-continuous	17.53/ 16.0	30.0/ 35.0/ 41.0	5.0-5.5/ 5.0-5.5/ 7.6-7.9	2.0/ 2.0/ 12	22-22.8/ 25-27.4/ 12-18.8	650-700 mL-CH$_4$/g-VS	95.96% COD	[101]

4. Current Application

The achievements of the AD not only solve the waste problems but also bring a new approach to energy production. For two recent decades, the total installed AD capacity has been increased five-fold worldwide from 2 million tons to 11 million tons a year [106]. Group of European countries has contributed the most proportion to the total biogas power generation with the increase of big biogas plants from 18 in 2001 to 418 in 2015 [10, 107]. In which, Germany has been a leader in using energy from biogas with a power capacity of 12,500 GWh by 2014 [10]. Beside European countries, the United States also contributes a significant proportion in using bio-waste for power generation with 181 AD facilities processing OFMSW (total capacity of 780,000 tons/y, 2015) [108]. In contrast, although Japan is also a developed country with high potential of technologies, the application of biogas technologies is still slow: from one large biogas by 2004 (150 Nm^3-gas/h) up to six plants by 2009 (total 2,400 Nm^3-gas/h) [107]. Meanwhile, for developing countries, low capacity digestion systems are commonly used that China is a typical example with the largest biogas program in the world. By the end of 2009, there had been over 30.5 million household biogas digesters (12.4 billion m^3-gas/y) and 25,012 large-scale biogas projects (7,225 million m^3-gas/y) [10, 109]. Another smaller program, SNV (a Netherland Development Organization) supported the domestic biogas programs for 17 countries in Asia and Africa. By the end of 2012, SNV installed total 479,609 household digesters for Asian countries and 24,990 small digesters for African countries [110]. The biogas program in Nepal and Vietnam counted for over 80% of this total. In general, because of lacking technologies and the financial burden then the wet low rate digesters are commonly used in the developing countries with a large quantity but small-capacity (mainly household scale and farm scale). Meanwhile, the European countries with technical potential have advantage conditions to build large-scale plants which have much higher power capacity than China and other developing countries [5, 111], also see Fig. S3. So based on current installed capacity, the contribution of AD systems in the European can represent a panorama in applying AD systems over the world. Fig. 4 shows the current application of various AD systems in Europe by 2014.
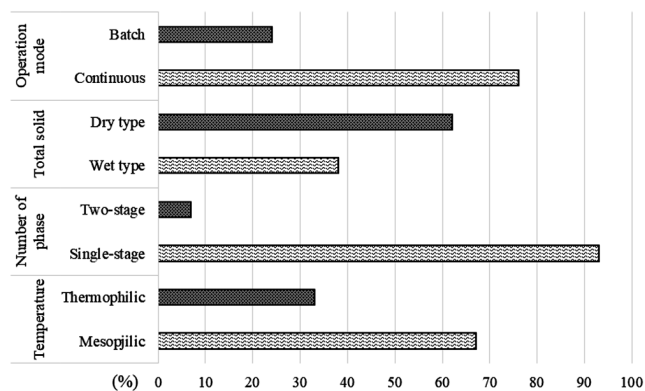

Fig. 4. Applications of AD systems in Europe based on cumulative capacity installed in 2014 [111].

The discovering a significant increase of OLR in the thermophilic condition compared to the mesophilic condition has brought many advantages such as reducing the volume of the reactor in a new design and raising the capacity of the old reactor. Although the thermophilic technology requires a high stability of waste stream because of the sensitivity of the thermophilic bacteria, this is no longer an obstacle to the development of current technology. Thermophilic AD systems have only been studied and applied in recent years, it takes a long time to become popular. Thus, mesophilic AD systems are still more dominant in the present. The dry single-stage digestion systems are more stable, more robust, have a smaller reactor, and produce less residual wastewater than the wet systems. It has been most applied in Europe to deal with OFMSW which accounts for the highest proportion of the biodegradable waste sectors. Thus, based on cumulative installed capacity, the dry single-stage digestion systems are still overwhelming applicated as shown in Fig. 4. The batch process often accompanies with disadvantages such as variable biogas production following time, long HRT, big reactor, and uncompleted degradation. However, it brings many advantages including simple material handling, less pre-processing requirements, easily controlling, and especially easy separation of hydrolysis and methanogenesis. Therefore, the batch process which has been soon being applied since 2009 for digestion of OFMSW, is estimated continuously to increase in use due to their simplicity and low cost [5, 111]. It is easy to understand the lack of three-stage system in Fig. 4 because the three-stage system has not been proved to have more methane yield than the two-stage system meanwhile it is much more complex in operating and also consumes higher energy.

5. Conclusions and Recommendations

The single-stage wet low-rate digestion systems are characterized by producing low biogas yield and low OLR (0.5-1.6 kg-VS/m^3.d^{-1}), requiring long RT (30-60 d) of the feedstock and big reactors, and depending on the outdoor temperature during the operation. However, they bring many advantages such as easy operation and cheap investment. In perspective economic benefits, they should only be applied for fine biodegradable waste sources (such as animal manures) without requiring any pretreatment step. Therefore, the wet low-rate system is a good solution for warm-climate rural areas, where agricultural land is available, and digestive products can turn back to serve for agricultural activities.

The single-stage wet high-rate digestion systems are much more effective than the wet low-rate systems with OLR up to 4-8 kg-VS/m^3.d^{-1}. However, the operation is more complex than the low-rate one because of the attached equipment and pre-treatment process sometimes. The feedstock with low solid contents (< 3-5%) is the best source for these systems with many selections of reactors such as UASB, FBR, EBR, EGBR, and CSTR. When TS > 5%, the CSTR is often used. In fact, these systems have been applied widely to deal with sewage sludge, agricultural products, and industrial wastewater. They can also be used to treat high solid feedstock (TS > 20%) such as OFMSW, but it requires to be diluted. At that time, water consumption and energy expense significantly

reduce the energy benefit of the system.

The single-stage dry digestion systems have brought a milestone for the AD technology, which can deal with high solid feedstock (TS = 30-35%) and high OLR (up to 15 kg-VS/m^3.d^{-1}). In case of too high solid contents in the feedstock (> 35%), it should be diluted. They are very suitable for handling OFMSW which accounts for a large amount of biodegradable waste sector. These systems can be distinguished by continuous operation mode or batch operation mode. For continuous mode, there are many advantages attached such as low water consumption, small reactor, very little wastewater production, cheap pre-treatment, waste residue well applied for composting, and less heat requirement. Thus, the continuous mode systems should be applied for urban areas. For batch mode systems, it costs lower for equipment invest and operation. However, it needs much larger space than the continuous mode systems and gas generation is variable. Therefore, the batch mode systems is a great solution for suburban areas.

The two-stage AD systems with separation of hydrolysis and methanogenesis in various reactors could have shorter HRT and higher OLR (up to 50 kg-COD/m^3/d) than the single-stage systems. They bring many advantages such as lower construction cost, much more operational flexibility, more robust, highest methane content, higher digestion efficiency, and especially well running with low C/N (< 10). The best application range for solid content of the feedstock is in the range of 5-15%. When TS < 5%, using the single-stage wet systems might better. When TS > 20%, the dilution of the waste stream causes a significant increase in energy needed for heating, pumping and expanding reactors, hence it is better to use the dry single-stage digestion system. The two-stage AD systems are suitable to deal with industrial wastewater.

The performance of the three-stage digestion systems has not been improved compared to the two-stage systems. Moreover, because it has more complex operation, more expensive investment, more energy for maintenance and operation compared to two-stage AD systems, application of the three-stage system for large scale is not a good selection at present. However, three-stage systems are still a good idea; its operation should be studied further to reduce the cost involved.

Ackowledgments

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