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JIANNING, DING
Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Sciences, Kyushu University

Eljamal, Osama
Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Sciences, Kyushu University

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Hydrogen production of anaerobic digestion: A review

DING JIANNING [1], Osama Eljamal [1*]

[1] Water and Environmental Engineering Laboratory, Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, 6-1 Kasuga-Koen Kasuga, Fukuoka 816-8580, Japan

*Corresponding author email: osama-eljamal@kyudai.jp

Abstract: *Low hydrogen production which is only 10% of the gas production is a disadvantage of bio-hydrogen production in an anaerobic digestion system. The two-stage AD process can achieve hydrogen production and methane production in two reactors respectively. In most studies, the pH of the hydrogen-producing reactor is controlled at 5.5, at this time the main hydrogen-producing bacteria Clostridium butyricum can have the maximum hydrogen production rate. In addition, HRT, temperature, and the concentration of external additives are also important operating conditions for the two-stage AD process. When the added concentration of NZVI is over 30 mM, the methane production process will be inhibited, but the hydrogen production can be promoted. Therefore, how NZVI promotes hydrogen production in the two-stage AD process is still worthy of attention in the future research.*

Keywords: Anaerobic digestion; Hydrogen; Methane

1. Introduction

Today where fossil fuels are still the main energy source, energy shortage and environmental pollution force us to seek a new alternative, clean and renewable energy. As a kind of promising energy, hydrogen has a high 122 kJ/g energy yield which was three times energy storage capacity per weight greater than the average liquid hydrocarbon, therefore the interest in hydrogen production has grown in recent years [1, 2, 3]. Hydrogen is a kind of renewable energy that could also achieve 100% renewable and zero carbon emission, so it has gained wider attention over the past few years[4, 5]. There are many processes for hydrogen production: seam reforming method, partial oxidation method, auto thermal reforming method, electrolysis, biological processes, thermochemical processes, and so on[6]. But some of this process has some disadvantages like high energy requirements. Bio-hydrogen production, such as anaerobic digestion technology, has huge socio-economic benefits due to its two advantages of organic waste management as well as renewable energy production (biogas). According to the reaction principle of anaerobic digestion, it also has great potential in hydrogen production[7].

Anaerobic digestion (AD) is one of the well-known and widely used technologies to treat organic solid waste[8]. It was considered as an eco-friendly technology because of its environmental benefits such as organic disposal and biogas production[4]. The AD process has four main sequential steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis (Fig.1)[5]. It obvious that the production of biogas includes methane, carbon dioxide, hydrogen and so on. But we can only detect small amounts of hydrogen in the biogas because hydrogen is an intermediate metabolite during the AD process[8]. And there are also some modeling was used in some pollution removal research, it is better for us to understand how the system works, so it may be another good way to be used in investigating the AD system[9, 10].

In recent years, much research has focused on hydrogen production by anaerobic digestion on how to increase hydrogen production. Zappi et al.[1] pointed out that in the operation of the anaerobic digestion process resulting in increased hydrogen production, it is extremely important to achieve inhibition of the conversion from hydrogen to methane. There are many studies that operate a two-stage anaerobic digestion system (as shown in Fig.2) the first reactor is the hydrogen production stage and the second one is the methane production stage. Some of them set a high feed rate (5.0 L d-1) so that hydrogen-producing bacteria became the dominant species and methane production was inhibited in the first reactor [11]. As a result, the total gas evolution

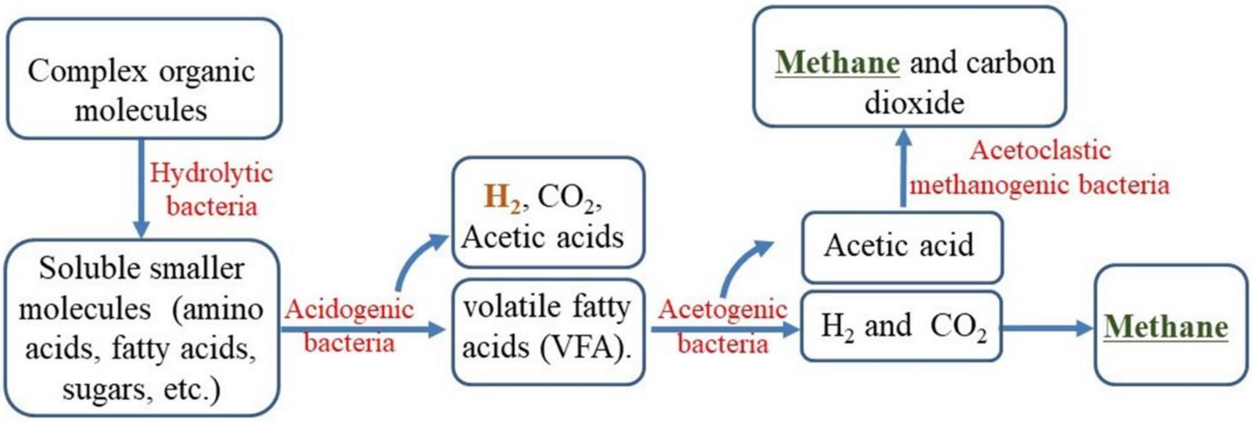


Fig.1 Anaerobic digestion processes four main steps.

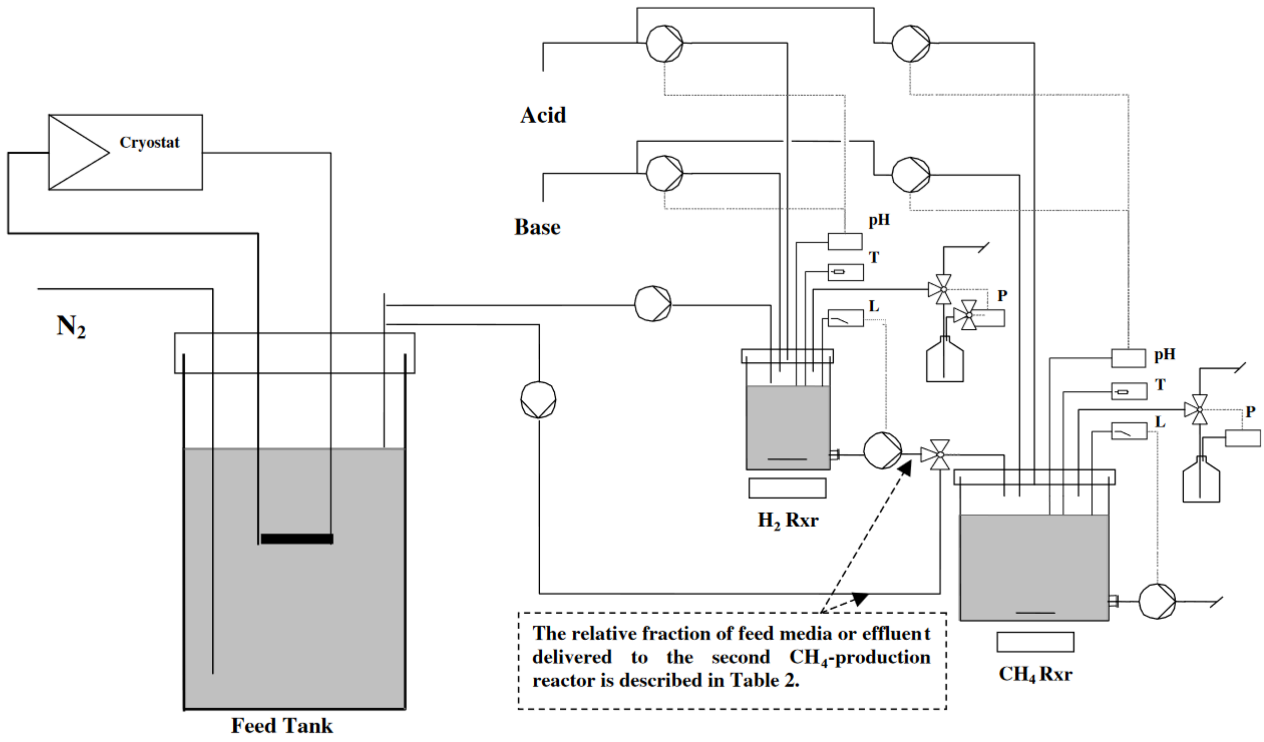


Fig.2 Two-stage anaerobic reactor system design[8].

rate was 139 ml h^{-1} which 35% were H_2 and only trace amounts of CH_4 in the hydrogen production reactor. In this system, many other factors could also increase the hydrogen yield. Guo et al.[12] shows that using high concentration of corn stalk as feedstock of anaerobic digestion combining with hydrogen fermentation could obtain the maximum hydrogen yield of 79.8 ml/g-cornstalk and hydrogen production rate of 3.78 ml/g-cornstalk/h.

As a renewable energy, hydrogen is better than methane because it doesn't have any carbon dioxide emission when it is burning. It is important to increase the production of hydrogen in the anaerobic digestion system. In fact, much research about hydrogen production by anaerobic digestion in recent years has mainly focused on two aspects: two-phase anaerobic digestion system operation and the method of increasing hydrogen production in the AD process. This review will focus on these two aspects to summarize the methods of improving hydrogen production in anaerobic digestion in recent years.

2. Two-phase anaerobic digestion system

In a two-phase anaerobic digestion system, the first hydrogen production reactor is also the acid production reactor combined with the second stage methanation reactor to establish a continuous flow bioreactor system. The two-phase AD system ensures the optimal growth conditions for the acidogenic bacteria and methanogenic bacteria through different but regular operating parameters [13]. The most important advantage of two-phase AD is that it is easy to cultivate different dominant bacterial species in two reactors so that hydrogen and methane gas can be generated in two reactors at the same time [14]. This means the hydrogen production will not be transferred to the methane production reactor [15]. In this section, recent research activities on the two-phase anaerobic digestion process are exclusively discussed.

Cooney et al.[11] sited the two bioreactors of 2 L (H_2 reactor) and 15 L (CH_4 reactor) working volume respectively in their two-stage continuous flow AD process study(Fig.2). Their study showed that selection pressure of pH, dilution rate, and the raised feed rate was

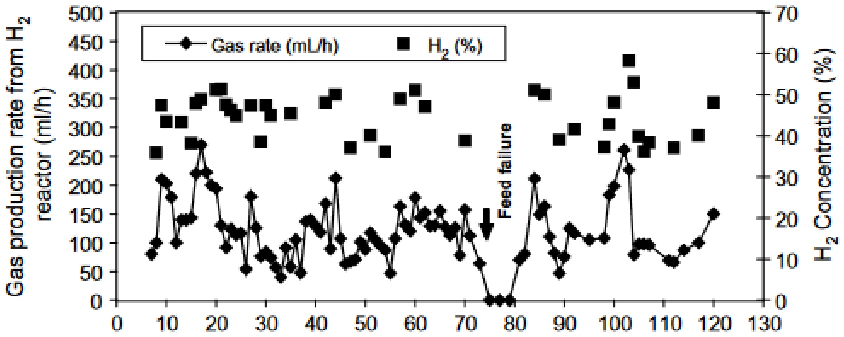


Fig.3 Biogas production in hydrogen reactor [13]

obviously increasing the hydrogen yield [11]. As a result, the percentage of hydrogen in the production gas was between 30% to 40% at the H_2 reactor.

Zhu et al.[16] got 270 ml/h (maximum) and 119 ml/h (average) gas production rate over 110 days in the H_2 reactor of their two-stage AD system when they controlled its pH at 5.5 and HRT at 6 h. Zhu et al.[16] also shows the average hydrogen concentration was 45% (v/v) in the gas production in the H_2 reactor (Fig.3). At the end of the H_2 reactor, the VS decreased with a 52% removal rate, revealing that almost half of the organic matter in sludge was stabilized and converted during when the hydrogen production and they present the stage of hydrogen production was an acetic–butyric type as shown in equation (1)[13].

$$C_6H_{12}O_6+H_2O$$
$$CH_3COO+1/2CH_3(CH_2)_2COOH+3H_2+2CO_2 \qquad (1)$$

Park et al.[17] detected three Clostridium species of hydrogen-producing bacteria (Clostridium butyricum, Clostridium letpum and Clostridium drakei) in the hydrogen production reactor in their two-stage AD process using molasses as the raw material. Clostridium butyricum is a kind of microbial species that widely exists in anaerobic sludge, so it has been highly concerned and studied. In many studies, it has been mentioned that bacteria can efficiently release hydrogen while a cell growing[18]. When the pH is 5.5, the best production capacity can be obtained, so most anaerobic digestion will control the pH at 5.5 in its production stage. As shown in table1, many studies focus on hydrogen production by two-phase AD process under different feedstock and operation conditions[19]. Although the hydrogen yield in different studies has significant differences, this table also shows two-phase AD system has huge potential to generate lots of hydrogen. The two-phase AD system will continue to be of interest in future hydrogen production research.

3. The mothed to increase the hydrogen production

Over the last two decades, many studies focus on how to increase the performance of biogas generation with AD system. There are three main methods which including pretreatment of feedstock, co-digestion, and external additives.

3.1 Pretreatment

The pretreatment method is one of the effective ways to improve the properties of sludge and improve the performance of sludge anaerobic digestion. It can prepare more available substrates from the raw material. There are many kinds of pretreatment technologies, such as mechanical, thermal, ultrasonic, aerobic, enzyme, alkali and acidic and so on[20].

Jia et al.[2] present eight methods including acid, alkali, heat, drying, ultrasound, aeration, sodium 2-

Table 1 production of bio-hydrogen and methane [19].

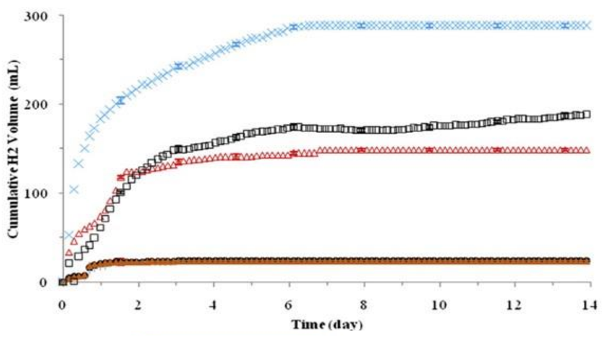
Production of Bio-hydrogen in two-stage food waste digesters.

Type of substrate	Type of digestion	Reactor volume (L)	Operating parameters of biohydrogen reactor	Operating parameters of biomethane reactor	Varying parameter	Hydrogen yield	Methane yield
Food waste	Two-stage	10, 40	Mesophilic (37 °C), 0.3 d, 47.8 g VS L^{-1}	Mesophilic (37 °C), 20 d, 37.2 g VS L^{-1}	–	99.8 mL g^{-1} VS	728 mL g^{-1} VS
Food waste	Two-stage	2, 4.5	Thermophilic (55 °C), 5 d, 15–18 kg VS $m^{-3}d^{-1}$	Mesophilic (35 °C), 9 d, 5.7 kg-VS $m^{-3}d^{-1}$	–	125 L kg^{-1} VS	526 L kg^{-1} VS
	Two-stage with recirculation		Thermophilic (55 °C), 5 d, 15–18 kg VS $m^{-3}d^{-1}$	Mesophilic (35 °C), 9 d, 5.7 kg-VS $m^{-3}d^{-1}$	Recirculation ratio - 0.3	135 L kg^{-1} VS	529.5 L kg^{-1} VS
					Recirculation ratio –0.5	113 L kg^{-1} VS	529.8 L kg^{-1} VS
					Recirculation ratio –1.0	43.8 L kg^{-1} VS	527.1 L kg^{-1} VS
Food waste	Two-stage	5,5	Mesophilic (35–37 °C), 0.5 d, 48 g COD L^{-1} d^{-1}, pH 6.0	Mesophilic (35–37 °C), 1 d, pH 7.0	–	292.7 mL g^{-1} VS	391.6 mL g^{-1} VS.
Raw food waste from restaurants and canteens	Two-stage	0.417,0.417	Mesophilic (35 °C), 2 d, pH 6.0	Mesophilic (35 °C), 24 d	–	43 mL g^{-1} VS	511.6 mL g^{-1} VS
Food waste + sewage sludge + glycerol	Two-stage	0.25,0.25	Mesophilic (35 °C), pH 5.5, 10 g VS L^{-1}	Mesophilic (35 °C), pH 7.0	1% Glycerol	140.2 mL g^{-1} VS	342 mL g^{-1} VS
Beverage waste water	Two-stage	1,1 (WV)	Mesophilic (35 °C), pH 5.5	Mesophilic (35 °C), pH 8.0, VS-37.7 g L^{-1}, TCOD – 70.2 g L^{-1}	HRT (d) - 12, 24	0.6 mL g^{-1} COD	4.7 mL g^{-1} COD
					HRT (d) - 8,24	1.3 mL g^{-1} COD	58.0 mL g^{-1} COD
					HRT (d) - 4,24	18.0 mL g^{-1} COD	7.0 mL g^{-1} COD
					HRT (d) - 2,24	172.0 mL g^{-1} COD	0.7 mL g^{-1} COD
Dairy processing waste	Two-stage	60,60	Thermophilic (60 °C), 3 d, OLR – 32.9 g COD L^{-1} d^{-1}	–	–	2358.3 mL H_2 L^{-1} d^{-1}	–
Cheese whey powder	Two-stage	2, 0.79	Mesophilic (37 °C), 0.25 d, OLR – 182 g COD L^{-1} d^{-1}	–	–	25,000 mL H_2 L^{-1} d^{-1}	–
Cheese whey	Two-stage	3, 15	Mesophilic (35 °C), 1 d, OLR – 47.4 g COD L^{-1} d^{-1}	–	–	2510 mL H_2 L^{-1} d^{-1}	–
Cassava waste water	Two-stage	4, 24	Thermophilic (55 °C), OLR – 12 g	–	–	54.22 mL H_2 g^{-1} COD applied	–

bromoethanesulfonate (BES), and chloroform and drying the sludge at 60 °C for 48 h could enhance fermentative hydrogen production. Guo et al.[21]present ultrasonic pretreatment increases methane production in anaerobic digestion products by increasing the hydrolysis process. Rafieenia et al. [22] demonstrate that aerobic pre-treatment has no positive impact on hydrogen production but it is a promising way to improve substrate conversion efficiencies and CH_4 production in the AD process.
The pretreatment method was widely used in improving biogas and methane production by AD system while no positive effect on hydrogen production.

3.2 Co-digestion

iron nanoparticles were used in removing pollution[35]. Yang et al.[36] proved that 30 Mm NZVI could inhibit methane production by disrupting cellular integrity. However, at the same concentration of ZVI powder, due to the slow release of hydrogen, it promotes the growth of hydrogenotrophic methanogens and improves methane production. In Fig.4, hydrogen production was increased with the increase of NZVI concentration. But it seems like the heat-killed pretreatment generated higher hydrogen. Although the hydrogen production of heat-killed pretreatment was higher than that of the AD system with NZVI, NZVI is the most applicable for the AD process due to the availability and affordability of the trace additive.



Cumulative hydrogen gas production profiles during mesophilic anaerobic glucose degradation in the presence of methanogenesis inhibitor BES, in the groups of negative control (no glucose, no iron, ○), positive control (glucose only, no iron, ●), 1 mM NZVI (+), 30 mM NZVI (△), and 30 mM ZVI (▲), respectively. A water sample containing 30 mM NZVI only served as an abiotic control to show the hydrogen production trend in Fig. 1b (×). A heat-killed sludge sample followed by the dose of 30 mM NZVI resulted in a different hydrogen production profile (□). Error bars represent the range of data from duplicate experiments. No methane was detected because of BES inhibition.

Fig.4 Cumulative hydrogen with different adding dosage [36]

Co-digestion is another effective way to improve the anaerobic digestion performance. Jokhio et al. [23] proved an anaerobic co-digestion system by treating banana plant waste (BPW) and sewage sludge (SS) could obtain better methane and hydrogen production. Dareioti et al. [24] give the optimal pH and hydraulic retention time (HRT) values at an anaerobic digestion system by treating a mixture of sorghum biomass solution with liquid cow manure (in a ratio of 95:5 v/v). When the pH was 5 and HRT was 5 d, the highest hydrogen yield was 0.92 mol H_2/mol carbohydrates consumed and 1.68 mol H_2/mol carbohydrates consumed respectively. Because it can not only treat a variety of organic wastes but also increase the production of biological methane and biological hydrogen, co-digestion is a sustainable and environmentally attractive method.

3.3 External additives

External additives have been used to increase the methane production rate by increasing the degradation efficiency of the organic matter of the waste sludge itself[25]. Biochar always be used as a sorbent material[26]. Biochar, as a kind of external additive, was investigated in the AD system. The biochar which has the important elements Fe, K, and Ca can increase the abundance of the H_2-producing bacteria Clostridium butyricum[27]. Sunyoto et al.[28] increased the 31% hydrogen yield in the AD system by adding the biochar.

On the other hand, more and more studies have focused on the effect of nano zero-valent iron in the anaerobic digestion process. ZVI has advantages such as operational simplicity and low cost[29, 30]. NZVI is widely used for contaminant removal and environmental remediation[31, 32, 33, 34]. NZVI and different kinds of

4. Conclusion

In the past decade, researchers have paid extensive attention to the two-phase anaerobic digestion system to increase the production of hydrogen and methane at the same time. In the two-phase anaerobic digestion system, it is divided into two reactors for hydrogen production (first stage) and methane production (second stage). During the operation of the two reactors, the pH was set to 5.5 and 7 respectively, and the feed rate was different, so that hydrogen production bacteria and methanogenesis bacteria were dominant in different reactors. Clostridium butyricum is detected in most anaerobic digestion systems. As the dominant hydrogen-producing bacteria when the pH is 5.5, the maximum hydrogen production rate can be achieved. In the anaerobic digestion system, pretreatment, co-digestion, and external additives are also three commonly used methods to improve gas production performance. Among these, NZVI can inhibit methane production because of its disruption of cell integrity. When the added concentration of NZVI is over 30 mM, the methane production process will be inhibited, but hydrogen production can be promoted. NZVI is the most applicable for the AD process due to its availability, it has great future development prospects in improving hydrogen production in the anaerobic digestion process.

Acknowledgments
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