Conversion of organic solid waste to hydrogen and methane by two-stage fermentation system with reuse of methane fermenter effluent as diluting water in hydrogen fermentation

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HIGHLIGHTS

- Two-stage system to convert organic solid waste to H_2 and CH_4 was investigated.
- (H_2 + CH_4-ASBR) system showed total biogas conversion of 78.6%.
- 2.03 L H_2/L_{system}/d and 1.96 L CH_4/L_{system}/d in (H_2 + CH_4-UASBr) system.
- Half of alkali addition reduction by using ASBR effluent as diluting water.

GRAPHICAL ABSTRACT
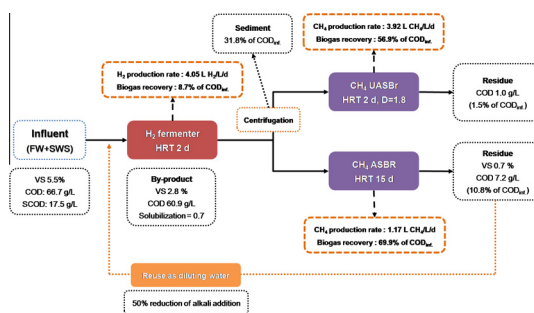
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ABSTRACT

In this study, a two-stage system converting organic solid waste (food waste + sewage sludge) to H_2 and CH_4 was operated. In the first stage of dark fermentative hydrogen production (DFHP), a recently proposed method that does not require external inoculum, was applied. In the second stage, anaerobic sequencing batch reactor (ASBR) and an up-flow anaerobic sludge blanket reactor (UASBr) were followed to treat H_2 fermenter effluent. (H_2 + CH_4-ASBR) system showed better performance in terms of total biogas conversion (78.6%), while higher biogas production rate (2.03 L H_2/L_{system}/d, 1.96 L CH_4/L_{system}/d) was achieved in (H_2 + CH_4-UASBr) system. To reduce the alkali addition requirement in DFHP process, CH_4 fermenter effluent was tested as a diluting water. Both the ASBR and UASBr effluent was effective to keep the pH above 6 without CH_4 production. In case of using ASBR effluent, H_2 production dropped by 15%, but alkali addition requirement was reduced by 50%.

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1. Introduction

Searching for alternative clean energy types to meet the ever-increasing energy demand and for the sustainable development of our modern society is an urgent issue. Specifically, hydrogen (H_2) offers tremendous potential as a clean and renewable energy currency. H_2 is strategically important, as it is associated with low CO_2 emission levels and is environmentally benign and sustainable. H_2 can be produced by fermentative bacteria under dark and light conditions, but the former (often called dark fermentative H_2 production, DFHP) expect paramount importance as an increasingly strategically superior process due to its fast reaction rate and lack of a requirement for light (Levin et al., 2004).

However, as DFHP alone can convert to H_2, even under an optimal condition, less than 33% of the electrons in hexose sugars, the second step of the process should be combined with a

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post-treatment. Although the three possible well-known processes of CH_4 fermentation, photo fermentation, and microbial fuel cell have been suggested as the second-step process (Tao et al., 2007; Li and Fang, 2007; Hawkes et al., 2007), the 'anaerobic $H_2 + CH_4$ process' is similar to the two-phase anaerobic digestion process, which has achieved enhanced stability, higher loading capacities and greater levels of process efficiency (Ke et al., 2005). This process may be practical if efficient biological modifications for H_2 production and stable H_2 production are provided in the first acidogenic reactor. An energy balance analysis confirmed that the anaerobic $H_2 + CH_4$ process led to greater positive net energy recovery than the single DFHP process (Ruggeri et al., 2010).

It is well known that DFHP is always accompanied by acid production and thus requires the addition of external alkali chemicals such as NaOH or KOH during fermentation to maintain a pH of 5.5–6.0. When CH_4 fermentation is followed by H_2 fermentation, the alkalinity increases because acid is removed and protein is further degraded to ammonia, leading to alkalinity. Thus, in order to decrease the amount of external alkaline solution that must be added, Chu et al. (2008), Lee et al. (2010), and Cavinato et al. (2011) reported a two-stage fermentation system that was successfully operated with the recirculation of CH_4 fermenter sludge. Taking into consideration the aforementioned issues, Kraemer and Bagley (2005) also recycled CH_4 fermenter effluent to H_2 fermenter; however, a H_2 production drop of 80% was observed due to methanogens activity, though it did allow the alkali addition to be reduced by 40%. Similarly, Jung et al. (2012) found that a drastic decrease of H_2 productivity was observed upon the utilization of non-heat pretreated CH_4 fermented effluent due to contamination by propionate-producing bacteria. This implies the need for a pretreatment when CH_4 fermenter effluent is added to DFHP process.

Therefore, in light of the above findings, the present study sought to develop a two-stage fermentation system for H_2 and CH_4 production. In the first stage of the process, an innovative developed batch process was employed which produces H_2 from heat-pretreated food waste without external inoculum addition (Kim et al., 2009). Sewage sludge was added as an auxiliary substrate to food waste, since it was already found that a slight amount of sewage sludge addition synergistically enhanced the DFHP performance owing to the much higher content of Fe and Ca in sewage sludge (Kim et al., 2011). In the second stage of the process, an anaerobic sequencing batch reactor (ASBR) and an up-flow anaerobic sludge blanket reactor (UASBr) were applied. The two-stage fermentation system was assessed in terms of total bioenergy recovery and chemical oxygen demand (COD) removal efficiency. In addition, it was attempted to use CH_4 fermenter effluent as diluting water in the H_2 fermentation process.

2. Methods

2.1. Feedstock preparation

The feedstock was a mixture of food waste and sewage sludge. Food waste collected from a school cafeteria in our university was shredded to particle size of less than 5 mm by a grinder. Sewage sludge was taken from a gravity sludge thickener line in a local wastewater treatment plant into which primary and secondary sludge were added at the same amounts by volume. The characteristics of food waste and sewage sludge used in this experiment are arranged in Table 1.

2.2. Two-stage fermentation system

The H_2 fermentation was conducted as described in our previous work (Kim et al., 2011). In detail, prior to addition, food waste

Table 1
Characteristics of food waste, sewage sludge, and their mixture (food waste:sewage sludge = 10:1 on COD basis, the detailed procedure to make a mixture is provided in Section 2.2).

Item	Unit	Food waste	Sewage sludge	Mixture
TCOD	g COD/L	136.1	40.2	66.6 ± 7.1
SCOD	g COD/L	39.7	3.9	17.5 ± 3.6
TS	g/L	117.5	20.5	58.7 ± 2.5
VS	g/L	113.7	15.5	54.8 ± 2.5
Carbohydrate	g COD/L	74.0	2.0	32.3 ± 3.2
TN	g N/L	3.0	1.5	2.3 ± 0.4
Ammonia	mg NH_4-N/L	236	258	132 ± 21
pH	–	4.9	7.2	5.4 ± 0.2

and sewage sludge were boiled at 90 °C for 20 min to prohibit methanogenic H_2-consuming activity. In the fermenter, first, food waste corresponding to 30 g Carbo. COD/L (g/L as carbohydrate COD) was placed in the fermenter. Then, sewage sludge equivalent to 10% of food waste on COD basis was added to the fermenter, and filled with tap water to reach an effective volume of 6.5 L. The increase of COD and volatile solids (VS) concentration was considerable by sewage sludge addition, but the carbohydrate concentration increase was negligible due to the low carbohydrate content in sewage sludge. After adding all substrates, N_2 gas was purged to provide an anaerobic condition. By using a pH sensor and pH controller, initial pH was adjusted at 8.0 ± 0.1 with 3 N KOH addition, and pH maintained 6.0 ± 0.1 during the fermentation. The produced gas was measured by a wet gas meter. In order to get the feed for the following CH_4 fermenter, the fermentation was repeated sixteen times, and each fermentation lasted for 2 d. The fermentation effluent was stored in refrigerator to block further microbial reaction.

For the CH_4 fermentation system, ASBR and UASBr were applied. One cycle period for the ASBR consisted 0.05 h filling, 18.9 h reaction, 5 h settling, and 0.05 h decanting (total 24 h). While H_2 fermenter effluent was directly fed to the ASBR, only the supernatant obtained by centrifugation at 10,500×g force for 10 min was fed to the UASBr. Organic loading rate (OLR) was controlled by hydraulic retention time (HRT) change (12–30 d) and dilution ratio (D = 1.8–10) in ASBR and UASBr, respectively. The HRT was set to 2 d in UASBr. The seed sludge of the ASBR was taken from the local anaerobic digester, while the CH_4-producing granule was collected from a local brewery wastewater treatment plant. All reactors were installed in a temperature controlled room at 35 ± 1 °C.

2.3. Reuse of CH_4 fermenter effluent as diluting water in DFHP

In this experiment, seven kinds of diluting water were prepared: (1) tap water; (2) ASBR effluent; (3) heat-treated ASBR effluent; (4) supernatant of ASBR effluent; (5) UASBr effluent; (6) heat-treated UASBr effluent; and (7) supernatant of UASBr effluent. Pretreatment was to inhibit methanogenic H_2-consuming reaction: heat treatment to inactivate non-spore forming microorganism and centrifugation to remove all particle matters including methanogens at high gravitational force. Heat treatment condition was 90 °C for 20 min and the supernatant was obtained by centrifugation at 10,500×g for 10 min. The pH, alkalinity and ammonia concentration of various diluting water are shown in Table 2. The HRT in ASBR and the dilution ratio (D) in UASBr when the effluent was taken were 15 d and 1.8 d, respectively. About 81 mL of food waste corresponding to 30 g Carbo. COD/L and 20 mL of sewage sludge were added and seven kinds diluting water was added to each batch bottle, reaching 200 mL. The temperature was maintained at 35 ± 1 °C using a water bath. The modified Gompertz equation