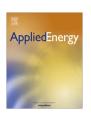
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Fermentative biohydrogen and biomethane co-production from mixture of food waste and sewage sludge: Effects of physiochemical properties and mix ratios on fermentation performance



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HIGHLIGHTS

- Microanalyses revealed food waste had more gelatinized organics and less mineral ash.
- Mixed food waste and sewage sludge at 5 ratios were used for H₂ and CH₄ co-production.
- Highest H₂ yield of 174.6 mL/gVS was achieved when food waste:sewage sludge was 3:1.
- Co-fermentation enhanced carbon conversion by strengthening hydrolysis of substrates.
- Energy yield rose from 1.9 kJ/gVS in H₂ to 11.3 kJ/gVS in H₂ and CH₄ co-production.

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ABSTRACT

The accumulation of increasingly generated food waste and sewage sludge is currently a heavy burden on environment in China. In this study, the physiochemical properties of food waste and sewage sludge were identified using scanning electron microscopy and Fourier transform infrared spectroscopy to investigate the effects on the fermentation performance in the co-fermentation of food waste and sewage sludge for biohydrogen production. The high gelatinized organic components in food waste, the enhanced bioaccessibility due to the dilution of mineral compounds in sewage sludge, and the balanced C/N ratio synergistically improved the fermentative biohydrogen production through the co-fermentation of food waste and sewage sludge at a volatile solids (VS) mix ratio of 3:1. The biohydrogen yield of 174.6 mL/gVS was 49.9% higher than the weighted average calculated from mono-fermentation of food waste and sewage sludge. Co-fermentation also strengthened the hydrolysis and acidogenesis of the mixture, resulting in a total carbon conversion efficiency of 63.3% and an energy conversion efficiency of 56.6% during biohydrogen production. After the second-stage anaerobic digestion of hydrogenogenic effluent, the energy yield from the mixed food waste and sewage sludge significantly increased from 1.9 kJ/gVS in the first-stage biohydrogen production to 11.3 kJ/gVS in the two-stage fermentative biohydrogen and biomethane co-production.

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1. Introduction

With rapid urbanization, the total production of municipal solid wastes (MSWs) in China amounted to 272 million tons with an annual growth rate of over 11% in 2013, but the delivering quantity of MSWs (173 million tons) only accounted for 63.6% of the total production [1–3]. Food waste (FW), which is the main component of the organic fraction of MSWs, occupied 51.8% of the total MSWs

* Corresponding author. E-mail address: juncheng@zju.edu.cn (J. Cheng). generated in China [2]. Similarly, the sewage sludge (SS) production in China reached up to 6.25 million metric tons of dry solids in 2013 and an average annual growth rate of 13% has been recorded during the past 6 years [4]. The accumulation of increasingly generated FW and SS significantly exacerbates their adverse effects on the environment.

In China, several FW disposal methods, including landfill, animal feed processing, aerobic composting, and anaerobic digestion [2], have been mainly applied. However, landfill has been thrown into doubt because of the environmental pollution and occupiedland-related problems. Feed processing has been gradually

forbidden because of the homologous pollution caused by feed derived from animal sources. Aerobic composting has also been controversial because FW yields high salinity [5] and ammonia emissions which result in secondary environmental pollution [6]. Similarly, the most commonly used SS disposal methods, such as sanitary landfill, incineration, and building material production [4], are either inefficient or energy-intensive. Nevertheless, anaerobic digestion not only reduces waste and greenhouse gas emissions, but also produces biomethane that is considered as an alternative energy carrier to traditional fossil fuels [7]. Therefore, anaerobic digestion has attracted increasing attention as an environmentally effective disposal method that combines waste reduction and bioenergy production [8].

Biomethane production through anaerobic digestion has been extensively investigated by using mono-substrate of FW or SS [9–14]. However, the high lipid contents often induce the accumulation of volatile fatty acids (VFAs) during the mono-digestion of FW. The lack of essential trace elements also inhibits methanogenesis in the long-term run [9]. Mono-digestion of SS is inefficient because of limited organic content, insufficient carbon sources [15], poorly biodegradable organic complex, and possible toxic compounds such as methanogen-inhibiting antibiotics [16]. Furthermore, the low energy values of SS and the limited organic loadings in mono-digestion of SS always result in a waste of processing capability of anaerobic digestion facilities [17]. Thus, the codigestion of FW and SS at suitable mixing ratios may synergistically enhance digestibility, biomethane production, and process stability by balancing the C/N ratio, offsetting the lack of nutrients, and diluting the possible toxic substances in the co-substrates [15,17-19]. Several investigations on the co-digestion of FW and SS have been carried out [15,20,21]. Kim et al. [21] added 50% of FW into SS on the basis of volatile solids (VS) and observed that the biomethane yield increased from 116 mL/gVS in monodigestion of SS to 215 mL/gVS in co-digestion. Another mixture of FW and SS at a VS ratio of 40:60 produced a biomethane yield of 200 mL/gVS due to the synergy of co-digestion and sequencing temperature-phased batch operation [15]. Koch et al. [20] found that the co-digestion of FW and SS not only increased the biomethane yield, but also accelerated the biomethane production

Although the co-digestion of FW and SS offsets nutrient imbalance and improves biomethane production, the prevailing pH (7-8), which benefits the methanogenic microbes in single-stage anaerobic digestion, provides a disadvantageous function condition for acidogenic bacteria. As a result, slow hydrolysis ensues and biomethane production rates are limited [14]. A two-stage process involving dark hydrogen fermentation in the first stage and anaerobic digestion in the second stage is supposed to be an alternative solution to achieve a positive net energy recovery and improve the process efficiency and stability [1]. In dark fermentation, acidic pH (4-6) favors the anaerobic fermentative bacteria (AFB) [22]. Hydrolysis and acidogenesis, which are two essential stages before acetogenesis and methanogenesis during anaerobic digestion, are optimized, and the degradation of the largemolecular-weight organics is facilitated. A large amount of biohydrogen as a clean alternative energy carrier and abundant soluble metabolic products (SMPs) such as short-chain VFAs and ethanol can be produced during the dark fermentation of carbohydrates [1,23]. Thus, the two-stage process can recover both biohydrogen and biomethane which can be utilized either separately or together as biohythane with enhanced combustion efficiency and emission performance [24,25].

The literature on the co-digestion of FW and SS typically includes two-stage biohydrogen and biomethane co-production [26–28]. However, the microstructural features of FW and SS for gaseous biofuel production through co-fermentation and co-

digestion have been rarely investigated. Hence, the effects of the distinct characteristics of each co-substrate on biohydrogen and biomethane co-production should be elucidated to enhance the co-fermentation and co-digestion performance at various mixing ratios of FW and SS. Thus, to fill in the gap in the state of the art, the objectives of the present study are to:

- (1) Identify the physicochemical properties of FW and SS using scanning electron microscopy (SEM) and Fourier transform infrared (FTIR) spectroscopy and analyze the effects on the subsequent fermentation performances.
- (2) Examine the effects of mix ratios on the first-stage fermentative biohydrogen and SMP production and the second-stage fermentative biomethane production.
- (3) Assess the overall energy yields of the gaseous biofuels during the two-stage process.

2. Materials and methods

2.1. Substrates and characterization

The substrates used in the experiments were FW and SS. FW was collected from a campus canteen in Zhejiang University, Hangzhou, China. Bones, plastic bags, and other non-biodegradable wastes were manually removed. FW was blended into a pulp in a blender. SS was collected from Jining Zhongshan Public Utilities Water Co., Ltd., Shandong Province, China. The raw substrates were cryopreserved at a temperature of $-20\,^{\circ}\text{C}$ before use.

2.2. Inocula

The inoculum for dark hydrogen fermentation was sourced from the anaerobic digestion sludge collected from a methane plant in Huzhou, Zhejiang Province, China. The original sludge was heated at 100 °C in an autoclave (Sanyo MLS-3780, Japan) for 30 min. After heat treatment, the methanogens were deactivated and the spore-forming AFB survived. Subsequently, the heat-treated sludge was acclimatized 3 times (3 days each time) using a modified culture medium under an anaerobic environment to enrich the spore-forming AFB. The first acclimatization process was used to activate the AFB, and the second and third ones were used to ensure that the AFB can fully function. The modified culture medium was detailed in a previous study [29]. Total solids (TS) and VS of the hydrogen inoculum were 94.3 and 47.7 g/kg, respectively.

The inoculum for anaerobic digestion was also from the anaerobic digestate of a methane plant in Huzhou, Zhejiang Province, China. The original digestate was filtered through a 1 mm sieve to remove all the big particles and then placed in an anaerobic workstation (Whitley DG250, UK) at 35 °C for 7 days to ensure that all the consumable residues were depleted before experiment. TS and VS of the methane inoculum were 58.5 and 31.8 g/kg, respectively.

2.3. Combined dark hydrogen fermentation and anaerobic digestion

Biohydrogen production through dark fermentation was conducted in 300-mL glass reactors. Five experimental groups according to different VS based ratios of FW to SS from 4:0 (Group 1), 3:1 (Group 2), 2:2 (Group 3), 1:3 (Group 4), to 0:4 (Group 5) were established with each one in triplicates. Substrates (5 gVS) and hydrogen inoculum (25 mL) were added into each reactor, and the total liquor volume was adjusted to 250 mL with distilled water. The initial pH was adjusted to 6.0 ± 0.1 with 6 M NaOH and 6 M HCl solutions. Subsequently the reactors were sealed using silicone rubber stoppers and purged with nitrogen gas for

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