ARTICLE IN PRESS

Bioresource Technology xxx (2017) xxx-xxx

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech



Long-term bio-H₂ and bio-CH₄ production from food waste in a continuous two-stage system: Energy efficiency and conversion pathways

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HIGHLIGHTS

- The continuous bio-H₂ and bio-CH₄ production was sustainable in long term.
- The optimized bio-H₂ and bio-CH₄ production parameters was obtained.
- The methane production was supposed to through SAO-HM pathway.

ARTICLE INFO

Article history: Received 9 April 2017 Received in revised form 25 May 2017 Accepted 26 May 2017 Available online xxxx

Keywords: Food waste Long-term experiment Hydrogen and methane Two-stage process

ABSTRACT

Anaerobic digestion is a well-established technology for treating organic waste, but it is still under challenge for food waste due to process stability problems. In this work, continuous H₂ and CH₄ production from canteen food waste (FW) in a two-stage system were successfully established by optimizing process parameters. The optimal hydraulic retention time was 5 d for H₂ and 15 d for CH₄. Overall, around 59% of the total COD in FW was converted into H₂ (4%) and into CH₄ (55%). The fluctuations of FW characteristics did not significantly affect process performance. From the energy point view, the H₂ reactor contributed much less than the methane reactor to total energy balance, but it played a key role in maintaining the stability of anaerobic treatment of food waste. Microbial characterization indicated that methane formation was through syntrophic acetate oxidation combined with hydrogenotrophic methanogenesis nathway.

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1. Introduction

The global economy is currently shifting from fossil energy supplies to renewable energy sources. Energy recovery from various organic wastes is of increasing interest owing to the large amounts available, low resource cost, and environmental benefits. Approximately 100 million Mg of food are wasted annually in Europe (Pagliaccia et al., 2016), and roughly 600 million Mg of food waste (FW) are generated yearly in China (Meng et al., 2015). High amounts of energy, i.e. 367 m³ of biogas per dry Mg, and an annual

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http://dx.doi.org/10.1016/j.biortech.2017.05.164 0960-8524/© 2017 Published by Elsevier Ltd.

yield of 894 TWh can be obtained from treating food waste by anaerobic digestion (Curry and Pillay, 2012).

Nevertheless, the bioenergy production of FW through anaerobic digestion (AD) needs optimization and the stability of the single stage AD process is still under challenge due to the low C/N ratio of FW and the unbalance between hydrolysis/acetogenesis and methanogenesis (Algapani et al., 2016). Two-stage anaerobic digestion technology separates the hydrolysis/acidification and methanogenesis into two optimized phases; this approach has been reported to guarantee a more stable and efficient process in comparison with the single-stage approach (Ghimire et al., 2015; Ariunbaatar et al., 2014). Higher biogas production in a temperature phased two stage AD process of FW has been widely reported (Wu et al., 2015; Zhang et al., 2014); Muhammad et al., 2012). High

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amounts of hydrogen could be obtained in the first hightemperature stage when the process was appropriately managed. The stability of methane production in the second stage would be improved as well by enhancing the balance between the production and consumption of volatile fatty acids. As a result, the co-generation of hydrogen and methane from FW can be a promising way to meet the multiple utilizations of the two energy gases separately or to use their mixture as bio-hythane (Thi et al., 2016). However, the co-generation of hydrogen and methane in a two-stage process has mostly been investigated in batch or short-term experiments (Rafieenia et al., 2017; Sunyoto et al., 2016; Pisutpaisal et al., 2014). Data on the optimization of the two-stage process towards energy production and process stability through a long-term experiment are scarce. Whether hydrogen and methane production were sustainable in the long term was still unclear. Studies investigating the anaerobic degradation of FW probably used artificial FW composed of rice, vegetable matter and meat (Nathao et al., 2013; Sunyoto et al., 2016). Whether the process performance would be as satisfactory with the normal variability of food waste characteristics was still not clear.

The simultaneous production of hydrogen and methane usually uses materials rich in carbohydrates such as sugarcane bagasse and syrup (Baêta et al., 2016; Nualsri et al., 2016), cassava wastewater (Intanoo et al., 2016) and molasses (Abd-Alla et al., 2014). However, FW is a very different substrate because of its high protein and lipid content, and information on the effects of lipids and proteins on the cogeneration of hydrogen and methane from FW is still limited and little investigated in long-term experiments (Thi et al., 2016).

The effluent from hydrogen fermentation contains high volatile fatty acids (VFAs) concentrations (Algapani et al., 2016). The methane formation reactor in the second stage of the process receives a material containing high acetate content, compared with the single process which needs hydrolysis and fermenting FW before getting acetate to produce methane. Theoretically, methane formation at elevated acetate concentration would thermodynamically favor the syntrophic methanisation of acetate: firstly the acetate would be oxidized into CO₂ and H₂ by syntrophic acetate-oxidizing bacteria and secondly converted into methane by hydrogen methanogens, i.e. utilizing the SAO-HM pathway (Westerholm et al., 2016). The elevated acids concentrations bring risks for reactor stability management. The stability management strategy would be renewed in SAO-mediated processes. The digester performance response to elevated acetate concentration fundamentally depends on the microbial community. Nonetheless, the possibilities of the SAO-HM pathway in hydrogen + methane system have not been investigated from the point of view of the microbial community.

The present study aimed to elucidate the co-generation of biohydrogen and bio-methane through two-stage anaerobic digestion of food waste with particular attention to the material and energy balance, process kinetics, and long-term process stability. Moreover, the microbial community structure in the two-stage reactors was investigated to reveal the bio-hydrogen and bio-methane production pathway.

2. Materials and methods

2.1. Characteristics of food waste

The FW in this study was collected every 2–3 weeks from the lunch residues from a canteen at China Agricultural University (Beijing, China). Undesirable compounds such as bones, waste paper and plastics were removed manually. Thereafter, the FW was ground for 5 min using a blender (Joyoung, JYLC012). The

FW slurry was then sealed in plastic bags and stored in a refrigerator at 4 °C. Before being fed into the digestion process, FW was diluted with tap water to obtain a total solid (TS) content of around 10% weight/weight (w/w). The 55 °C inoculums were taken from a full scale thermophilic reactor treating maize straw and the 35 °C inoculums were taken from a full scale mesophilic reactor treating sewage sludge. The inoculums were put into the hydrogen reactor that was then operated by shortening step by step the HRT. The hydrogen inocula were not pretreated before feeding to the reactor. The characteristics of the FW slurry and inocula at 55 °C and 35 °C in the digester are listed in Table 1.

2.2. Continuous two-stage anaerobic system

Two laboratory-scale continuous stirred tank reactors were used as the reactors for the hydrogen and methane fermentation. The first stage hydrogen reactor was maintained at 55 °C and the second stage methane reactor was maintained at 35 °C. The reactor temperature was maintained using a water bath (HH-60) which provides an accuracy of ±0.1 °C. Each reactor had a working volume of 4.5 L, and the slurry inside was stirred at a range of 50-90 rpm. The timer-controlled stirrer was automatically operated for 10 min every 2 h. The gas volumes produced were recorded with gas meters. The hydrogen reactor was operated as a pretreatment reactor of different HRT (15, 10, 5 and 3) days, and the optimized conditions for hydrogen production were: HRT of 5 days, pH of 5-5.5 and organic loading rate (OLR) of 16.3 kg-VS m⁻³d⁻¹, authors previous research (Algapani et al., 2016). Characteristics of the FW feeding the hydrogen reactor were tested every 3-6 days and average results are reported in Table 2. With the two reactors working together, the hydrogen reactor operated at HRT 5 days and the methane reactor was operated at HRTs of 30, 20, 15, 12, and 8 days. Trace metals were added into the methane reactor to maintain the following concentrations: 20 mg L⁻¹ of FeCl₂-4H₂O and 1 mg L⁻¹ of CoCl₂-6H₂O, NiCl₂-6H₂O, CuCl₂-2H₂O, ZnCl₂, MnCl₂-4H₂O, H₃BO₃, Na₂SeO_{3.} Na₂WO₄-2H₂O, and NaMoO₄-2H₂O. The 24-h experiment was carried out for the hydrogen reactor (HRT 5 days) when the reactor was in a stable condition. For the methane reactor, the 24-h experiment was done during the HRT 15 d period. The gas volume and composition were tested irregularly during the 24 h subsequent to feeding. The continuous two-stage system configuration is shown in Fig. S1 (Supplementary Materials).

2.3. Biogas production potential of hydrogen reactor effluent

The biogas yield and production rate from the hydrogen reactor effluent, the supernatant and the solid of the effluent were tested at 35 °C by using the biochemical methane potential (BMP) procedure. The mesophilic inocula were obtained from the methanogenesis reactor, which operated at a hydraulic retention time (HRT) of 15 days. Fifty mL of inocula were added to 120-mL bottles containing 0.5 g-COD TFW. The bottles were purged with an N_2 gas and incubated in a water bath shaker at 35 °C; two bottles without substrate were used as a control. The biogas produced from each bottle was analyzed with a flexible frequency in accordance with the volume of produced biogas. Biogas potential was simulated by the modified Gompertz model in Eq. (S1), and gas production rate was calculated by the first-order kinetic model in Eq. (S2) (Supplementary Materials). The detailed BMP text procedure was in accordance with previous research (Li et al., 2015).

2.4. Chemical analyses

TS, volatile solids (VS), suspended solids (SS), volatile suspended solids (VSS), chemical oxygen demand (COD), soluble COD, and ammonia ($NH_{2}^{+}N$) were measured following the

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