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A new method for the simultaneous enhancement of methane yield and reduction of hydrogen sulfide production in the anaerobic digestion of waste activated sludge

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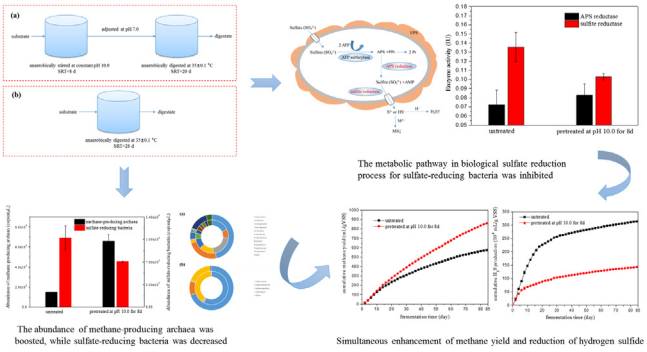
HIGHLIGHTS

- An approach was used to enhance the CH_4 yield while decreasing the H_2S production.
- Characteristic APS reductase and sulfite reductase were detected in this study.
- Alkaline fermentation had an adverse impact on the abundance of SRB.
- Alkaline fermentation made the acetotrophic methanogens more dominant.

GRAPHICAL ABSTRACT



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ABSTRACT

The biogas generated from anaerobic digestion (AD) also includes undesirable by-product such as hydrogen sulfide (H_2S), which must be removed before the biogas can be used as a clean energy source. Therefore, it is necessary to find an appropriate strategy to simultaneously enhance the methane yield and reduce H_2S production. An efficient strategy—pretreating sludge at pH 10 for 8 d and adjusting the system at neutral pH to produce methane for 20 d—is reported for the synchronous enhancement of methane production and reduction of H_2S production during AD. The experimental results showed that the cumulative methane yield was 861.2 ± 6.1 mL/g volatile solids (VS) of sludge pretreated at pH 10 in semi-continuous stirred anaerobic reactors for 84 d, an increase of 49.6% over the yield in the control. Meanwhile, the cumulative production of H_2S was 144.1×10^{-4} mL/g VS, 54.2% lower than that in the control.

1. Introduction

Waste activated sludge (WAS) is an undesirable byproduct of many municipal wastewater treatment plants (WWTPs) with activated sludge processes. WAS usually has a high organic matter content, and the degradation and stabilization of this organic matter to produce methane can be achieved; thus, the treatment of WAS by microbial anaerobic digestion (AD) has been applied (Wan et al., 2016). Typically, AD is performed at 35 °C or a thermophilic (55 °C) temperature. However, compared with thermophilic AD, mesophilic AD is more applied mainly because of its lower energy requirements and higher system stability (Gavala et al., 2003). Moreover, the mesophilic AD of WAS has

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many restrictions, such as a low degradation rate of organic matter, long sludge retention time (SRT), and especially low methane production (Braguglia et al., 2015; Dai et al., 2016b). Therefore, the improvement of methane production in sludge AD has become a topic of considerable interest. Previous studies reported that both hydrolysis and acidification were significantly enhanced when WAS was anaerobically pretreated with alkaline (pH 10.0 for 8 d), and high methane yield was obtained at neutral pH in batch experiments under mesophilic AD (Zhang et al., 2010). Nevertheless, whether alkaline fermentation can ultimately improve the methane yield is still unknown in a subject of semi-continuous experiments.

In addition to methane and carbon dioxide as its major components, biogas from AD fermentation includes trace compounds that must be removed before it can be used as clean energy. Hydrogen sulfide (H_2S) is the key pollutant in biogas and is produced by the reduction and degradation of inorganic and organic sulfur-containing compounds present in both primary and secondary sludge (Diaz et al., 2015). In the AD process, sulfate-reducing bacteria producing H_2S will oxidize sulfate as an electron acceptor in the oxidation of energy substrates. These sulfate-reducing bacteria can compete with methanogens for substrates (e.g., H_2 and acetate) in anaerobic fermentation (Elferink et al., 1994). However, H_2S can easily diffuse through the cell membrane into the cytoplasm (Chen et al., 2008), damaging methanogens, especially acetotrophic methanogens (Maillacheruvu and Parkin, 1996; Uberoi and Bhattacharya, 1997). Therefore, the presence of sulfate-reducing bacteria and H_2S will limit the generation of methane during AD.

Methods for removing H_2S from biogas can be divided into physicochemical and biological methods. The need for additional equipment and chemicals and high operating conditions (e.g., temperature and pressure) makes most physicochemical methods uneconomical and environmentally hazardous (Appels et al., 2008). Hao et al. (2016) operated a tubular zeolite/TiO_2 photocatalytic reactor and achieved a 98.1% H_2S removal rate and a 16.1% increase in bio-methane yield. Moreover, in the biological method, the application of micro-aerobic conditions during AD has been proven to be an effective method for removing H_2S from biogas (Diaz et al., 2015). Most researchers inject a small amount of air or oxygen into the liquid or gas phase of the anaerobic digester and achieve a high H_2S removal rate; however, the methane yield remains stable or even decreases (Diaz et al., 2010; Fdz-Polanco et al., 2009; Krayzelova et al., 2015).

Previous studies have focused on improving the in situ removal rate of H_2S from biogas, rather than enhancing methane yield, when facing these two challenges. Thus, it is necessary to find a more economical and effective way to significantly increase methane yield while reducing H_2S production in biogas. The purposes of this study were as follows: first, to determine whether alkaline fermentation can eventually increase methane yield in semi-continuous AD reactors; second, to elucidate the inhibitory effects of alkaline fermentation on H_2S production; third, to evaluate the effect of alkaline fermentation on changes to extracellular polymeric substances (EPSs) and Fe in the exchangeable fractions; fourth, to reveal the effect of alkaline fermentation on the activity of certain key enzymes (such as APS reductase and sulfite reductase) during sulfate reduction; and fifth, to elaborate the functional populations of bacteria and archaea using high-throughput 16S rDNA sequencing technology under the alkaline fermentation conditions of semi-continuous AD reactors. Real-time fluorescence quantitative polymerase chain reaction (PCR) technology was used to quantify the abundance of methanogens and sulfate-reducing bacteria to detect their changes under alkaline fermentation conditions compared to the control.

2. Materials and methods

2.1. WAS and inoculum

The WAS used for AD was obtained from the sludge-thickening tank of a municipal wastewater treatment plant (WWTP) in Shanghai, China, which operates using a traditional activated sludge process. The sludge from the thickening tank was concentrated by gravity thickening, and the main characteristics (average data plus standard deviations in duplicate tests) of the feedstock were as follows: pH 6.57 ± 0.20, total solids (TS) 25.5 ± 0.1 g/L, volatile solids (VS) 672.3 ± 31.4 g/kg-TS, TCOD 984.4 ± 10.4 g-COD/kg-TS, SCOD 27.1 ± 1.0 g-COD/kg-TS, total carbohydrate 101.6 ± 5.1 g-COD/kg-TS, total protein 628.4 ± 17.4 g-COD/kg-TS, and sulfate 9.28 ± 0.04 g/L. The WAS was collected every 30–40 d and kept at 4 °C. Before anaerobic fermentation, all substrates were mixed sufficiently and preheated to approximately 35 °C.

The mesophilic seed sludge was obtained from an anaerobic reactor with no feeding for approximately one month following the continuous treatment of dewatered sewage sludge for approximately six months. The main characteristics of the inoculum were as follows: pH 7.50 ± 0.12, TS 20.6 ± 0.8 g/L, VS 530.1 ± 25.2 g/kg-TS, TCOD 740.9 ± 28.1 g-COD/kg-TS, SCOD 32.0 ± 1.2 g-COD/kg-TS, total carbohydrate 194.0 ± 8.3 g-COD/kg-TS, and total protein 274.7 ± 10.1 g-COD/kg-TS.

2.2. Operation of semi-continuous stirred anaerobic reactors

Two sets of semi-continuous stirred anaerobic reactors with a working volume of 1.5 L were operated (see Fig. 1). Each set had three replicate reactors. In set one, 1 L of raw sludge was pretreated at pH 10.0 ± 0.2 by the addition of 4 M calcium hydroxide ($Ca(OH)_2$) or 4 M hydrogen chloride (HCl) before the sludge was added to the first-stage reactor, and every day, 125 mL of the digestion mixture was manually withdrawn from the first-stage reactor. The same amount of pretreated mixture was added, which means that the HRT (hydrolytic retention time) was 8 d (Yuan et al., 2006), while 50 mL of the digested mixture withdrawn from the first-stage reactor was added to the following second-stage reactor to maintain the HRT as 20 d. In set two, 1 L of raw sludge was directly added to the single-phase reactor without pretreatment. Every day, 35.7 mL of the fermentation substrate was manually withdrawn from the single-phase reactor, and the same amount of sludge was added, which means that the HRT was 28 d (8 + 20 d). Each reactor was inoculated with 200 mL of AD sludge before being flushed with nitrogen gas to maintain an oxygen-free environment and then sealed with rubber stoppers. The temperature of the semi-continuous-flow experiments was controlled at 35 ± 1 °C, and the agitator speed was maintained at 120 ± 5 rpm (revolutions per minute). The biogas yield, H_2S content, methane content in biogas, and pH of these reactors were measured every second day for the entire period. When these reactors became stable after 84 d of AD, the key enzyme activities and functional microbial community and populations in the anaerobic reactors were analyzed.

2.3. Batch experiments for inhibitory effects of alkaline fermentation on H_2S production

To explain the inhibitory effects of alkaline fermentation (pH 10 for 8 d) on H_2S production, two batches of experiments (untreated substrate and pretreated substrate) were performed to simplify the AD system using a synthetic solution. The organic fermentation substrate was 20.0 g of sodium acetate, and the organic matter was dissolved in 300 mL of synthetic solution in each serum bottle containing (mg/L) 200 Na_2SO_4, 405 $NaHCO_3$, 155 $K_2HPO_4{\cdot}3H_2O$, 50