Chemical Engineering Journal 259 (2015) 787–794

Contents lists available at ScienceDirect

Chemical Engineering Journal

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

Enhanced production of methane from waste activated sludge by the combination of high-solid anaerobic digestion and microbial electrolysis cell with iron–graphite electrode

Yinghong Feng, Yaobin Zhang *, Shuo Chen, Xie Quan

Key Laboratory of Industrial Ecology and Environmental Engineering (Dalian University of Technology), Ministry of Education, School of Environmental Science and Technology, Dalian University of Technology, Dalian 116024, China

HIGHLIGHTS

- High production of methane was achieved using iron–graphite electrode with 0.3 V.
- 0.3 V also increased the transformation of SCOD and the conversion of VFA.
- Energy consumption at 0.3 V could be neglected compared to the incremental methane.
- Further increase the voltage to 0.6 V led to the accumulation of hydrogen.
- Iron–graphite electrodes had bioaugmentation effect for both archaea and bacteria.

ARTICLE INFO

Article history:
Received 30 May 2014
Received in revised form 15 August 2014
Accepted 17 August 2014
Available online 23 August 2014

Keywords:
Waste activated sludge
Microbial electrolysis cell (MEC)
Anaerobic digestion
Iron
Methane production

ABSTRACT

Batch tests were operated to investigate the bioelectrochemical enhancement of methane production from the high-solid anaerobic digestion of waste sludge in the microbial electrolysis cells (MEC) with iron–graphite electrode. Compared with the control tests, methane production in the MEC with iron–graphite electrode increased by 22.4% and VSS removal rate increased by 11% at an applied voltage of 0.3 V. However the methane production decreased and hydrogen was cathodically produced when increasing the voltage to 0.6 V. At the higher voltage, the excessive utilization of H^+ in the cathode led to the alkaline pH to inhibit the methanogenesis. The applied voltages of 0.3 V could also enhance the removal of suspended and volatile suspended solids. The input of energy at 0.3 V could be neglected compared to the incremental energy generated from the methane. Denaturing gradient gel electrophoresis analysis revealed that the operation at 0.3 V had a bioaugmentation effect for both archaea and bacteria in the high-solid anaerobic digestion of waste sludge, which might be useful for enhancing VFA formation and methane production.

1. Introduction

Waste activated sludge is the byproduct during waste water treatment process, and its treatment and disposal cost about 50% of operating cost of many wastewater treatment facilities [1]. With the increasing population in cities and towns and construction of new waste water treatment plants, the yield of waste activated sludge has increased continuously in the recent decades. In China, over 11.2 million tons of dry sludge is generated annually and almost 80% of it has not obtained necessary stabilization [2]. To minimize the volume of waste sludge, most municipal sludge before discharge has been dewatered to become high-solid sludge with total solid content typically greater than 10% (w/w).

Anaerobic digestion is an appropriate technique for the treatment of sludge before final disposal and it is employed worldwide as efficient and sustainable technology to stabilize sludge. Recently, the high-solid anaerobic digestion (solid content > 10%) has gotten much attention since its smaller reactor volume, lower energy requirements for heating, less material handling compared to traditional anaerobic digesters with low-solid [3]. Generally, anaerobic digestion consists of three stages: hydrolysis, acidification and methanogenesis [4]. Methane is the final produce which can help cut down the operation cost of treatment plants. However, the yield of methane is usually limited by slow hydrolysis of sludge [5,6].

* Corresponding author. Tel.: +86 411 8470 6460; fax: +86 411 8470 6263.
E-mail address: zhangyb@dlut.edu.cn (Y. Zhang).

http://dx.doi.org/10.1016/j.cej.2014.08.048
1385-8947/

Iron, as a cheap reductant, has been proved to enhance the performance of waste sludge anaerobic digestion [7,8]. Iron could serve as electron donor to decrease oxidative-reductive potential (ORP), thus creating a more favorable environment for anaerobic microorganisms [9]. The releasing Fe^{2+} enhanced the activity of the enzymes associated with hydrolysis-acidification [10]. It was also reported that the addition of iron into the anaerobic system could boost the growth of H_2-utilizing methanogens to consume H_2 produced in anaerobic digestion which is an unfavorable intermediate in thermodynamics [7]. Despite the enhanced performance of anaerobic sludge digestion with addition of iron in the lab scale, this method is challenged in the full scale by the way of adding iron. Direct addition of scrap iron or powder iron into anaerobic digester might give rise to either iron precipitation in the bottom or washing out from the digester along with the effluent. As a result, the iron added cannot function sustainably.

MEC is a novel technology that may enhance the decomposition of organic matters coupled with the production of fuels and chemicals, such as hydrogen or ethanol with an external electricity supplement [11]. It has recently reported that methane was produced from cathodic reduction of CO_2 using microorganisms as the catalysts in MEC [12]. In the methane-producing MEC, bacteria (exoelectrogens) oxidized the substrate and released electron to anode, and then electron was transferred to cathode to produce methane. The reaction at the cathode is catalyzed by electrochemically active microorganisms, that is, hydrogenotrophic methanogens [13].

In this study, iron was set as the anode of a single-chamber MEC to enhance the anaerobic digestion of waste sludge. We assumed that the iron electrode could sustainably serve as a source of electron donor to enhance the anaerobic digestion to address the difficult of adding iron. Also, the anodic oxidation of organics is a biocatalytic process driven by electrogens which are generally categorized to iron-reducing bacteria (IRB) [14,15]. IRB can transfer the electron from organic oxidation to solid mediums, such as iron oxides or electrodes [16]. We assumed that the iron anode was possibly helpful for enrichment of the IRB because Fe^{2+} released from the iron electrode might be oxidized into Fe(III) oxides even at a quite low concentration of oxidants such as oxygen, SO_4^{2-} and NO_3^- [17,18]. The main objectives of the research were to investigate whether or not improvement of waste sludge anaerobic digestion occurred by using MEC with iron–graphite electrode and to study its mechanisms. Also, the microorganism communities functioning in the anaerobic digestion were identified and explored.

2. Materials and methods

2.1. Characteristic of waste sludge

Raw sludge from the Chunliu Wastewater Treatment Plant (Dalian, China) was used as substrates for this study. The seed sludge were collected from an UASB reactor at laboratory. Before the digestion, the raw sludge was mixed with the seed sludge with a ratio of 9:1. The characteristics of raw sludge, seed sludge and sludge mixture are listed in Table 1.

2.2. Batch experiment

A pair of Fe tube electrode (Φ 100 × 180 mm, anode) and graphite pillar electrode (Φ 8 × 180 mm, cathode) was inserted into a cylindrical acrylic plastic batch anaerobic reactor (Φ 110 × 250 mm) to form an electric-biological reactor (hereafter referred to as MEC-anaerobic reactor). The working volume of the reactor was 2.0 L. The graphite pillar electrode was located in the axes of Fe tube electrode, and they were connected to a DC power source through an electric wire. The voltage supplied was fixed at 0, 0.3 and 0.6 V. The control experiments were conducted in a common reactor that was the same as MEC-anaerobic reactor but without electrodes. Before digestion, oxygen was removed from the headspace by exchanging it with nitrogen gas for 10 min, and then sealed the reactors. A silica tube across the cap of reactors was connected to the gasbag. During the digestion, the biogas produced from each reactor was collected into gasbag. The biogas in gasbag was drawn out by a syringe every day for measuring volume and component. The reactors were operated as a batch mode and the digestion was lasted for 22 d. The reactors were stirred at 120 rpm with magnetic stirrers during the fermentation. All experiments were operated at 35 °C.

2.3. Analyses and calculations

During the operation, 5 mL of sludge samples were taken from the reactors every 2 or 3 days for analyzed. The samples were centrifuged at 8000 rpm for 10 min and immediately filtered through a 0.45 μm pore size cellulose membrane filters for analysis of SCOD, soluble protein, soluble polysaccharide and VFAs. TSS, VSS, TCOD and SCOD were determined according to Standard Methods for the Examination of Water and Wastewater [19]. Proteins were measured with Lowry's method using bovine serum albumin as a standard solution [20]. Polysaccharide was measured with phenol–sulfuric acid method using glucose as a standard solution [21]. The equivalent relationships between COD and substrates were as follows: 1.5 g-COD/g protein, 1.06 g-COD/g carbohydrate, 1.07 g-COD/g acetate, 1.51 g-COD/g propionate, 1.82 g-COD/g butyrate and 2.04 g-COD/g valerate [22]. The current during fermentation was recorded by a multimeter. The pH was recorded using a pH analyzer (Sartorius PB-20, Germany).

The concentration of methane and hydrogen in the biogas was analyzed with a gas chromatograph (Shimadzu, GC-14C) equipped with a thermal conductivity detector and a 1.5 m stainless-steel column (Molecular Sieve, 80/100 mesh). The temperatures of injector, detector and column were kept at 100 °C, 105 °C and 60 °C according to Zhao et al. [23]. Nitrogen was used as the carrier gas at a flow rate of 30 ml/min. VFAs (acetate, propionate, butyrate and valerate) were measured in another gas chromatograph

Table 1
Characteristics of the raw sludge, seed sludge and sludge mixture.

Parameters	Raw sludge	Seed sludge	Sludge mixture
pH	7.3 ± 0.1	7.7 ± 0.1	7.5 ± 0.1
TSS (total suspended solids, g/L)	117.9 ± 8.1	62.6.2 ± 6.3	103.2 ± 5.9
VSS (volatile suspended solids, g/L)	77.2 ± 3.9	33.7 ± 2.3	62.7 ± 3.8
TCOD (total chemical oxygen demand, g/L)	114.2 ± 9.2	50.0 ± 7.7	106.5 ± 8.1
SCOD (soluble chemical oxygen demand, g/L)	4.1 ± 0.6	5.2 ± 0.4	4.3 ± 0.3
VFA (volatile fatty acid, g/L)	0.9 ± 0.2	1.6 ± 0.2	1.1 ± 0.2
Total protein (g/L)	29.0 ± 1.8	12.7 ± 1.2	27.5 ± 1.6
Total polysaccharide (g/L)	13.5 ± 1.0	4.8 ± 0.4	12.4 ± 0.8

Average data and standard deviation obtained from three tests.