INTERNATIONAL JOURNAL OF HYDROGEN ENERGY XXX (2014) 1–9

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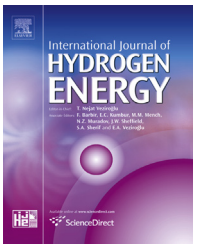

Optimization of high-solid waste activated sludge concentration for hydrogen production in microbial electrolysis cells and microbial community diversity analysis

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ARTICLE INFO

Article history:
Received 11 July 2014
Received in revised form
13 September 2014
Accepted 29 September 2014
Available online xxx

Keywords:
Waste activated sludge (WAS)
High-solid
Microbial electrolysis cell (MEC)
Hydrogen production
Microbial community

ABSTRACT

To enhance hydrogen recovery from high-solid waste activated sludge (WAS), microbial electrolysis cells (MECs) were used as an efficient device. The effects of WAS concentrations were firstly investigated. Optimal concentration for hydrogen production was 7.6 g VSS/L. Maximum hydrogen yields reached to 4.66 ± 1.90 mg-H_2/g VSS and 11.42 ± 2.43 mg-H_2/g VSS for MECs fed with raw WAS (R-WAS) and alkaline-pretreated WAS (A-WAS) respectively, which was much higher than that obtained traditional anaerobic digestion. Moreover, no propionic acid accumulation was achieved at the optimal concentration. Effective sludge reduction was also achieved in MECs feeding with A-WAS. 52.9 ± 1.3% TCOD were removed in A-WAS MECs, meanwhile, protein degradation were 50.4 ± 0.8%. The 454 pyrosequencing analysis of 16S rRNA gene revealed the syntrophic interactions were existed between exoelectrogen *Geobacter* and fermentative bacteria *Petrimonas*, which apparently drove the efficient performance of MECs fed with WAS.

Introduction

As a result of fast development of cities in China, over 60 billion tons waste water is discharged nowadays. Waste activated sludge (WAS) is the main byproduct of wastewater treatment plants (WWTPs), the annual production is estimated to be over 30 million tons in China [1]. Due to the expensive cost of WAS management and disposal for WWTPs [2], directly discard or landfill cause serious environmental issues. Anaerobic biological method, as an environmentally beneficial and sustainable way, has been commonly used to produce methane from WAS [3]. Yet, increasingly attention has been drawn on bio-hydrogen production from WAS due to its high energy yield (142.9 kJ/g) and being a carbon-neutral energy carrier [4,5]. It is known that biohydrogen conversion efficiency of WAS was limited due to complex carbons which were degraded slowly in anaerobic fermentation. Protein,

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http://dx.doi.org/10.1016/j.ijhydene.2014.09.163

Please cite this article in press as: Sun R, et al., Optimization of high-solid waste activated sludge concentration for hydrogen production in microbial electrolysis cells and microbial community diversity analysis, International Journal of Hydrogen Energy (2014), http://dx.doi.org/10.1016/j.ijhydene.2014.09.163

which accounts for ~40% of WAS, is difficult to be directly utilized by fermentative bacteria to produce hydrogen.

Recently, development of microbial electrolysis cells (MECs) has been showing a promising solution to enhance hydrogen production [6,7]. Electrons, which generated by exoelectrogenic bacteria from substrate degradation on the anode, were transferred to cathode and combined with the protons to form hydrogen under a small applied voltage (0.2–0.8 V) [8]. Various biomass had been proven to be the feasible substrates for MECs, such as domestic wastewater, corn stalk fermentation liquid and winery wastewater, etc. [9–12]. Recently, WAS is proven to be an ideal feedstock for hydrogen production in MECs [13–16]. Previous study showed that H_2 yields increased from 3.89 ± 0.39 mg-H_2/g-DS (raw WAS) to 6.78 ± 0.94 mg-H_2/g-DS (alkaline pretreated WAS) in two-chamber MECs [11]. Guo et al. (2013) used raw WAS as substrate and produced only 7.7 mL hydrogen in a 300 mL-MECs [16]. Xu et al. (2013) also obtained 46 ± 1 mL H_2 (0.8 V applied voltage) from WAS fermentation liquid, that is obtained by combined bi-frequency ultrasonic and alkaline pretreated WAS fermentation, in single-chamber MECs [14].

The organic concentration of the substrates is one of the major interests in bioelectrochemical systems [17]. Although the performance of MECs using WAS or pretreated WAS had been recently investigated and documented in researches, few studies had drawn attention on the effects of WAS concentration regarding to maximum hydrogen yield, organic removal and functional communities in MECs. Previous researches mainly used fermentation liquid of WAS as substrate to produce hydrogen in MECs [13,14,18]. Few researches put solid WAS directly into MECs for sludge reduction or hydrogen production because the complex organics, like protein [19], were also fermented simultaneously, which cause competitions or syntrophic interactions between fermentative bacteria and exoelectrogens. Up to now, the effect of the concentrations of solid WAS on hydrogen production and microbial community structure had not been reported in the literature before.

In our present study, the feasibility of hydrogen production in the MECs feeding with high-solid WAS was investigated. In terms of maximum H_2 production and sludge reduction, the effect of influent WAS concentrations were firstly discussed. Meanwhile, we used 454 pyrosequencing of 16S rRNA gene to access syntrophic interactions of microbial communities and to characterize both the microbial phylogenetic and functional communities in the anode biofilm of MECs feeding with high-solid WAS.

Materials and methods

Raw and alkaline-pretreated WAS

WAS was collected from the secondary sedimentation tank of the Harbin Wenchang wastewater treatment plant. After 24 h-precipitation at 4 °C, the supernate and impurities were removed and the remaining sludge was used as raw WAS (R-WAS). Based on previous study [20], we used alkaline pretreated WAS as substrate for MECs in this study to improve hydrogen recovery from WAS. The pH of alkaline-pretreated WAS (A-WAS) was initially adjusted to 12 by 4 M NaOH. The specific alkaline dosage for A-WAS was 7.27 ± 0.35 g NaOH per gram of volatile suspended solids (VSS). After precipitation for another 24 h, the pH of A-WAS remained at 9.49 ± 0.22. The characteristics of R-WAS and A-WAS were shown in Table 1.

MECs set-up and operation

Single-chamber MECs with the volume of 25 mL were used in our study as described in previous literature [6]. Carbon cloths coated with Pt/C catalyst were used as cathode, and graphite fiber brushes were used as anodes. An anaerobic tube linked with a gas bag was glued to the top of the reactor for gas collection. Voltages across the resistance (10 Ω) were recorded by the Keithley 2700 data system.

The applied voltage of MECs was 0.6 V and the temperature was 20 ± 2 °C. Six parallel MECs were constructed and divided into two groups. One group with three reactors (R-WAS MECs) used R-WAS as inoculum and substrate, and one of them was running as open circuit control. The other three used A-WAS as inoculum and substrate (A-WAS MECs) with one of them as open circuit control. All the MECs were directly started up in fed-batch MEC mode [7]. Three WAS concentrations were prepared by diluting R-WAS or A-WAS with 50 mM PBS (V_{WAS}:V_{PBS} of 1:1, 2:1, 3:1), which were 5.7 g VSS/L, 7.6 g VSS/L and 9.5 g VSS/L, respectively. The composition of 50 mM PBS was stated as before [21]. All reactors were refilled when the voltage decreased to ~8 mV. After all the MECs achieved stable performance, the influent and effluent were collected for chemical analysis.

Analyses and calculations

The TCOD, SCOD, SS, TS, VS and VSS were measured according to the standard methods [22]. The TN and TOC in the sludge samples was analyzed using TOC-5000 Total Organic Carbon Analyzer (Shimadzu, Kyoto, Japan). Phenol-sulfuric method was used to obtain the carbohydrate concentration with glucose as the standard [23]. Protein concentration was measured by Bicinchoninic Acid Assay with bull serum albumin as the standard (BCA Protein Assay Kit, Pierce) [24]. The pH

Table 1 – Characteristics of buffered R-WAS and A-WAS.

	R-WAS	A-WAS
Suspended solids (SS, g/L)	20.25 ± 0.16	20.26 ± 1.33
Total solids (TS, g/L)	23.98 ± 1.60	23.03 ± 1.06
Volatile solids (VS, g/L)	14.09 ± 0.01	16.32 ± 0.82
Volatile suspended solids (VSS, g/L)	11.00 ± 0.23	12.17 ± 0.59
Total chemical oxygen demand (TCOD, mg/L)	25,150 ± 1364	27,533 ± 1236
Soluble chemical oxygen demand (SCOD, mg/L)	501 ± 58	3189 ± 240
Total carbohydrate (mg COD/L)	43 ± 2	460 ± 18
Total protein (mg COD/L)	9034 ± 363	9653 ± 380
Soluble protein (mg COD/L)	605 ± 23	2931 ± 149
pH	6.85 ± 0.09	9.49 ± 0.22
Volatile fatty acids (VFAs, mg COD/L)	412 ± 3	1236 ± 79
Total organic carbon (TOC, mg/L)	236 ± 3	2436 ± 23
Total nitrogen (TN, mg/L)	109 ± 5	319 ± 20
Moisture content (%)	97.78 ± 1.70	99.31 ± 0.87
Conductivity (mS/cm)	3.67 ± 0.19	5.03 ± 0.03

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