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Article

## Effects of Electric Current on Performance of Bio-hydrogen Production

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Abstract: Fermentation is the most beneficial process for hydrogen (H<sub>2</sub>) production. The previous studies were mostly focused on medium, microorganisms, pH control, temperature, and hydraulic retention time. In this study, we used electric current to measure the H<sub>2</sub> production rate and its concentration. We observed the microorganism morphology to determine the effect of electric current on the microorganism growth state. The results showed the range of H<sub>2</sub> production rate and its concentration were 1,200–1,350 mL/L·hr and 32–42% with the current of 1.05–3.66 mA/cm<sup>2</sup>. H<sub>2</sub> concentration decreased to 2.09 mA/cm<sup>2</sup>. However, with the shorter rod-shaped microorganisms, the production time was shorter than the original strain.

Keywords: Fermentative process, Hydrogen production, Microorganism, Electric current

#### 1. Introduction

Alternative energy has been paid much attention in recent years. Fossil fuels are expected to be in shortage, and global warming is accelerated. In the pursuit of clean and pollution-free energy alternatives, related research in recent years has been conducted extensively. Hydrogen gas is an excellent alternative energy source as only water is generated after using it as a fuel [1-4]. Hydrogen is produced from methane by the physical and chemical or biological reaction or by electrolysis of water. Both production processes require a lot of energy and high manufacturing costs, while biological production using metabolism consumes less energy. Electrolyte-supported cells allow the lower degradation of cells at lower currents, which lowers the hydrogen production cost [5]. By using anaerobic bacteria mixed with organic waste sludge, a high concentration of hydrogen is produced showing the future development potential. The varieties of hydrogen biological products are shown in Table 1. In this study, we applied an electrical current to speed up microbial metabolism and capacity. The relationship of hydrogen-producing bacteria and the current was observed, too.

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Process	Organisms	Light	Electron Source	Products	Advantages	Representative Organism	Maximum Reported Rate(Mm olh <sup>2</sup> /L H)
Direct biophotolysis	Green algae	Yes	H <sub>2</sub> O	H2, O2	Can produce H <sub>2</sub> directly from water and sunlight Solar conversion energy increased by tenfold as compared to trees, crops	Chlamydomonas reinhardtii	0.07
Indirect biophotolysis	Cyanobacteria	Yes	H <sub>2</sub> O	H2, O2	Can produce H <sub>2</sub> from water can fix N <sub>2</sub> from the atmosphere	Anabaena variables	0.36
Photo- fermentation	Phototrophic bacteria	Yes	Organic compounds	H <sub>2</sub> , CO <sub>2</sub>	A wide spectral light energy can be used by these bacteria Can use different waste materials like distillery effluents, waste, etc.	Rhodobacter spheroids	0.16
Dark- fermentation	Fermentative bacteria	No	Organic compounds	H2, O2 VFA	It can produce H <sub>2</sub> all day long without light A variety of carbon sources can be used as substrates It produces valuable metabolites such as butyric, lactic, and acetic acids as byproducts. It is an anaerobic process, so there is no O <sub>2</sub> limitation problem.	Enterobacter cloacae DM 11 Clostridium sp. strain No. 2	75.6 64.5

#### Table 1. Types of biological hydrogen production method.

### 2. Materials and Methods

#### 2.1. Environmental Factors and Sludge

In the experiment, we used sewage sludge collected from a school field (Table 2). The electric current was applied to increase the strength of the hydrogen-producing bacteria and accelerate biochemical metabolic reactions.

Sludge Source	Sewage Treatment Plant				
Substrate concentration	20g COD/L				
Temperature	35°C				
pH	5.5				
RPM	100				
ORP	-300500mV				
Nutrients	Endo formula				
Pre-Treatment	Thermal screening method				
Current strength	20 mA-70 mA				
HRT	6 hr				
Current density	$1.05 - 3.66 \text{ mA/cm}^2$				

Table 2. Conditions of various environmental parameters factors

#### 2.2. Bacteria Screening and Pre-treatment

Pre-treatment and screening of hydrogen-producing bacteria were conducted in an anaerobic environment using the hot sieve method. Anaerobic hydrogen-producing sludge was put in a water bath of boiling water for an hour and cooled to room temperature to inhibit the conversion of methane to hydrogen by fermentation. To evaluate the water quality, composition, and volatile organic acids and alcohols including propionic acid, butyric acid, valeric acid, and ethanol were analyzed using GC-FID (Shimadzu 14A).

### 3. Results and Discussion

#### 3.1. Water Quality in Microbial Hydrogen Production

Water quality affects hydrogen production (Table 3). Hydraulic retention time (HRT) was six hours. Alkalinity, volatile suspended solids (VSS), and chemical oxygen demand (COD) did not significantly change with different currents. pH and carbohydrate degradation changed during the hydrogen production process (Fig. 1). pH maintained between 6.20–6.40, and the acidification response was not significant regardless of the current (Fig. 2). Due to the acetic acid hydrolysis producing carbon dioxide and hydrogen ions on the anode, and the conversion of hydrogen ions into hydrogen gas on the cathode, pH did not change even with acidification.

Current strength (mA)	HRT	Degradation rate of sucrose (%)	Alkalinity (mg/L)	VSS (g/L)	COD (mg/L)	рН
0	6	83.5	1250	8.47	16600	5.50
20	6	81.7	1300	7.78	14000	6.36
30	6	82.4	1300	8.08	15200	6.25
40	6	82.1	1258	8.62	15700	6.25
50	6	63.8	1250	7.46	16200	6.33
60	6	72.5	1300	7.90	15600	6.26
70	6	69.3	1253	8.24	16700	6.32

Table 3. Water quality parameters in different electric currents.



Fig. 1. Effect of convection on sludge.



Fig. 2. Current effect on pH in water.

### 3.2. Current in Hydrogen Production

Electric current affected the production of hydrogen gas (Table 4). Gas production increased as the current increased. The highest hydrogen concentration was observed at the current of 40 mA and decreased while the concentration of nitrogen ( $N_2$ ), methane (CH<sub>4</sub>), and carbon dioxide (CO<sub>2</sub>) increased continuously with the increase in the current. When the current of 20–40 mA, total gas production was doubled(Fig. 3). At the current of 40 mA, hydrogen-producing bacteria were most active. At the same current, electrolytic hydrogen production began.

Current strength (mA)	HRT	H2 (mL)	N2 (mL)	CH4 (mL)	CO2 (mL)	Total gas production amount (mL)	Electrolysis of water production gas amount (mL)	H2 (%)	N2 (%)	CH4 (%)	CO2 (%)
0	6	1590	ND	16	2144	3750	0	42.41	ND	0.42	57.17
20	6	3049	ND	33	4116	7200	0	42.36	ND	0.46	57.18
30	6	3003	ND	40	4156	7200	0	41.72	ND	0.56	57.73
40	6	3030	ND	43	4126	7200	60	42.09	ND	0.61	57.31
50	6	2739	ND	215	4545	7500	100	36.53	ND	2.87	60.60
60	6	2546	ND	133	5120	7800	130	32.65	ND	1.71	65.65
70	6	2583	ND	139	5376	8100	160	31.90	ND	1.72	66.38

Table 4. Microbial production of hydrogen and other gases at different currents.



Fig. 3. Gas products at different currents.

#### 3.3. Products of Microbial Hydrogen Production

In the microbial reaction of hydrogen production, glucose was converted to acetic acid. The maximum hydrogen production was observed for 4 mol  $H_2$ /mol glucose. When the metabolite was butyric acid, the hydrogen production rate was 2 mol  $H_2$ /mol glucose. When using butyric acid, the hydrogen production capacity was limited due to the anaerobic digestion process and methanation. At a current of 20–40 mA, methanation began to increase due to the growth of methane bacteria which used organic acids as a carbon source (Fig. 4). When the current exceeded 40 mA, this advantageous environment was not available and less carbon-organic matter was used. Thereby, the amount of ethanol, butanol, acetic acid, and butyric acid was lowered. The reaction formulae is as follows.

$$C_6H_{12}O_6 + 2H_2O \rightarrow 2CH_3COOH + 4H_2 + 2H_2O$$

$$\tag{1}$$

$$C_6H_{12}O_6 \rightarrow CH_3CH_2CH_2COOH + 2H_2 + 2CO_2$$
(2)

$$CH_3COOH \rightarrow CH_4 + CO_2$$
 (3)



Fig. 4. Liquid and hydeorgen production at different currents.

### 3.4. Impact of Current on Hydrogen Production Bacteria

The bacterial phase was observed using electron microscopy (SEM) (Fig. 5). With electric currents, the bacteria phase changed as shown in Figs. 6 and 7. At a current of 30 mA, the bacteria phase began to lengthen, and at a current of 70 mA, it became shorter. Hydrogen production bacteria showed the highest activities at 20–40 mA, while at 50–70 mA, their activities were retarded.



Fig. 5. Bacteria phase without current.



Fig. 6. Bacteria phase at current of 30 mA.



Fig. 7. Bacteria phase at current of 70 mA.

#### 4. Conclusions

The experiment results showed that the rate of hydrogen concentration was 625 mL/L·hr consisting of 42% of the whole produced gas without electric current. With the current of  $1.05-3.66 \text{ mA/cm}^2$ , the rate increased to 1200-1350 mL/L·hr showing 32-42% of hydrogen gas in the whole gas. In hydrogen production, a positive relationship between electric current and total gas production was observed. At the current of  $2.09 \text{ mA/cm}^2$ , the highest hydrogen production rate was observed. However, the electric current higher than  $2.09 \text{ mA/cm}^2$  slowed hydrogen production. In the normal process of hydrogen production, bacteria conduct an acidification reaction. Therefore, NaOH is necessary to adjust pH. With electric currents applied, pH was maintained at between 6.25-6.35. Although there was an acidification reaction, the acid hydrolysis into CO<sub>2</sub> and hydrogen ions on the cathode occurred, which did not lower pH and NaOH was not used. The results showed that at the current of 30 mA, the bacteria showed longer phases, while at the current of 70 mA, they had shorter ones. The microbial activities of the bacteria reached the maximum at the current of 20-40 mA. At 50-70 mA, the hydrogen production. At the current of  $2.09 \text{ mA/cm}^2$ , complete methanation occurred. When the electric current was higher than  $2.61 \text{ mA/cm}^2$ , the concentrations of acetic acid and butyric acid were lowered, while those of ethanol and butanol increased. The results showed the effect of the electric current on the microbial activity to speed up the metabolic reaction and produce more hydrogen gas.

Author Contributions: C.-F.Huang: Writing—original draft; T.-W.Jang: Formal analysis, Investigation; T.Yin: Data curation; T.Zhang: Software; A.-C.Huang and T.-J.Wan: Supervision, Conceptualization, Methodology. All authors have read and agreed to the published version of the manuscript.

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