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Application of bioelectromethanation using an electroactive methanogen for the biogas upgrading

Jisun Lee†, Jaesung Chun†, Okkyoung Choi* and Byoung-In Sang*
Department of Chemical Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea

Bioelectromethanation was tested using an isolated strain, *Methanothermobacter* sp., for biogas upgrading. The investigated method showed faster bioelectrochemical conversion of carbon dioxide to methane with higher coulombic efficiency than previously reported without additional hydrogen and mediator supplementation. Bioelectromethanation can utilize carbon dioxide, unlike gas separation methods, and actually requires a less negative potential than in water electrolysis. The isolated methanogens showed a relatively fast conversion to methane compared to the previously reported methane production rate and current intensity. Through further research on electroactive methanogens and the development of operation technology, bioelectromethanation can be applied for biogas upgrading with a lower energy requirement but without CO_2 emissions.

Keywords: Bioelectrochemical conversion, Electrogenotrophic methanogen, Biogas upgrading, Methanation, CO_2 utilization.

Introduction

Efforts to reduce carbon dioxide (CO_2) emissions are in progress worldwide in response to climate change issues. Carbon capture and utilization (CCU) is an effective technology for reducing CO_2 emissions. The reuse of CO_2 would reduce the need for CO_2 extraction from natural sediment resources such as petroleum and enhance the circular CO_2 economy [1]. Methane (CH_4) is a reduced chemical for CO_2 utilization and can be used as a substitute for natural gases (95% methane, 5% ethane) to generate electricity and make energy [2]. Highly concentrated methane can be injected directly into a prevailing natural gas network to efficiently store energy when needed and can stabilize renewable energy supply systems with intermittent supply characteristics [3, 4]. Just as natural gas is used as a vehicle fuel, a mixture with more than 95% methane can be used as a transportation fuel [5-7]. In addition, methane is likely to be used as a multipurpose chemical building block to make more value-added materials [8-11].

Biogas from anaerobic digestion contains approximately 40% CO_2 and 60% CH_4 [12]. Recently, biogas upgrading technology has attracted attention for CO_2 utilization and waste-to-energy applications beyond the concept of gas purification, such as absorption, adsorption, and membrane separation [13, 14]. The reduction of CO_2 requires reducing agents, generally hydrogen. To convert CO_2 to CH_4, the amount of hydrogen needed is 4 times the amount of CO_2.

$$2H^+ + 2e^- \rightarrow H_2 \quad E = -0.414 \text{ V vs NHE} \tag{1}$$

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \tag{2}$$

Electrical energy made from renewable energy, such as solar and wind, can be converted into chemicals such as hydrogen through water hydrolysis. The electrical power equivalent of hydrogen (lower heating value) is 33.3 kWh/kg H_2 [15]. Methanation eq. (2) can be carried out on both chemical and biological catalysts. Hydrogenotrophic methanogens are used as a biological catalyst [16-18].

Several studies have suggested the bioelectromethanation (bioelectrochemical) technique for biogas upgrading [19-23]. The interaction between the microbes and cathode determines the product.

$$CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \quad E = -0.244 \text{ V vs. NHE} \tag{3}$$

In electromethanogenesis, electrogenotrophic methanogens can directly use electrons from the cathode [22]. Bioelectromethanation is operated at a less negative potential (-0.244 V), but the methane production is much lower than those obtained with hydrogenotrophic methanogens.

This study used the recently isolated strain *Methanothermobacter marburgensis,* which is strictly hydrogenotrophic and thermophilic, to investigate its potential for ex situ bioelectrochemical biogas upgrading. This study showed the bioelectrochemical conversion from CO_2 to CH_4 by one isolated electroactive methanogen, evaluated of pH effect on methane production, and suggested the application of bioelectromethantion for biogas upgrading based on faster conversion rate than

*Corresponding author:
Tel: +82-2-2220-4717, Fax: +82-2-2220-4716
E-mail: okgii77@hanyang.ac.kr (Okkyoung Choi)
Tel: +82-2-2220-2328, Fax: +82-2-2220-4716
E-mail: biosang@hanyang.ac.kr (Byoung-In Sang)
†The first two authors listed share first authorship.

previously reported.

Methods and Materials

Inoculums

The hydrogenotrophic and thermophilic methanogen denoted as *Methanothermobacter* sp. THM-2 (NCBI taxonomy ID 2606912), which was isolated from a thermophilic anaerobic digester and is phylogenetically related to *Methanothermobacter marburgensis* DSM 2133T, was used as the inoculum. The preculture was performed in a 1 L bottle at 60 ºC with 200 mL ATCC medium containing (per liter) K_2HPO_4 2.04 g, KH_2PO_4 1 g, NH_4Cl 1 g, NaCl 1 g, $MgCl_2 \cdot 6\ H_2O$ 0.1 g, $CaCl_2 \cdot 2H_2O$ 0.06 g, $NaHCO_3$ 4 g, resazurin (0.1% w/v) 0.5 mL and trace elements 10 mL. The trace element solution contained (per liter) nitrilotriacetic acid 12.8 g, $FeSO_4 \cdot 7H_2O$, $MnSO_4 \cdot H_2O$ 0.085 g, $CaCl_2 \cdot 2H_2O$ 0.1 g, $ZnCl_2$ 0.1 g, H_3BO_3 0.01 g, NaCl 1 g, $NiCl_2 \cdot 6H_2O$ 0.15 g, $Al(SO_4)_2 \cdot 12H_2O$ 0.098 g, $CoCl_2 \cdot 6H_2O$ 0.024 g, $CuCl_2 \cdot 2H_2O$ 0.025 g, $Na_2MoO_4 \cdot 2H_2O$ 0.024 g, $NaSeO_4$ 0.026 g, and $Na_2WO_4 \cdot 2H_2O$ 0.25 g. The medium was flushed with Ar gas for at least 30 min, and after autoclaving at 121 ºC for 20 min, $Na_2S \cdot 9H_2O$ (0.5 g/L) was added. During the preculture, the headspace of the bottle was exchanged with CO_2/H_2 (20/80) daily. The preculture was operated for 3 weeks, vigorously mixing between gas-liquid at 500 rpm using magnetic stirring.

Bioelectrochemical system (BES) start up and operation

Graphite felt (6 cm×19.5 cm×0.5 cm) was used as the cathode and anode and was inserted into the reactor in tubular form (area, 253.5 cm^2). A three-electrode system (Fig. 1) was used, and a 3 M KCl saturated Ag/AgCl electrode (+199 mV vs. SHE) was used as a reference electrode. The electrode was attached to a titanium wire (1 mm in diameter) using carbon paste and connected to a potentiostat (WMPG1000, South Korea). The two-chamber reactor consisted of cathode and anode compartments and had a 4-port cap sealed with butyl rubber on the side. The cathode and anode were separated by a proton exchange membrane (Nafionâ 117, Du Pont). A pH meter (InLab Easy BNC, Mettler Toledo, USA) was inserted into the cathode compartment. The volume of each chamber was 1.4 L, the headspace was 0.7 L, and the working volume was 0.7 L. After assembly, the reactor with the electrode was autoclaved at 121 ºC for 20 min. After anaerobic treatment with 100% CO_2 gas purging, 200 mL of *Methanothermobacter* sp. THM-2 cells were inoculated, and the initial optical density at 600 nm was approximately 4. Gas exchange was carried out once a day with CO_2. To control the pH, 3.5 N H_2SO_4 and ammonia solution (28% NH_3 in H_2O) were used. The experiment was performed at potentials of -0.85, -0.90 and -0.95 V vs Ag/AgCl and different pH conditions (pH 6.9 ± 0.1, pH 6.5 ± 0.1).

Analysis and calculations

The gas (CO_2, H_2, or CH_4) from the cathode was analyzed using GC-TCD (gas chromatography-thermal conductivity detector, 7890A, Agilent, USA) with a Porapak Q Column (Supelco, Inc, 6 ft × 1/8 in, SS, 80/100 mes), the oven temperature ranged from 40 to 80 ºC (10 ºC/min), the injector temperature was 100 ºC, and the detector temperature was 200 ºC. The ammonium

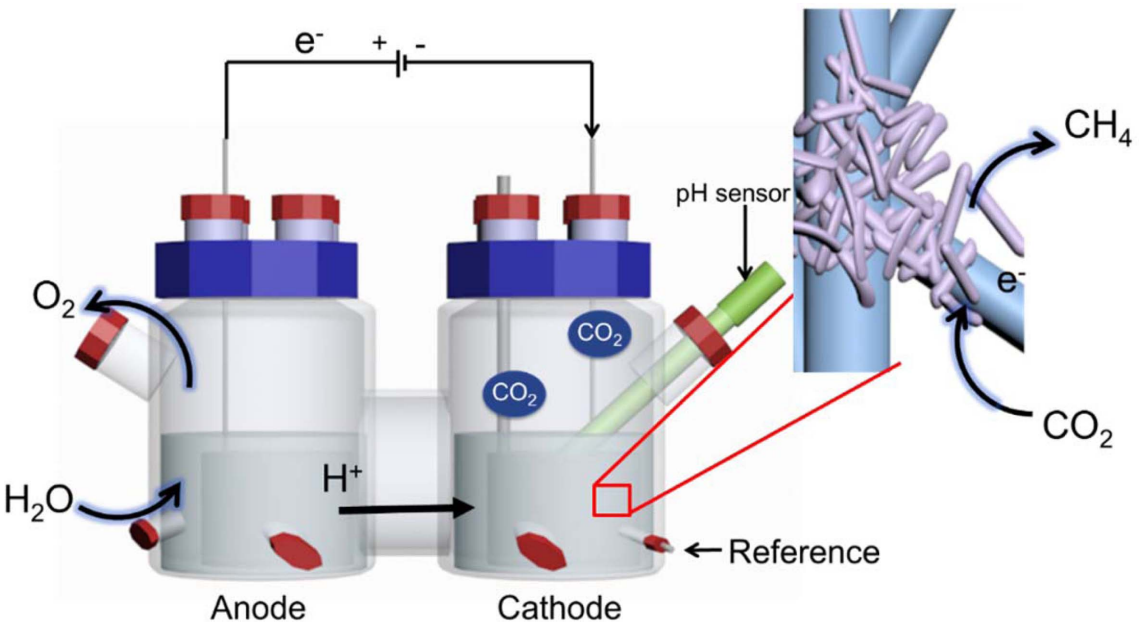

Fig. 1. Bioelectromethanation reactor. The insert shows a schematic of the expected bioelectrochemical reaction at the interface of the electrode and hydrogenotrophic methanogens.

ion (NH_4^+) content was measured using a RQflex 10 reflectometer (Merck, Germany) and a Reflectoquant® Ammonium Test (Merck, Germany) kit. Coulombic efficiency was calculated using the ratio of the electron number for the formation of the product to current consumption.

$$\text{Coulombic efficiency CE, \%} = \frac{mnF}{\int_0^t Idt} \quad (4)$$

where m is the number of moles of methane generated, n is the number of electrons required for the formation of methane (8 eq./mol CH_4, eq. (3)), F is the Faraday constant (96,485 C/mol of electrons), and I is the circuit current at time t.

Results and Discussion

Bioelectrochemical conversion of CO_2 to CH_4

After inoculation into the bioelectrochemical (BES) reactor, the CO_2 conversion to methane was detected. Under abiotic conditions, methane production and current consumption were not observed (data not shown). The methane production per unit working volume was 0.05~0.23 L/L/d at pH 6.5 but 0.006~0.08 L/L/d at pH 6.9 and was enhanced at lower pH (Fig. 2). The methane production per unit cathode surface area was 6.36 L/m^2/d at -0.95 V vs. Ag/AgCl and pH 6.5 (Fig. 2). Studies on the bioelectrochemical conversion of CO_2 reported methane production rates of 1 L/m^2/d (273.15 K, 1 atm) in a pure culture of *Methanothermobacter thermoautotrophicus* [24] and 0.26 L/m^2/d with *Methanobacterium palustre* [25] (Table 2). In general, pure methanogen culture applications of BES resulted in lower reported methane production rates than mixed cultures, such as anaerobic sludge, but this study showed a high methane production rate similar to that of mixed cultures, reported as 1.5~9.8 L/m^2/d [26].

The optical density (600 nm) was approximately 4 at the initial time, and the value decreased slightly but did not change significantly (data not shown). Methanogens are sensitive against acidification, and the optimal pH range was previously reported to be pH 6.5~8.0 [27]. The pH effect was tested at pH 6.5 vs. pH 6.9. Interestingly, the bioelectrochemical conversion of CO_2 to methane was improved at pH 6.5 compared with that at pH 6.9 (Fig. 2 and Table 1). The pH difference was small, but the methane production was significantly different. Even at the same voltage, the bioelectrochemical conversion of CO_2 to methane was higher at pH 6.5 than at pH 6.9 (Fig. 2). There have been no studies about the pH effect on BES in pure cultures of hydrogenotrophic methanogens. The reason why methane production increased at lower pH is still unknown, but it is speculated that electron transfer between the electrode and methanogens is more active at pH 6.5 than at pH 6.9. As a follow-up study, we plan to further characterize the isolated electrotrophic methanogen strain to determine why these results were observed and develop operating conditions for increased methane production.

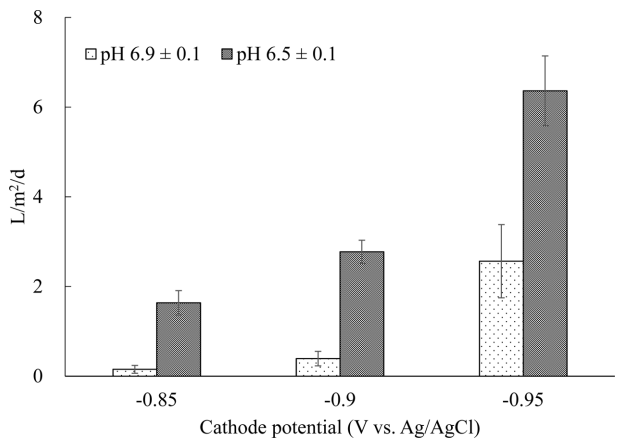

Fig. 2. pH-dependent methane production in the bioelectrochemical system.

Coulombic efficiency

In addition, the average current density was 0.06~1.7 A/m^2 (Table 1). This current density was also higher than that previously reported for pure culture

Table 1. The experimental conditions at various potentials and pH values

Potential (V vs. Ag/AgCl)	pH	Average current density (A/m^2)	Maximum coulombic efficiency (%)
-0.85 V	pH 6.9 ± 0.1	0.061 (± 0.006)	89.1
	pH 6.5 ± 0.1	0.36 (± 0.04)	135.0
-0.9 V	pH 6.9 ± 0.1	0.098 (± 0.008)	125.5
	pH 6.5 ± 0.1	0.8 (± 0.1)	115.1
-0.95 V	pH 6.9 ± 0.1	0.68 (± 0.15)	101.0
	pH 6.5 ± 0.1	1.7 (± 0.2)	117.9

Table 2. The comparison of methane production rate and coulomb efficiency reported previously

Electroactive methanogen	Methane production rate (L/m^2/d at STP)	Coulomb efficiency (%)	Reference
Methanobacterium-like archaeon strain IM1	0.08	80	Beese-Vasbender *et al.* [36]
Methanobacterium palustre	0.26		Chen *et al.* [25]
Methanothermobacter thermoautotrophicus	1	96	Sato *et al.* [24]
Methanothermobacter thermautotrophicus strain ΔH	2.7	20	Hara *et al.* [37]
Methanothermobacter sp. THM-2	6.36	100	This study

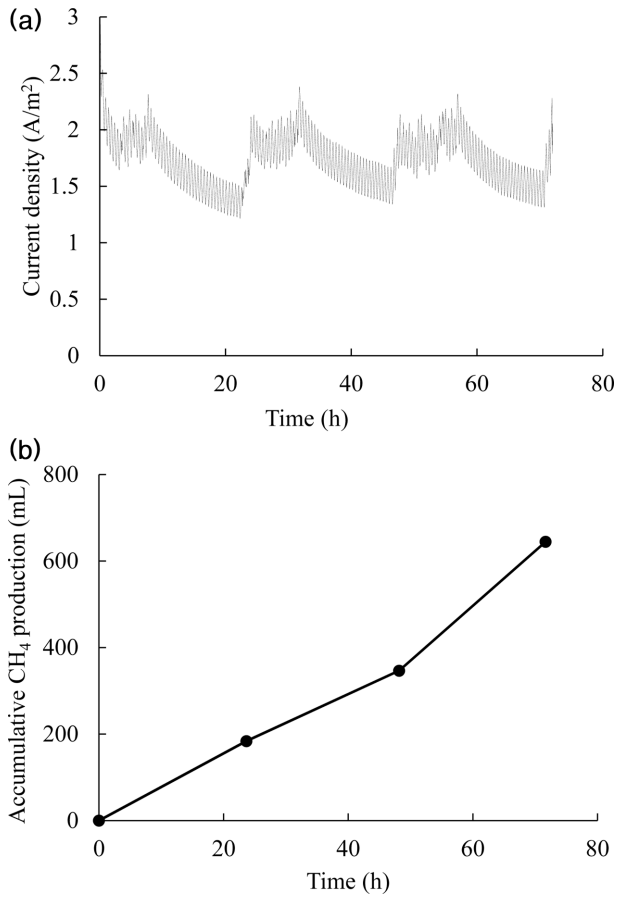

Fig. 3. Current density (a) and cumulative methane production (b) obtained when operating the BES at -0.95 V vs. Ag/AgCl and pH 6.5. CO_2 charging into the bioelectrochemical reactor was performed three times at the times of current decrease.

methanogens, < 0.5 A/m^2 [28]. Fig. 3 shows the current density and methane accumulation over time at -0.95 V vs Ag/AgCl and pH 6.5. After CO_2 injection, the current increased rapidly but decreased with CO_2 consumption. Under all conditions, the amount of methane produced was found to be linearly related to current consumption, and the ratio of approximately 1 in Fig. 4 indicates nearly 100% coulombic efficiency (eq. (4)). High methane production and current density increased the coulombic efficiency to near 100%; the slope value is 1.1 in Fig. 4. According to past research data, the reported highest level of coulombic efficiency for methane, CH_4, is approximately 64% when using a copper single-crystal catalyst [29]. The abiotic electrochemical reduction of CO_2 exhibits a lower energy efficiency on metal electrodes [30]. In a bioelectrochemical system, the reported coulombic efficiencies of a mixed culture and pure culture were 82.6±15.7% and 55±27%, respectively, and the mixed cathodic culture showed higher methane production [26]. However, in the case of microbial fuel cells that were studied much earlier, a pure inoculum with defined electroactive microorganisms showed higher efficiency [31]. Our study suggests the possibility of better efficiency when using a pure culture than

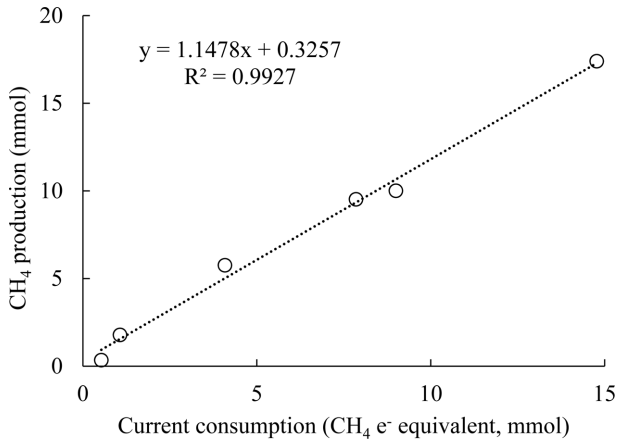

Fig. 4. Methane production with respect to current consumption. The current consumption is indicated in methane electron equivalents, i.e., the number of moles of electrons divided by 8 (eq. (2)).

mixed cultures in the cathode after operational parameter development.

In general, the higher the voltage is, the higher the methane production [26]. A more negative potential increases the hydrogen production when converting CO_2 to CH_4, i.e., indirect electron transfer occurs using H_2 as an electron carrier. At -0.95 V vs. Ag/AgCl, the amount of hydrogen detected was only ~5.5% of the CH_4 amount. Because 4 H_2 molecules are needed for 1 CH_4 molecule, the electron number required for 1 CH_4 generation is the same in both direct (8 e^-) and indirect electron transfer (4 × 2 e^- = 8 e^-). Additionally, the current consumption required for methane production is the same if both the direct (electron) and indirect (H_2) processes have the same efficiency. The observed high coulombic efficiency at -0.95 V vs. Ag/AgCl indicates efficient electron transfer at the given potential.

Application of bioelectrochemical conversion for biogas upgrading

This study shows the possibility of applying the bioelectrochemical conversion of CO_2 to CH_4 using an electroactive methanogen for biogas upgrading. Despite having the advantage of operating at less negative voltages (eq. (1) vs. eq. (3)), the slow methane production rate and low current densities of the bioelectrochemical system [19, 28] are the main reasons making this system unsuitable for real methanation applications. Nevertheless, the methane production rate is much lower than that of biomethanation using thermophilic hydrogenotrophic methanogens. The highest methane production rate reported was 288 L/L/d under atmospheric pressure [17]. Bioelectrochemical conversion occurs at the surface of the electrode, and the methane production rate is indicated per unit surface area, not volume. Therefore, it is difficult to directly compare bioelectromethanation with biomethanation. In the bioelectromethanation approach in this study, the rate of methane

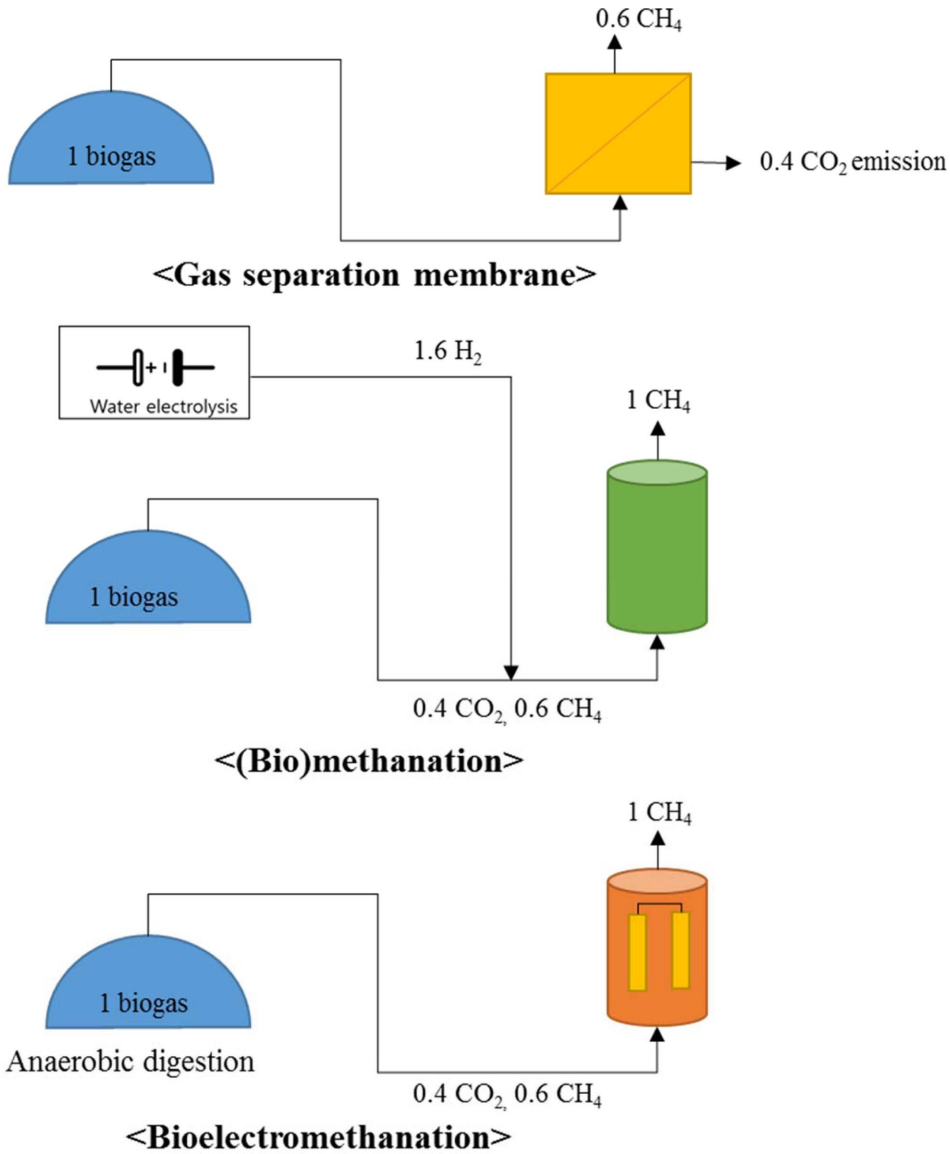

Fig. 5. A comparison of gas separation, biochemical methanation and bioelectromethanation as biogas upgrading methods. Bioelectromethanation can convert CO_2 to CH_4 directly using electricity.

Table 3. Biogas upgrading technology comparison

Biogas upgrading methods	Gas separation membrane	(Bio)methantion	Bioelectromethanation
CO_2 emission	High	Low	Low
Pressure (bar)	5-10	(1-10) 50-200	1
Process specifics	CO_2 adsorption and inevitable CO_2 generation	Requires a reducing agent such as H_2, which can be supplied through water electrolysis	Bioelectrochemical conversion by microbe-electrode interaction
Hydrogen	No hydrogen required	Hydrogen required	No hydrogen required

production per electrode surface was 6.4 (±0.8) L/m^2/d, and the rate per working volume was 0.23 (±0.03) L/L/d at -0.95 V vs. Ag/AgCl and pH 6.5. A low conversion rate is difficult to apply to processes with a high gas hourly space velocity (GHSV) treating a large amount of CO_2. The typical biogas generation rate is 0.3-0.5 L/L/d, and biogas contains approximately 40% CO_2 [32]. Based on previous data, the amount of CO_2 treated for conversion to CH_4 is 0.1-0.2 L/L/d. Therefore, we propose a biogas system combined with bioelectromethanation for biogas upgrading that does not require a fast conversion rate (Fig. 5). Table 3 shows characteristics of three biogas upgrading technology. Biogas upgrading using a gas separation system emits CO_2 and produces a relatively small amount of methane (Fig. 5(a)). The theoretical external voltage of the cathode

for bioelectromethanation (eq. (3)) is less than that for hydrogen production (eq. (1) and Fig. 5(b)) in water electrolysis [21, 24]. Additionally, there is no need for an additional reactor that produces a reducing agent, such as H_2 (Fig. 4(b)), because electricity supply and conversion take place in one reactor (Fig. 5(c)). The substrate for bioelectromethanation is only CO_2, and a pure culture can prevent competition between methanogens (-0.24 V vs. Ag/AgCl) and acetogens (-0.29 V vs. Ag/AgCl) [33, 34]. Because autotrophic methanogens are used, no organic carbon is required, and only ammonia (nitrogen source), sulfide (enzyme precursor), and trace metal (enzyme cofactor) are needed for bioelectromethanation. Therefore, bioelectromethanation is relatively easy to apply as a module in anaerobic digesters (Fig. 5(c)). The bioelectromethantion can be simply installed as a module to increase methane concentration of biogas. Based on our conversion rate, bioelectromethanation can be applied for biogas upgrading (~0.02 GHSV, gas hourly space velocity) requiring lower GHSV than biological methanation (~100 GHSV) or chemical methanation (500~5,000 GHSV) [35].

Conclusions

This study showed fast conversion rate of CO_2 to CH_4 by pure methanogen-electrode reaction, up to the level applicable to actual process and it represented that it can be applied to actual processes such as biogas upgrading system. Bioelectromethanation has the advantage of not requiring expensive hydrogen, but it has been reported that the reaction rate and efficiency was low for practical application. However, our results suggest that a pure electrogenotrophic methanogen can be used for bioelectromethanation, especially biogas upgrading, with relatively faster conversion rate than previously reported value. This approach allows for sustainable energy systems in anaerobic digestion plants, and the environmental incentives are expected to evolve.

Acknowledgments

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Conflicts of Interest

The authors declare no conflict of interest.

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