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Conversion of CO_2 to CH_4 by a Pulsed Hydrogen Plasma Shower Method

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Authors' contributions

The experiments were performed in collaboration with both authors. The data were analyzed by author KA under discussions with author SI. Both authors were approved the final manuscript.

Article Information

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Original Research Article

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ABSTRACT

Aims: To suppress the emission of CO_2 to the environment and to save the consumption of fossil fuels, CO_2 was converted to CH_4 by a newly developed hydrogen shower method with a hydrogen pulse plasma.
Study Design: Research study.
Place and Duration of Study: This study was performed for 2013 - 2015 at Department of Electrical Engineering, Tohoku University, Sendai, Miyagi, Japan.
Methodology: The experiment was carried out in a small chamber which was divided into two parts by an orifice disc of 3-mm-thickness stainless plate with one 0.5-mm-diameter hole at the center. Hydrogen gas was supplied from the left part, where hydrogen radicals of H* and H_2* were produced by a pulse discharge. Hydrogen radicals were supplied through the orifice from the left part to the right part as a hydrogen radical shower. Carbon dioxide was directly supplied to the right reaction part, where CO_2 was able to collide with hydrogen radicals and as a result CH_4 was produced.
Results: Dependences of CO_2 decomposition ratio α, methane selectivity β, and energy efficiency γ on hydrogen flow rate, electrode distance, discharge tube diameter, applied voltage, electrode diameter, and gas feeding type were investigated. Methane was produced from carbon dioxide by

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using a hydrogen radical shower method. Methane was only organic species produced from CO_2. Only CO was detected as non-organic by-product. It was found that the decomposition ratio α, methane selectivity β, and energy efficiency γ were α = 32%, β = 37%, and γ = 1.6 L/kWh, respectively, under optimized condition at the flow rate ratio of CO_2: H_2 = 1:2, gap distance of d = 6 mm, and input power of P_{in} = 4.6 W (1.2 kV, 3.8 mA) with a use of 6-mm-diameter electrode.

Conclusion: Energy efficiency in our case was fairly improved. Hydrogen radical shower method was very effective for the conversion of CO_2 to CH_4.

Keywords: Carbon dioxide; methane; hydrogen radical shower; pulse discharge.

1. INTRODUCTION

Carbon dioxide CO_2 is one of the man-made greenhouse gases that are emitted by combustion of fossil fuels, such as coal, oil, and natural gas. Carbon dioxide is emitted from many power plants for generating electricity, power vehicles, heat homes, cook food and much more. However, fossil fuels are essentially a non-renewable energy source. Within the next 100 years it is widely believed that the cost of finding and extracting new underground resources will be much more expensive for everyday use. It might be also serious that CO_2 would cause global warming by absorbing and emitting radiation within the infrared range.

Therefore, the suppression of emission of carbon dioxide into the environment and the reduction of consumption of fossil fuels are crucial subject that must be settled urgently.

In order to suppress the emission of CO_2 into the environment from electrical power plants, for example, it might be desirable that CO_2 is collected before exhausting to convert it to methane, if any surplus electric power exists. This means that surplus electric energy can be converted to chemical bonding energy of methane. That is, the surplus electric energy can be stored as methane [1]. This method is superior to batteries, because the electric energy stored in batteries will be gradually lost by a natural discharge. On the contrary, the energy stored in methane will be conserved without any loss for many years.

In order to reduce CO_2 with hydrogen various experiments were carried out by using discharge system [2-9]. In most cases, CO_2 was reduced by CH_4 to form syngas of CO and H_2, because methane is also one of the greenhouse gases [10-16]. Eliasson et al. [2] investigated the production of CH_4 by a dielectric barrier discharge with H_2 in detail. Mixed gas of CO_2 and H_2 was employed for CH_4 production. However, for an efficient formation of methane a new innovative method has been expected.

The purpose of this study is to investigate fundamental process of the reduction of carbon dioxide by hydrogen radicals that were produced in H_2 discharge. Hydrogen radical shower was supplied to the plasma-free downstream reaction space where CO_2 was supplied. Since CH_4 production region was separated spatially from H_2 discharge region, a preferable conversion rate was expected, because deformation of CH_4 was able to be avoided in the plasma-free reaction space. Our method proposed here is quite unique to generate beneficial and reusable organic materials like methane by using low-pressure hydrogen discharges [17-19].

2. EXPERIMENTAL DETAILS

Schematic of the experimental apparatus is shown in Fig. 1. The chamber was divided into two parts by an orifice disc of 3-mm-thickness stainless plate with one 0.5-mm-diameter hole at the center. The left part was a hydrogen plasma source for the hydrogen radical production, consisting of a glass tube of 4 mm in inner diameter. Hydrogen gas was supplied from the left side into the plasma source region. The right part was a narrow reaction space consisting of a glass tube of 10 mm in inner diameter, terminated by double glass tubes, consisting of an inner glass tube of 4 mm in outer diameter and 2 mm in inner diameter, and an outer glass tube of 10 mm in outer diameter and 8 mm in inner diameter. Axial length of the reaction space between the orifice plate and the end of the double tube can be varied from 3 mm to 10 mm. Usually, it was set at 5 mm. There was no discharge in the reaction space. Carbon dioxide was fed directly into the reaction space through an inner tube of the double glass tubes from the right side. A stainless rod electrode of 1 mm in diameter was inserted into the glass tube from the left side. Hydrogen plasma was produced between the tip of the electrode and the metal orifice plate grounded electrically. Here, hydrogen radicals of H* and H_2* were produced through the following reactions.

$$e + H_2 \rightarrow H_2^*, \quad (1)$$

$$e + H_2 \rightarrow H^* + H^* \quad (2)$$

These radicals were injected into the reaction space through the orifice hole as a hydrogen radical shower. In this way, CO_2 was able to collide with H* and H_2* radicals in the reaction space and as a result CH_4 was produced. The gas produced was evacuated by a rotary pump through a circumferential gap between the outer and inner tubes of the double glass tube. Gas flow directions are indicated by arrows in Fig. 1.

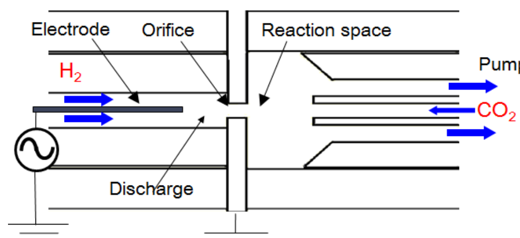

Fig. 1. Schematic of the experimental setup

The gas flow rate ratio of carbon dioxide to hydrogen and the total gas flow rate were controlled by mass flow controllers, independently. Total pressure was fixed at 200 Pa. Here, we employed a negative square-pulse voltage that was supplied to a small electrode. Pulse duration was fixed at 5 μs. Repetition frequency of the square pulse was also fixed at 7.8 kHz. The gas after passing through the discharge region was sampled and analyzed by Fourier transform infrared spectroscopy (FTIR) by comparing the gas species before and after the discharge [17-19].

3. RESULTS

The results were evaluated by the following quantities.

(i) CO_2 decomposition ratio α:

$$\alpha = 1 - [CO_2]_1 / [CO_2]_0 \quad (3)$$

(ii) CH_4 selectivity β:

$$\beta = [CH_4] / [\text{all carbon species produced}] \quad (4)$$

(iii) Energy efficiency γ (L/kWh) for CH_4 production:

$$\gamma = [CH_4 \text{ produced in litter}] / (\text{electric energy consumed by the discharge}) \quad (5)$$

Here, [x] denotes amount of x, and suffix 0 and 1 correspond to the values before and after the discharge, respectively. These quantities show how much carbon in CO_2 has been converted to methane. γ is an important factor to realize a suitable commercial system for producing methane in high efficiency. Using α and β, γ can be expressed as follows.

$$\gamma = \alpha\beta\Gamma / (\text{electric power for the discharge (W)}) \times 60 \quad (6)$$

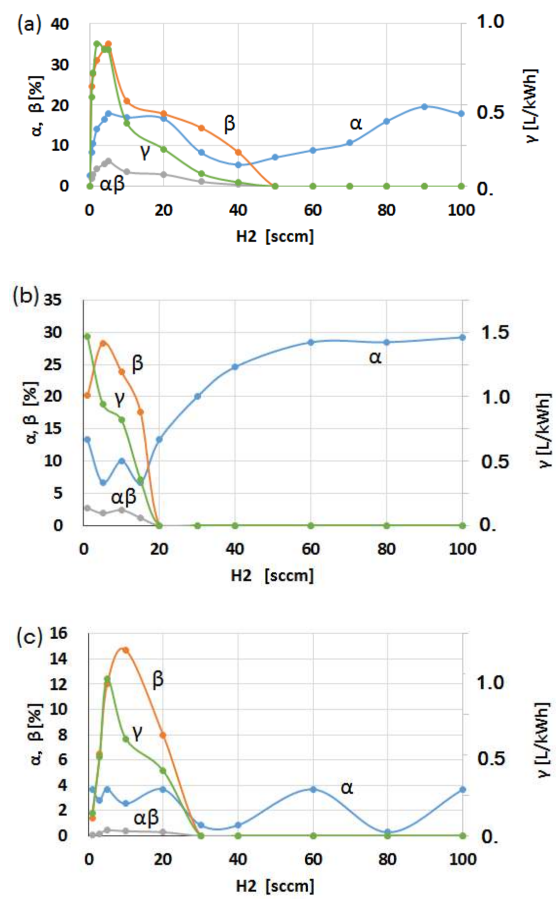

Fig. 2. Variations of α, β, αβ, γ as a function of H_2 flow rate in cases of CO_2 = (a) 1 sccm, (b) 5 sccm, and (c) 20 sccm in a 4-mm-diameter glass tube. Electrode distance is d = 2 mm

Here, Γ is initial gas flow rate of CO_2 [scc/min; standard cubic centimeter per minute]. The change of gas species measured by FTIR showed that main carbon products were CH_4 and CO through the whole experiment. Here, CO might come from the following dissociation reaction by hydrogen radicals in the reaction space.

$$CO_2 + H_2^* \rightarrow CO + H_2O \quad (7)$$

Hydrocarbon species was only CH_4, and the other species like HCOH and CH_3OH were not detected and/or were negligibly small. We could not detect other C_2 organic materials such as ethane, ethylene, and acetylene. But, the production of steam H_2O was detected. Therefore, it was shown that methane was only a hydrocarbon produced from CO_2 in this system. Therefore, in this case, the reaction was rather simple and the methane selectivity β could be simply expressed by $\beta = [CH_4] / ([CH_4]+[CO])$.

3.1 H_2 Flow Rate Dependence

First, dependences of CO_2 decomposition ratio α, methane selectivity β, product αβ, and energy efficiency γ on hydrogen flow rate were shown in Fig. 2 with CO_2 flow rate as a parameter. Here, electrode diameter was 1 mm, electrode distance was d = 2 mm and the applied voltage was 1.25 kV under the total pressure of 200 Pa. The discharge took place in a glass tube of 4 mm in inner diameter.

When CO_2 flow rate is 1 sccm, α increases with H_2 flow rate and attained to a broad maximum of 17 – 18% in the range of H_2 = 5 – 20 sccm. Then, α decreases with H_2 and eventually increased again to about 20% as shown in Fig. 2(a). The variation of β was similar to that of α in the range H_2 < 5 sccm. However, β was simply decreased to zero in the range of H_2 =10 – 50 sccm. No methane was produced in the range H_2 > 50 sccm. The maximum of β was about 35% in this case. Then, the maximum of the product αβ was about 6.3% at H_2 = 5 sccm. That is, 6.3% of CO_2 was converted to CH_4. The energy efficiency for methane production was also varied like αβ and its maximum attained was 0.9 L/kWh at H_2 = 5 sccm.

The properties described above were not much changed when CO_2 flow rate was increased to 5 sccm and 20 sccm, as shown in Figs. 2(b) and (c), respectively. Methane production was observed in a limited range of H_2 < 20 sccm and < 30 sccm in the cases of CO_2 = 5 sccm and 20 sccm, respectively. The maximum of α in this range was 14% in the case of CO_2 = 5 sccm, and 4% in the case of CO_2 = 20 sccm. On the other hand, the maximum β was 28.5% in the case of CO_2 = 5 sccm, and 14.8% in the case of CO_2 = 20 sccm. These values were smaller than 35% t in the case of CO_2 = 1 sccm. The product αβ was also decreased with an increase of CO_2 flow rate. We got maximum αβ = 2.5% and 0.08% in the cases of CO_2 = 5 sccm and 20 sccm, respectively. The variation of energy efficiency γ was not so simple. We got γ of 1.0 – 1.5 L/kWh in the cases of CO_2 = 5 – 20 sccm.

3.2 Electrode Distance Dependence

The discharge took place under the condition with applied voltage of 1.25 kV and total pressure of 200 Pa. Fig. 3 shows variations of α, β, αβ, and γ as a function of the electrode distance d. Decomposition of 40% was obtained when the electrode distance d was 2-3 mm. However, α was decreased with an increase of d (> 4 mm). On the contrary, CH_4 selectivity β was increased with an increase of d (> 4 mm).

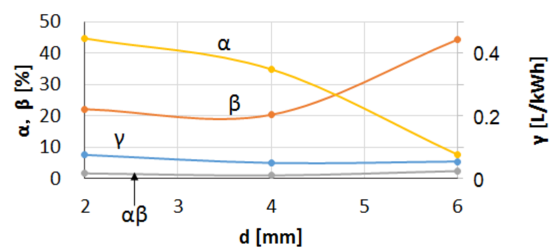

Fig. 3. Variations of α, β, αβ, γ as a function of electrode distance d in case of CO_2/H_2 = 1 sccm/10 sccm in a 4-mm-diameter glass tube

When d was short, the density of hydrogen plasma increases by an increase of the strength of electric field between the electrode and the orifice. Then, such increase of hydrogen radical density might give rise to an enhancement of CO_2 decomposition. On the contrary, with an increase in the electrode distance d, β became relatively high. This might be due to that methane synthesis was proceeded with a relatively low density H_2^* radicals, where decomposition of CO_2 by H_2^* collision was reduced. These different dependency of α and β on d for d > 4 mm was almost cancelled for d > 4 mm, then the change of αβ was very small with an increase of d. Therefore, the energy efficiency γ was also not much changed by the electrode distance d.

3.3 Effect of Discharge Tube Diameter

As shown in Fig. 3, the methane selectivity β increased with the discharge length, although CO_2 decomposition was decreased. In order to clarify the effect of the discharge volume on the methane production, the inner diameter of glass tube in the hydrogen discharge region was changed from 4 mm to 6 mm under the fixed discharge length at d = 6 mm. Fig. 4 shows the variations of α, β, and γ as a function of hydrogen flow rate. Here, the electrode diameter was 1 mm and CO_2 flow rate was 1 sccm. As shown in Fig. 4, CO_2 decomposition α was 15% for H_2 flow rate < 5 sccm, then α was decreased to about 10% with an increase of H_2. Finally, α became almost constant for H_2 > 10 sccm. Methane selectivity β was increased with an increase of H_2 flow rate, and attained to a maximum value of 54% when H_2= 20 sccm. Energy efficiency γ was also increased with H_2 and attained to the maximum of 0.45 L/kWh at H_2 = 5 sccm. By comparing these values with the results in Fig. 3 for the discharge with d = 6 mm in the 4-mm-diameter tube, it was found that the discharge tube diameter was not so important for the improvement of the parameters α and β. In both cases, we got α ~ 9% and β ~ 43%.

3.4 Effect of Applied Voltage

Dependence of α, β, αβ, and γ on the applied voltage is shown in Fig. 5. Here, CO_2 flow rate was 1 sccm and H_2 flow rate was 5 sccm. We got a large β when higher voltage was applied to the electrode. On the contrary, α was decreased in the higher applied voltage regime. Eventually, the product αβ was saturated at around 10% for the applied voltage > 1.0 kV. On the other hand, γ became maximum with a decrease of the applied voltage. We got 1.4 L/kWh when the applied voltage was 1.0 kV.

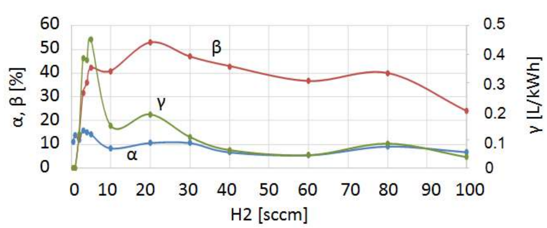

Fig. 4. Variations of α, β, and γ as a function of H_2 flow rate in case of CO_2 sccm in a 6-mm-diameter glass tube. Electrode distance d = 6 mm

3.5 Effect of Electrode Diameter

Finally, the diameter of the electrode for the hydrogen discharge was changed in the 6-mm-diameter glass tube. Dependence of α, β, and γ on hydrogen flow rate is shown in Fig. 6(a), (b), and (c) for the electrode diameter of 1mm, 3 mm, and 5 mm, respectively. We found that α, β, and γ were dependent on the diameter of the discharge electrode. For a small diameter electrode methane selectivity β became large. The maximum of β was about 54% at H_2 flow rate of 20 sccm. However, β was slightly decreased to 42% at H_2 = 5 sccm. On the other hand, the maximum of CO_2 decomposition rate α was about 15% at H_2 flow rate of 5 sccm as shown in Fig. 6(a).

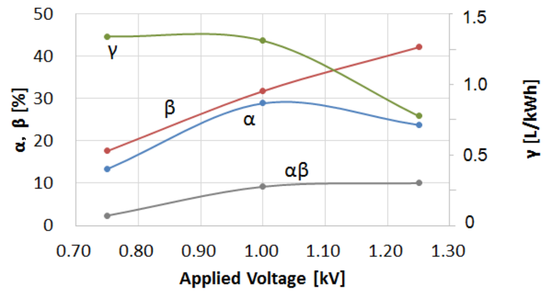

Fig. 5. Variations of α, β, αβ, and γ as a function of applied voltage in a 6-mm-diameter glass tube. CO_2/H_2 = 1 sccm/5 sccm

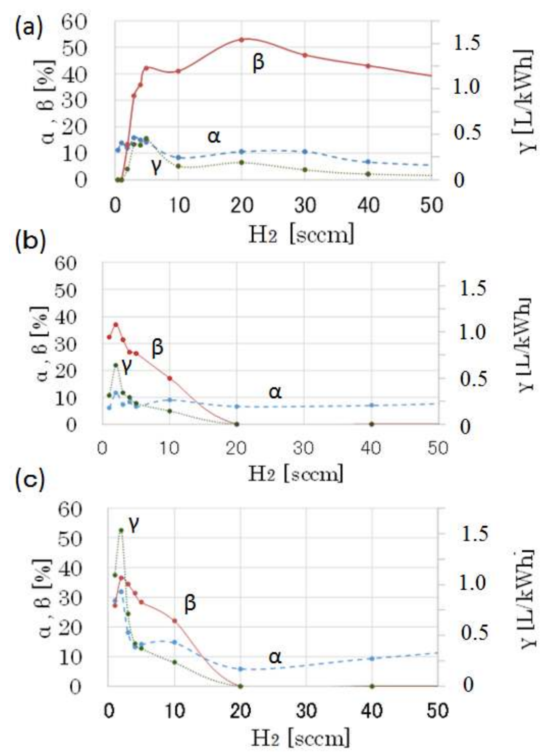

Fig. 6. Variations of α, β, and γ as a function of hydrogen flow rate in three different cases of electrode diameters. (a) 1 mm, (b) 3 mm, and (c) 5 mm in a 6-mm-diameter glass tube. Applied voltage is 1.25 kV. CO_2 = 1 sccm

In the cases of the electrode diameter of 3 mm and 5 mm, the maximum of α and β took place for a smaller H_2 flow rate regime around H_2 = 2 sccm. A big difference was observed in α and β when the electrode diameter was 5 mm as shown in Fig. 6(c). α was increased from 12% to 32% when the electrode diameter was increased from 3 mm to 5 mm, although β was not much changed. The maximum β attained to about 38% in both cases of 3 mm and 5 mm. When the electrode diameter was 5 mm, the energy efficiency γ attained to 1.6 L/kWh with α = 32% and β = 37%, which was the most optimized condition in our experiment. It was also worthwhile noting that such optimum condition took place in a low hydrogen flow rate of 2 sccm. This was very important for saving hydrogen consumption for the production of methane.

3.6 Effect of Gas Feeding Style

As shown in Fig. 1, the gas feeding of H_2 and CO_2 is separated, i.e., H_2 was fed from the left side and CO_2 was fed from the right side. In order to study the effect of gas feeding style, CO_2 was mixed with H_2 before feeding to the experimental apparatus, and the mixed gas was fed to the discharge region from the left side. In this case, the gas feeding from the right hand side was closed. The mixed gas was evacuated to the right side through a circumferential gap between the inner and outer glass tubes of the double glass tube. Fig. 7 shows H_2 flow rate dependence of α, β, and γ for the cases of (a) separated gas feeding and (b) mixed gas feeding at CO_2 flow rate of 1 sccm. The variations of f α, β, and γ were drastically changed in the regime H_2 < 5 sccm. In the separated case (a), both α and β were increased and attained to the local maxima at H_2 ~ 1 sccm, then these values were decreased with H_2 flow rate. α and β were 9% and 28% at H_2 = 1 sccm, respectively. In this case, γ also became the maximum of 0.5 L/kWh. On the other hand, in the case of the mixed gas feeding (b), no methane production was observed for H_2 < 3 sccm. That is, at least 3 sccm of H_2 was necessary for the CH_4 production. We got that α was 37%, β was 18%, and γ was 0.5 L/kWh at H_2 = 5 sccm. That is, in the case of the separated gas feeding style, almost same energy efficiency γ was obtained by using a small amount of hydrogen consumption, i.e., 1/5 H_2 flow rate, compared to a mixed gas feeding style. It was found that the separated gas feeding style was quite effective for reducing H_2 consumption for the generation of CH_4.

4. DISCUSSION

In our hydrogen radical shower method two processes are considered. One is a process for hydrogen radical production in the hydrogen plasma in the upper stream discharge space. The other is a process for CH_4 production by a reaction of CO_2 with hydrogen radicals in the downstream reaction space.

The production efficiency of hydrogen radicals was strongly dependent on the electron energy distribution function and electron density in the hydrogen plasma. It was found that the discharge length d gave an effect for CO_2 decomposition and CH_4 selectivity as shown in Fig. 3. When d was short, high energy tail electrons, accelerated by a large electric field between the electrodes, might excite and decompose H_2. And the reaction in Eqs. (1) and (2) were proceeded. Then, CO_2 decomposition was enhanced together with CH_4 production in the reaction space. On the other hand, when d was long, electron energy in the discharge space diminished, resulting in a decrease of CO_2 decomposition and an increase of CH_4 selectivity. However, it should be noted that an increase of the applied voltage did not simply result in an increase of CO_2 decomposition as shown in Fig. 5. When the supplied power was increased, a power loss by heating the electrode tip was not negligible, which might cause a plasma density decrease and an eventual decrease of the energy efficiency γ.

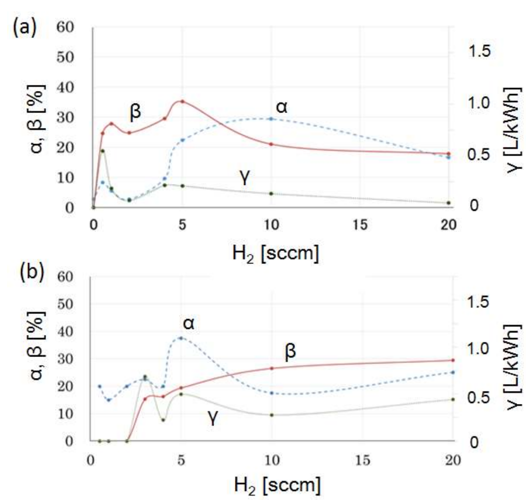

Fig. 7. Variations of α, β, and γ as a function of hydrogen flow rate for (a) separated and (b) mixed gas feedings. Applied voltage is 1.25 kV. CO_2 = 1 sccm

Electrode diameter was found to be very important for the methane production. The optimum condition was obtained when CO_2/H_2 flow rate was 1sccm/2 sccm, when the hydrogen plasma was produced with 5mm diameter electrode in a 6 mm diameter glass tube as shown in Fig. 6(c). We got that CO_2 decomposition ratio α was 32% and the CH_4 selectivity β was 37%, where the product αβ attained to approximately 12%. The energy efficiency γ for the CH_4 production was 1.6 L/kWh. Flow rate ratio of CO_2/H_2=1/2 was quite preferable, because only small amount of hydrogen was required for CH_4 production. We can save the hydrogen consumption. This point was confirmed by introducing a mixed gas feeding discharge as shown in Fig. 7(b), where no methane was produced in the lower H_2 flow rate regime. The reason why the suitable gas mixing ratio of CO_2/H_2 =1/2 was different from the stoichiometry ratio of 1/4, i.e., $CO_2 + 4H_2 \rightarrow CH_4 + H_2O$, might be due to an increase of H_2^*/H_2 ratio in the pure hydrogen plasma. The more H_2^* radicals were produced in the hydrogen discharge, the less input amount of H_2 was necessary for proceeding the reaction with CO_2, i.e., $CO_2 + H_2^* \rightarrow CO^* + H_2O$.

The energy efficiency γ described above did not include the energy for generating H_2 from the water, for example. Electrolysis is a promising option for hydrogen production from renewable resources. Industrial electrolyzer have a nominal hydrogen production efficiency of around 70% [20,21]. As described above, in our experiment, CO_2 was decomposed to form CO* by H_2^* and H* radicals in the plasma-free reaction space. Then, CO* was reduced further by H_2^* and H*, and finally CH_4 was produced. This process was quite similar to Sabatier reaction, where CO_2 was dissociated to $CO^* \rightarrow C^* + O^*$ on a heated Ni surface at 200 – 400℃. Then, H_2 reacted with C* and O* on Ni surface to generate CH_4 [22,23]. In our case, the hydrogen radicals produced in hydrogen plasma played a similar role as a catalysis effect of Ni.

Finally, we discuss a carbon balance. As mentioned above, the materials containing carbon, produced by the discharge, were simply CH_4 and CO. Methanol was scarcely produced. The other carbon materials such as HCOH and C_2 hydrocarbons like ethane, ethylene, and acetylene were not detected. Visible carbon film deposition was not detected. This might be due to that the reaction among CH_4 for hydrocarbon polymerization was restricted by the sufficient amount of hydrogen radicals injection into the downstream reaction space. Therefore, the carbon balance was simply expressed as $\alpha[CO_2] \sim \alpha\beta[CH_4] + \alpha(1-\beta)[CO]$.

It should be also noted that γ in our discharge system (α = 32%, β = 37%, and γ = 1.6 L/kWh in Fig. 6(c)) was much higher than that of conventional discharges. The energy efficiency in the case of high-pressure dielectric-barrier discharge (DBD) was reported to be 0.06 L/kWh, where α = 12.4%, β = 3.2%, and total flow rate Γ = 500 sccm (CO_2: H_2 = 1:3) for the input power of 500 W [2]. For a low pressure microwave discharge, γ = 0.027 L/kWh was reported with α = 81% and β = 1.2% at input power of 3 kW [5]. Therefore, energy efficiency in our case was fairly improved.

5. CONCLUSION

Methane was produced from carbon dioxide by using a hydrogen radical shower method. Methane was only organic species produced from CO_2. Only CO was detected as non-organic by-product. We found that the decomposition ratio α, methane selectivity β, and energy efficiency γ were α = 32%, β = 37%, and γ = 1.6 L/kWh, respectively, under optimized condition at flow rate ratio of $CO_2 : H_2 = 1 : 2$, gap distance of d = 6 mm, and input power of P_{in} = 4.6 W (1.2 kV, 3.8 mA) with a use of 6-mm-diameter electrode. Hydrogen radical shower method was a quite effective for the conversion of CO_2 to CH_4.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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