Analysis of Relationship between Reactor Temperature and Power Consumption on Methane Production System

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Abstract

Natural gas (methane) is a lower cost option and can apply in industrial sector. It can save energy and affect in both economic and environmental factors. Methane (CH_4) Production by combining Hydrogen (H_2) from water (H_2O) by electrolysis method and carbon dioxide (CO_2) from outer source has been an innovative technology for decades. There are two major theoretical and conceptual frame work for Power-to-Gas (P2G) technology. The first one is water electrolysis method and the other is Sabatier Process with two fixed bed reactors. Previous work in University of Miyazaki was examined and pursue a new method by changing reactor temperatures and using gas chromatography machine to achieve high methane concentration. In this study, a number of significant contributions to power consumption was revealed. After a series of experiments, it was found that 97.51% of methane concentration at temperature 180°C and 260°C for reactor 1 and reactor 2 for 4 hours operating time.

Keywords: P2G technology, Water electrolysis, Sabatier process, Photovoltaic, Methane, Hydrogen

1. INTRODUCTION

In recent years, all nations from all over the world are trying to keep the raise in global temperature that was happened from several sectors including carbon emission. Basically, all synthetic products we use every day are by burning fossil fuels. Carbon dioxide is released and we are rapidly warming up the earth. To deal with the impact of climate change, many developing countries are also encouraged to sign in their agreement and cooperate the rules and regulations in Global Climate Action Agenda.

Generally, it can be seen that decarburization has become a mainstream topic for the energy industry since natural gas has the lowest carbon dioxide emissions among fossil fuels[1)]. Therefore, to replace fossil fuels by sustainable resources for example renewable electricity and to store large scale of its energy for instance in summer to be able to use in winter, we use CO_2 and sustainably produce electricity with hydrogen that is produced by splitting water.

Power to fuel system can satisfy the increased demand of decarbonized energy and also is very useful in the transport system. In the P2G technology, the promising energy carrier is hydrogen as it can be easily produced by electrolysis of water[2)]. Further conversion of hydrogen to methane by CO_2 from outer source has done with thermal reactors.

The challenging problem which arises in this domain is choosing the best reactor and catalyst to outcome high methane concentration efficiency. Since the methanation process is an extremely exothermic reaction, an effective method of temperature management is to control the reactor inlet gas stream; this may cause undesired energy losses. In addition, heat management in a fixed-bed reactor may be difficult. Therefore, in order to manipulate the reactor with temperature controlling medium system, at least two reactors are needed to connect in series for good control of reaction temperature in a fixed bed reactor to maximize the CH_4 generation[3)]. Further major point is that Ni-ZrO_2 is an active Ni-based catalyst for CO_2 methanation as a result of enhanced Ni dispersion resulting from the presence of Zr. The other advantages of this are of its low cost, highest activity, and highest selectivity for methane formation. In all cases, a catalyst is required that is capable of increasing reaction rates within the temperature range of 25°C to 500°C[3)].

In this study, we investigate methods for improving methane conversion efficiency and solar-to-methane (StM) efficiency. Therefore, we changed the temperature of both reactors and analyzed. After that, to calculate the methane concentration, we used gas chromatography to measure methane concentration. It has significant benefits while evaluating the efficiency.

On the other hand, power consumption is also vital in response to the challenges of the increasingly prominent problems such as climate change and

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environmental pollution, which severely threaten human society and development. This study will compare the power consumption by utilizing low reactor temperature and high reactor temperature. Only 0.156 kWh in 4 hour operating time for both reactors was needed and higher methane concentration was achieved. This has particular advantages over other systems. In power consumption section, the relationship between methane concentration and power consumption will be demonstrated.

2. Related Work

The former researchers evaluated the Sabatier reaction utilizing hydrogen produced by concentrator photovoltaic (CPV) modules and used quadrupole mass spectrometer (QMS) to measure methane concentration. They used an established technique, namely solar-to-hydrogen (StH) conversion system to analyze its efficiency with the performance of CPV modules. At University of Miyazaki, three highly efficient CPV modules were installed and connected in parallel with four sets of DC/DC converters and EC cells. Sunlight was converted to electricity by CPV modules and was supplied to EC cells[3,4].

For the current work, in order to point out PtG technology with different reactors temperatures, power supply was utilized instead of using photovoltaic modules while measuring the composition of methane with different components of reactants. After then, utilizing CPV modules, methane production was analyzed with variable of reactors temperature. In this study, we used gas chromatography (GC; GC 7100, J-Science) to measure methane concentration. It has significant benefits while evaluating the concentration efficiency. The results were more accurate.

3. Experimental Procedure

Figure 1 shows the summary of StM process. Firstly, photovoltaic modules collect the sun light and convert them into electricity. The power supply to electrochemical (EC) cells was performed by using 4 DC/DC converters which was connected in parallel with 4 EC cells. DC/DC converters convert lower DC voltage into higher levels by storing the input energy and then releasing into the output at a higher voltage level. EC cells generate hydrogen by chemical reactions. Water was split electrochemically into H_2 and O_2. Two reactions occurred and the anode reaction is oxygen evolution reaction. The water was oxidized to oxygen. At the cathode reaction, hydrogen evolution reaction was taken placed. They are shown in equations (1) and (2)[5].

$$2H_2O \longrightarrow O_2 + 4H^+ + 4e^- \quad (1)$$

$$4H^+ + 4e^- \longrightarrow 2H_2 \quad (2)$$

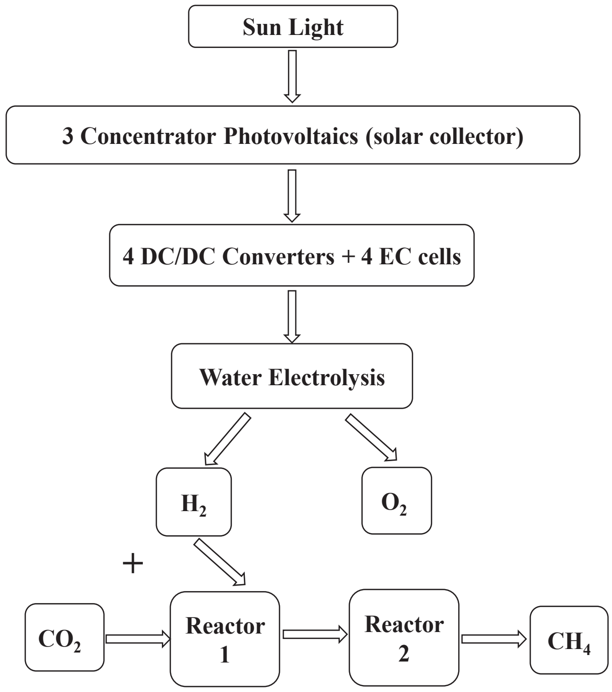

Fig. 1. Flow chat of the summary of Solar-to-Methane conversion process.

The next step is Sabatier process. Methane can be obtained by the following exothermic reaction.

$$CO_2 + 4H_2 \longrightarrow CH_4 + 2H_2O \quad \Delta H = -165 \text{ kJ/mol} \quad (3)$$

The stoichiometric ratio of CO_2 and H_2 is 1:4[6,8]. To investigate this statistically, we changed 0.02 point of CO_2 in every 30 min interval measurements to achieve the highest methane concentration at 260°C of each reactor. The evolved hydrogen from Polymer Electrolyte Membrane (PEM) electrolysis method was filtered with dryer (silica gel) and dry pure gas was put into the reactors. At that time, CO_2 was introduced from the outside source. Reaction was occurred in Reactor 1 first, and then, the unreacted gas flew through to the reactor 2.

Fig. 2. Two fixed bed reactors.

Consequently, since the Sabatier process is an exothermic reaction, it is limited by the thermodynamic equilibrium. Typically, lower temperatures from 250°C to 400°C is desirable for higher methane selectivity[7]. In this study, we set the reactor temperatures as shown in table 1 in order to get higher reaction rate from reactor 2 to product higher concentration of CH_4.

Table 1. Reactor temperature set.

	Reactor 1 temperature (°C)	Reactor 2 temperature (°C)
Low temperature condition	180	260
High temperature condition	260	260

As catalyst effects the product yield[6], Ni/ Y-doped ZrO_2 catalyst which support the reaction activity was utilized. Firstly, the two gases were feed into the reactors and they diffused in the catalyst. And then, the reactants, the left side of equation (3), are absorbed on an active site. Hydrogen molecules and carbon dioxide molecules have been absorbed and they undergo the Sabatier reaction. After that, they desorb from the catalyst surface as methane molecules and diffuse out to leave the catalyst[9]. Moreover, N_2 gas is used as gas carrier in methanation system and it also performed as cleaning process to refresh the system after the experiments were finished.

To analyze the amount of methane composition, we used an established technique, namely gas chromatography (GC) as shown in figure 3. It is a common type of chromatography used in analytical chemistry for separating and analyzing compounds. By utilizing it, the relative amount of components can be determined. Firstly, standard gases were allowed to pass through the system and determined. Argon (Ar) is used as carrier gas. And then, mixed elements are separated into single elements while being carried out by Ar gas. In interface section, carrier gas was removed and elements were lead to ion source and formed into ions, accelerated and sent to analysis section. Finally, detector catch and name the quantity of compounds.

In our research, the produced gases were analyzed in every 30 min interval time for 4 hours.

Fig. 3. Gas Chromatography.

4. ANALYSIS AND RESULTS

To determine the highest methane concentration yield, we analyze the different stoichiometric ratio. As shown in figure 4, 0.92: 4 ratio of CO_2: H_2 was the best efficiency outcome. Accordingly, the later experiments were done with this proportion.

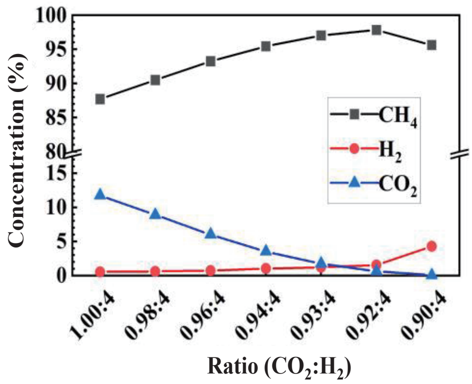

Fig. 4. Different stoichiometric ratio of CO_2 and H_2.

We carried out on June 22, 2020 (low-temperature condition) and June 23, 2020 (high-temperature condition) to evaluate the methane production and to measure the power consumption of reactors under actual outdoor conditions. The normal direct irradiance (DNI), which was converted to electricity by using the CPV module, was stable in two-days as shown in figure 5. The StM system was operated between 10:00 a.m. and 2:00 p.m.

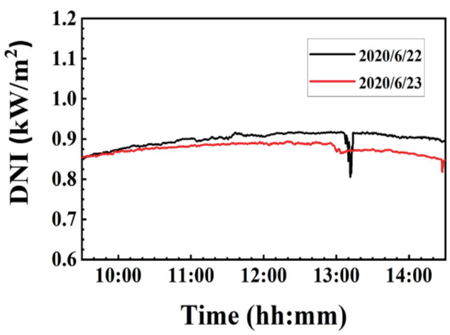

Fig. 5. DNI during the experimental period.

Figure 6 shows the concentration of (a) CH_4, (b) H_2 and (c) CO_2 on different reactors temperature conditions. Methane concentration declined during system operation (as shown in figure 6 (a)) when hydrogen concentration was increased and carbon dioxide concentration was reduced.

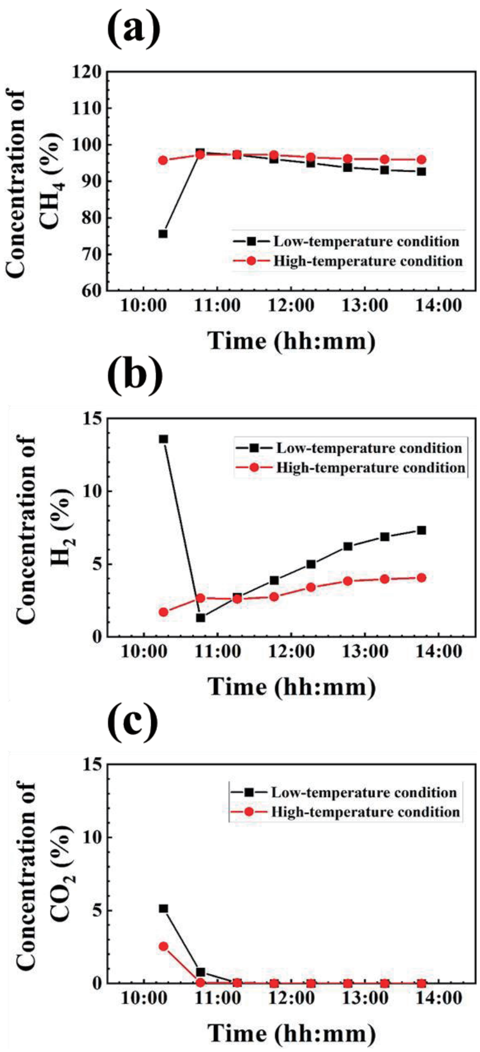

Fig. 6. Concentration of (a) CH_4, (b) H_2 and (c) CO_2 on different reactors temperature.

Figure 7 shows the time dependence on surface temperature ($T_{surface}$) of reactors 1 and 2. The dot lines are also shown in the target temperature. By comparing the two-days data of reactors changed, it can be seen that in figure 7 (a) 180°C of reactor 1 and 260°C of reactor 2, temperature of reactor 1 was reached 180°C at about10 am and then raised over 300°C. It was because the catalyst action released heat and surface temperature is increased. At that time, reactor 2 temperature was firstly increased over targeted temperature 260°C and then, maintained at the setting temperature. For Figure 7 (b) 260°C for both reactors, the reactor 2 temperature was stable and reactor 1 was nearly the same with the former one.

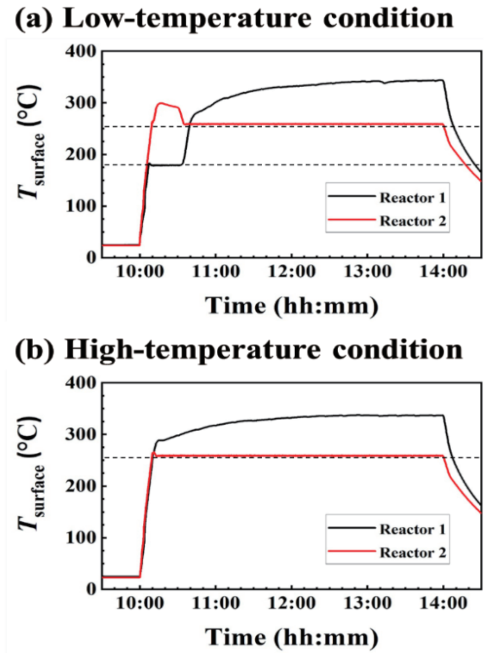

Fig. 7. Time dependence on surface temperature ($T_{surface}$) of reactors 1 and 2.

Aside from that, we examined the various temperature effects. The key point of the adjustment in temperature is that to get lower power consumption with high methane concentration. Furthermore, high feed temperatures will effect in reactor overheating, negatively resulting product distribution of the exothermic Sabatier reaction process[10].

Therefore, the other fact such as fluctuations of temperature are unavoidable in operating time. Figure 8 shows the relationship between reactors temperature and its power consumption. It has significant benefit on lowering temperature of reactor 1. Not only the total power consumption of reactors was low, but also the higher methane concentration was achieved as shown in table 2.

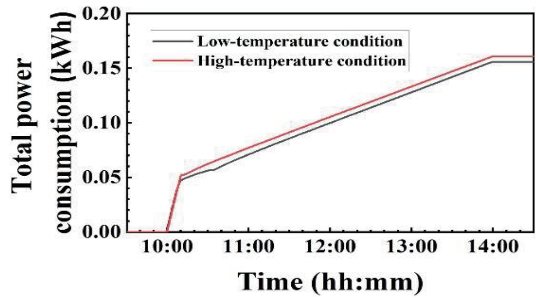

Fig. 8. The relationship between reactors temperature and its power consumption.

Table 2. Effect of power consumption depending reactor on temperature

Date	Time	Power Consumption		Total Power Consumption of reactors (kWh)
		Reactor 1 (kWh)	Reactor 2 (kWh)	
2020.06.22	10 am to 2 pm	0.031	0.125	0.156
2020.06.23	10 am to 2 pm	0.028	0.133	0.161

The assumption of the above mentioned table is supported by the fact that methane concentration reached at maximum peak with 97.51% when we utilized the lower reactor 1 temperature with 180°C. Moreover, the power consumption was also obtained lower when comparing the result of using 260°C for both reactors. On the flip side, solar to methane efficiency (η_{StM}) can be defined as the following equation (4)[3].

$$\eta_{StM} = \frac{|\Delta H^{o}| \times \text{amount of } CH_4}{E_{DNI} + E_{reactors}} \quad (4)$$

where ΔH^{o} = -802 kJ/mol, the integrated irradiance is defined as E_{DNI} and $E_{reactors}$ is expressed as the energy consumed by the reactors in the methanation system[3]. Therefore, it can be seen that, StM efficiency increase when the electrical energy is reduced in case of low temperature reactor.

5. CONCLUSION

Overall, our results demonstrate a strong effect of the best outcome methane concentration 97.51 % at 0.92: 4 ratio of carbon dioxide and hydrogen, which was produced by electrolysis method by the Sabatier reaction at the temperature of 180°C and 260°C from reactor 1 and 2 respectively. Power consumption of reactors was analyzed and it can be seen that the results are outstanding. Higher methane concentration with lower power consumption was achieved.

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