An Environmental Analysis of Bio-H₂ Production
Introducing the 2-step PSA and Waste Heat Recovery

Haruka NAKAYAMA※1†, Mitsuo KAMEYAMA※2, Yin LONG※1, and Kiyoshi DOWAKI※1

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Hydrogen (H₂) is expected to be one of the most promising secondary energy resources that can contribute to the mitigation of global warming. H₂ can be synthesized from biomass feedstock as Bio-H₂, which can contribute to greenhouse gas emission mitigation. However, the low energy density of Biomass limits the collection of the feedstock based on the life-cycle assessment methodology. Furthermore, for successful operation, small-scale Bio-H₂ plants must be used. Thus, a Bio-H₂ plant faces two challenges: 1) the need for auxiliary power negatively affects production efficiency and increases eco-burden. In particular, pressure swing adsorption (PSA) used for H₂ purification requires high power consumption, 2) a large amount of heat is lost. To overcome these limitations, we improved a previous Bio-H₂ plant by introducing 2-step PSA and a waste heat recovery system (WHR) to reduce the auxiliary power consumption and to compensate the auxiliary power demand. It was found that, the improved plant reduced the auxiliary power demand by 18.9%, and 7.2% of the required power was compensated by the WHR system. Furthermore, all environmental indicators were improved compared to those for the conventional plant.

1. Introduction

Hydrogen (H₂) is considered to be a clean energy resource 1). However, fossil fuels are currently the major source of hydrogen 2), giving rise to environmental problems. Therefore, alternative approaches for hydrogen production are expected to be developed to provide a cleaner energy source 3).

Biomass feedstock is a renewable resource, that does not increase greenhouse gas (GHG) emissions because of its carbon neutral lifecycle 4). However, biomass feedstock has low energy density, which is unfavorable because its use requires the collection of a large amount of biomass materials. Therefore, based on life-cycle assessment (LCA), the use of biomass for hydrogen generation will increase GHG emissions even if the biomass material itself is carbon

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Key Word

Waste heat recovery, Organic Rankine Cycle, Biomass gasification, Life Cycle Assessment (LCA)

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※1 Department of Industrial Administration, Graduate School of Science and Technology, Tokyo University of Science, Japan
※2 Japan Blue Energy Co., Ltd., Japan
† Corresponding author: teniss.haruka@gmail.com
neutral. Furthermore, for economic plant operation, the plant size is strictly limited to small scale.

The conversion of biomass to $\text{H}_2$ can be carried out through various processes, with gasification being one of the most promising technologies. The indirect pyrolysis gasification process in a moving bed is a biomass gasification process in which the biomass feedstock is converted into syngas with a high concentration of $\text{H}_2$ through the use of a gasifier. Then, the $\text{H}_2$ refining process in which $\text{H}_2$ is purified by pressure swing adsorption (PSA) is carried out.

The $\text{Bio-H}_2$ gasification plants used for $\text{Bio-H}_2$ production using the above-mentioned process are limited to small scale and face two challenges: 1) the need for auxiliary power has a negative impact on production efficiency and eco-burden. In particular, PSA for purification of $\text{H}_2$ consumes 58% of the power required for plant operation. 2) A large amount of heat is lost.

To reduce the auxiliary power demand in PSA, Dowaki et al. developed the 2-step PSA process. This process enables a reduction of the required power by lowering the operating pressure while achieving the same $\text{H}_2$ refining efficiency as conventional one. Kondo et al. showed that the introduction of 2-step PSA enables the plant to produce $\text{Bio-H}_2$ with 12.6% lower auxiliary power than the plant using conventional PSA, mitigating the eco-burden (e.g., global warming potential (GWP) and abiotic depletion potential (ADP), etc.). Additionally, carbon dioxide ($\text{CO}_2$) is recovered prior to $\text{H}_2$ purification in 2-step PSA. This dramatically reduces GHG emissions, assuming that the produced $\text{CO}_2$ can be used for fertilization in agriculture because the promotion of photosynthesis by the application of $\text{CO}_2$ increases crop yields, as shown for paprika horticulture.

However, eco-burden related to auxiliary power was still significant; therefore, further reduction in the auxiliary power is required. Here, power generation from exhaust heat is considered to be an effective approach because it may address both challenges simultaneously. That is, the recovery of the heat loss energy increases the total energy efficiency and mitigates GHG emissions.

For a plant scale of 12 t-dry/d, waste heat of approximately 4 kWh/kg $\text{H}_2$-product at medium-low temperatures (between 200 and 300 °C) is produced, in contrast to the power of 7.4 kWh required to generate 1 kg of the $\text{H}_2$ product.

Organic Rankine cycle (ORC) is an effective approach for the utilization of waste heat in the medium-low temperature range. ORC has been applied to geothermal, waste heat, and bottoming cycle combined with gas turbines. Compared to water, organic fluids are advantageous for use because of their requirement of low maximum temperatures and/or small power plants. At low temperatures, organic fluids enable a higher cycle efficiency than water. The use of ORC in small plants is also advantageous from legal and economic standpoints. Water shows good efficiency at high pressures, but requires increased safety measures, whose implementation is not economically feasible for small plants. Thus, the use of ORC is beneficial for biomass applications.

Therefore, in this study, a $\text{Bio-H}_2$ gasification plant with 2-step PSA and a waste heat recovery (WHR) system was designed, and its production efficiency and eco-burden were investigated and compared to those of a conventional plant based on the LCA concept. We carried out the process design using the process simulator of Aspen Plus Ver. 11 and perform the calculation of the eco-indexes based on the LCA using the LCA software package of SimaPro Ver. 8.5.

2. Process design and evaluation methods

2.1 Process design

Generally, for $\text{Bio-H}_2$ production from a gasification plant, biomass feedstock is decomposed in a pyrolyzer, generating pyrolysis gas. The pyrolysis gas reacts with $\text{H}_2\text{O}$ (steam) and produces syngas with a high concentration of $\text{H}_2$ (called reforming). Then, syngas moves through the gasifier into the desulfurizer to remove the sulfur compounds, and the clean syngas is converted to the gas with a high concentration of $\text{H}_2$ through the water gas shift (WGS) process. Finally, the produced $\text{H}_2$ fuel is refined to pure $\text{H}_2$ with the purity of 99.99 vol.% using the PSA equipment.

In this study, we assumed the conventional Blue Tower process with the scale of 12 t-dry/day using Japanese cedar as the feedstock. In this plant, the heat required for syngas generation from biomass feedstock is supplied by the circulation of alumina balls of heat carriers (HCs). The plant consists of the pyrolyzer, reformer, and pre-heater vessels. In each vessel, multiple alumina balls called heat carriers (HCs) move to transfer the heat volume (see Fig. 1). Assuming that the plant scale was 12 t-dry/day, the circulated weight of $4.1 \times 10^3$ kg/h and the HIC diameter of $\varnothing = 9.5$ mm were suitable conditions. The thermal conductivity of HIC was considered to be 3.50 W/(m·K). In addition, the residence time of the balls in each vessel was assumed to be 60 min. Under these conditions, besides the heat transfer in use of HCs can be executed in each vessel, the tar, char (mainly carbon content) and ash are likely to be almost separated in the pyrolyzer. On the other hand, as mentioned above, the heat transfer of HCs at high temperature will generate a high heat loss, even in the presence of suitable insulation.
Thus, the heat recovery process is important not only for increasing the energy efficiency but also for eco-burden mitigation.

To overcome the two challenges of the auxiliary demand and heat loss, the following two cases were compared with regard to the production efficiency and eco-burden (see Fig. 1)

- Case 1: Conventional process
- Case 2: Improved process (2-step PSA, WHR)

The differences between the two cases lie in the details of the PSA process, and the use or lack thereof of the heat recovery generator. The specifications for PSA are shown in Table 1. Only in Case 2, a WHR system based on ORC was assumed to be introduced. The main components of this system are an evaporator, a condenser, a pump, a turbine and a generator. In this study, R245fa with a low boiling point, no flammability, and no ozone depletion potential is used as the working fluid. Table 2 shows the properties of R245fa. The fluid is assumed not to leak because the power generation equipment without any fluid leak has already been developed. In the plant, exhaust heat generated after the heating of alumina balls in a

| Table 1 Conditions and performances of conventional PSA and 2-step PSA |
|---------------------------------------------------------------|
|                  | Conventional PSA | 2-step PSA       |
| Adsorption Temp. [°C] | 40               | 30 (1st.) 30 (2nd.) |
| Pressure [MPaG]      | 0.8              | 0.4 (1st.) 0.4 (2nd.) |
| H₂ recovery eff. [%] | 74%              | 55 (2nd.)        |
| CO₂ recovery eff. [%] | –               | 97 (1st.)        |
temperature of R245fa. Therefore, it is better to lower the temperature to 230 °C by blowing air before the heat exchange with the working fluid. The specifications of WHR are summarized in Table 3.

The process parameters for the process simulations that are identical for both cases are shown in Table 4.

2.2 Definition of Efficiencies

The thermal efficiency of WHR, \( \eta_{\text{WHR}} \), is calculated as follows:

\[
\eta_{\text{WHR}} = \frac{W_{\text{NET}}}{\Delta Q_{\text{in}}} = \frac{W_{\text{out}} - W_{\text{aux}}}{\Delta Q_{\text{in}}},
\]

(1)

where \( \Delta Q_{\text{in}} \) [kJ/h], \( W_{\text{NET}} \) [kJ/h], \( W_{\text{out}} \) [kJ/h], and \( W_{\text{aux}} \) [kJ] are the heat input in the evaporator, net power through the WHR generator, gross output, and the auxiliary power (such as the required pump work), respectively.

Next, the H2 energy efficiency is defined based on the primary energy \( \eta_0 \) as follows:

\[
\eta_0 = \frac{Q_{\text{the}}}{Q_{\text{bio}} + Q_{\text{grid}}},
\]

(2)

where \( Q_{\text{the}} \) [kJ/h], \( Q_{\text{bio}} \) [kJ/h], and \( Q_{\text{grid}} \) [kJ/h] are the lower heating value of the H2 product, lower heating value of the biomass feedstock, and conventional grid power for auxiliary power applications, respectively. The power efficiency for grid power in Japan is considered to be 36.9% \(^{22}\).

2.3 Methodology of LCA evaluation

LCA is a methodology for accounting for the life cycle environmental performance of a product during extraction of raw materials, manufacturing and transportation, and, finally, disposal \(^{23}\). The functional unit used in this study is 1 kg/h of Bio-H2. The system boundary is illustrated in Fig. 2. Six subsystems were defined as chipping (SS1), transportation (SS2), drying (SS3), syngas production (SS4), hydrogen purification (SS5), and WHR (SS6) (only Case 2).

For the subsystem of chipping, the required electric power and diesel fuel were set to 0.0136 kW/kg-feedstock

- The preheater is available, and therefore, we designed a WHR system using exhaust gas from the preheater (see Fig. 1 (Case 2)). The exhaust gas temperature is 251 °C, and this is over which is higher than the thermal decomposition...
and 1.033 g/kg-feedstock, respectively \(^{24}\). Based on the plant scale assumption, it was reported that the collection area of biomass feedstock is 5–50 km \(^{25}\). Here, the average of these two values (27.5 km) was used as the transportation distance. The energy consumption for drying, syngas production, and hydrogen purification and the generated power were estimated using a process simulator.

The background data were obtained from the Ecoinvent 3.2 database and using SimaPro (ver.8.5.0.0), and CML-IA baseline \(^{26}\). There are 11 impact categories for impact indicators; we performed environmental impact assessment using 6 indicators: cumulative energy demand (CED), global warming (GWP), ozone layer depletion (ODP), photochemical oxidant formation (POFP), acidification, and eutrophication (EP).

Next, to identify the subsystems that provide substantial improvement, we analyzed the contribution of each subsystem to the total reduction amount. Assuming that the indicator of the \(i\)-th subsystem, the \(j\)-th impact category and the \(k\)-th case is \(x_{ijk}\), the reduction ratio \(X_{ij}\) is expressed as follows:

\[
X_{ij} = \frac{x_{ijk} - x_{ij1}}{\sum_k (x_{ijk} - x_{ij1})}
\]

\[
\sum_j X_{ij} = -1
\]

3. Results and Discussion

3.1 Performance and efficiency

In our simulations, the amounts of the Bio-H\(_2\) product were \(5.04 \times 10^{-2}\) and \(4.94 \times 10^{-2}\) kg-H\(_2\)/kg-biomass/h from 1 kg/h biomass feedstock in Cases 1 and 2, respectively. We can obtain \(4.15 \times 10^{-4}\) kg/kg/h of CO\(_2\) with 99.5 vol.% purity only in Case 2 because the CO\(_2\) is recovered prior to H\(_2\) purification in 2-step PSA.

The performance and efficiency results are shown in Table 5. Based on our results in Case 2, the auxiliary power demand was reduced by 18.9% compared to that for Case 1.

For WHR, the generated power was 14.5 kW, compensating 7.2% of the auxiliary power required for the entire plant. In the H\(_2\) refinery process, the use of 2-step PSA reduced 17.5% of the auxiliary power demand.

Additionally, the primary energy efficiency was improved by 0.9%. This means that Bio-H\(_2\) can be produced with a lower primary energy input. For the practical implementation of a WHR generator, a generator on the 20 kW scale that has already been developed is suitable \(^{23}\).

3.2 Error analysis of the primary energy efficiency

Next, the error analysis was carried out to assess the significance of the primary energy efficiency gain. The significance with or without the countermeasures of 2-step PSA and WHR was evaluated.

In this study, we focused on the fluctuation of the operating temperatures. In particular, the pyrolizer, the reformer and the pre-heater are most impacted in terms of heat loss because the heated HCs are circulated into these vessels. Therefore, using the temperature at the reformer as a control parameter, we compared the efficiencies in the presence of the fluctuation.

Based on the previously reported demo-plant test data, we considered the temperature variation between 910 and 963 °C. In Table 6, we estimated the increase in the primary energy efficiency due to the heat recovery in the reforming temperatures of 910, 936 and 963 °C. We considered that the initial design temperature is 950 °C.

Table 6 shows the results for the primary energy efficiency gain. For each result, the gain is almost the same, even though the amount of the available waste heat varies at the different reforming temperatures, that is, the gain is almost the same even in the presence of a temperature fluctuation.
deviation. This means that a gain in the efficiency will be obtained due to the use of 2-step PSA and WHR.

3.3 Environmental Impacts

Finally, we discuss the environmental impact analysis results. **Fig. 3** shows the percent contribution of each indicator. The environmental impact of the Bio-H\textsubscript{2} is mainly due to the use of fossil fuel (e.g. for grid power). For the 2-step PSA and WHR system that used external grid power as an auxiliary power source, the external fossil fuel input will be reduced by our proposed system. In particular, the following 6 impact categories: cumulative energy demand (CED), global warming potential (GWP), ozone layer depletion potential (ODP), photochemical oxidant formation potential (POFP), acidification potential (AP) and eutrophication potential (EP) will be improved because coal, gas and oil are the main sources of the fuel used for the grid power generation in Japan. Thus, due to the reduction in the grid power consumption, direct emissions of nitrogen dioxide and sulfur dioxide and CO\textsubscript{2} into the atmosphere will be mitigated. Here, GWP has improved by 25% (see **Fig. 4**), and a reduction of 1.0 kg CO\textsubscript{2} eq./kg-H\textsubscript{2} in GWP is obtained by introducing 2-step PSA and WHR, corresponding to 658 kg CO\textsubscript{2} eq.

Moreover, assuming the use of CO\textsubscript{2} product as a co-product fertilizer in agriculture, GHG emissions will be dramatically mitigated in Case 2, with GWP of 4.4 kg CO\textsubscript{2} eq in Case 1 decreasing to -12.9 kg CO\textsubscript{2} eq in Case 2. In this case, the use of Bio-H\textsubscript{2} reduces GHG emissions.
4. Conclusions

The Bio-H₂ gasification plants limited to small scale were investigated with regard to the following two challenges: 1) the recovery of exhaust heat by a heat recovery generator, and 2) the abatement of the eco-burden of Bio-H₂ production. To achieve these goals, a plant with 2-step PSA and WHR power generation was designed.

Our simulations showed that the improved plant will reduce the auxiliary power demand by 18.9% compared to the conventional plant. The use of WHR enables a reduction of 7.2% in the auxiliary power required for the entire plant, and the use of 2-step PSA reduces the power demand in the refinery process by 17.5%. This indicates that Bio-H₂ with a 0.9% decrease relative to the primary energy basis can be produced. Similarly, all environmental indicators were improved. In particular, GWP which is considerably affected by the conventional grid power usage, was improved by 25%.

The WHR system designed in this study will result in a strong improvement of the process of Bio-H₂ production in terms of efficiency and environmental performance.

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