A System Analysis of Storage Alloy for Bio-H₂ in Consideration of the Purification Performance

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The environmental performance of hydrogen production via indirect gasification of Blue Tower including the use phase by a fuel cell (FC) cell phone was evaluated following a Life Cycle Assessment (LCA) approach. Foreground data for this study were provided mainly from process simulation in consideration of the adsorption and gasification experiments. Also, the problems in the whole system were clarified, and the countermeasures for them were proposed.

In the production phase, it is necessary to reduce the auxiliary power by the installation of 2-step pressure swing adsorption (2-step PSA) unit and remove the contaminants of H₂S. Here, it found that H₂S would be able to be entirely removed at the storage. In the use phase, sufficient H₂ storage capacity and stable power supply of a FC device, which has poor load-following operation due to large over-potentials, are absolutely required in order to meet various demands. Based on the specification of a FC-cell phone with electric double-layer capacitors (EDLCs), the Life Cycle CO₂ (LCCO₂) emission was estimated. Bio-H₂ was generally found to be a promising hydrogen fuel, with reduced greenhouse gas emissions. Especially, the negative emission had significantly impact even if the direct emission was small in comparison with the direct one.

Key Words
Biomass gasification, 2-step PSA, LCCO₂, EDLCs

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1. Introduction

Hydrogen can play a leading role in attaining environmental goals. However, this is conditioned by the primary energy and the conversion technology used to produce hydrogen. In this respect, urgency in global warming mitigation may result in a growing interest in bio-

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balance of GHG emissions has to be evaluated from a life-cycle perspective and requires an appropriate choice of both the biomass feedstock and the biomass conversion technology. For instance, from the viewpoint of production technology, indirect biomass gasification has proven to involve a favorable life-cycle performance when compared to conventional, fossil-based hydrogen from steam methane reforming (SMR) in terms of global warming and cumulative non-renewable energy demand. Hence, implementing CO2 capture in this type of Bio-H2 production system could be a promising energy strategy.

Furthermore, Life Cycle Assessment (LCA) has been widely taken as an effective analytical tool to investigate the total environmental impacts in all phases of an industrial activity, from raw materials acquisition to the final disposal. In recent researches related to the LCA of H2 fuel, the eco-burden of H2 fuel is compared to those of resources from other origins, e.g. the lowest specific GHG emissions of H2 fuel from renewable resources are produced by wind. For instance, there is a report that emission from biomass has been known at approximately 25% of those of conventional H2 production systems from natural gas origins. Besides, the H2 supply from biomass feedstock has some advantages of a steady supply and a character of carbon-neutral.

For mitigating Life Cycle CO2 (LCCO2) emission of H2 production of biomass feedstock origin, Susmozas A. et al. proposed the indirect gasification process with CO2 capture. CO2 capture is operated in a two-stage gas separation membrane. Also, using the off gas from pressure swing adsorption unit (PSA), the electricity is produced in a steam cycle. Due to these equipment, CO2 mitigation is able to be achieved with significant advantage. While, the productivity of H2 is worse than that of conventional system of natural gas origin.

Considering the practical use of H2 fuel from biomass feedstock, both better productivity of Bio-H2 and mitigation of LCCO2 have to be achieved. Also, in the LCA estimation on Bio-H2 production, besides the productivity of H2, the quality improvements of purification in consideration of contaminant removal, and the usability for an application performance and storage capability should be considered. Thus, in this paper, the following topics were argued on 2-step pressure swing adsorption (2 step-PSA) unit: (1) estimation of productivity of Bio-H2 through biomass gasification process and specific CO2 emission of Bio-H2 on basis of LCA, (2) probability of H2S removal through PSA unit, and (3) performance of H2 storage and usability of a FC-cell phone application.

2. Process design

2.1 Performance of Bio-H2 fuel

Here, based on our prior studies, the Bio-H2 was assumed to be synthesized through an indirect gasification process of Blue Tower (BT) and PSA. First, using the process parameters of 2-step PSA whose performance on basis of experiment was introduced, the productivity of Bio-H2 was analyzed. The advantage of this case is reduction of auxiliary power, especially, in PSA operation. Simultaneously, CO2 is also captured. The whole system consists of a refining process of pure H2 fuel and an extraction of CO2 as a by-product through 2-step PSA based on a BT process (see Fig. 1). Note that the conventional case

![Fig. 1 Schematic design of Bio-H2 production system with CO2 capture](image-url)
of single step PSA was estimated for comparison. In this analysis, the $H_2$ fuel production performance was evaluated in use of the process simulator of Excel VBA, which was developed on basis of experimental data\textsuperscript{6,11-13}.

In this study, the biomass feedstock is Japanese cedar. Tables 1 and 2 show the measured chemical properties of biomass feedstock and char, respectively. Note that char is discharged from the reactor. The initial moisture content of feedstock is assumed to be 50 wt. %. However, the moisture content at the inlet of gasifier is pre-treated by drying from 50\% to 20\% due to hot flue gas. Although the contaminants of sulfur and/or chlorine were contained in biomass feedstock, they are out of consideration in this section in order to investigate the productivity of Bio-$H_2$ only. Hence, the influences due to operating conditions of PSA into which the syngas with a high concentration of $H_2$ excluding the contaminants is fed were argued.

Here, the simulation parameters of this plant which was set by our previous studies are shown in Table 3. Note that Hydroxyl Aluminum Silicate-Clay (HAS-Clay) at 1st stage and Zeolite A-5 at 2nd stage in 2-step PSA were stuffed as adsorbent. The properties of each adsorbent are shown in Table 4.

HAS-Clay can absorb mainly $CO_2$ gas and $H_2$ gas can pass through the reactor of Zeolite-A5, and then pure $H_2$ fuel can be obtained. Based on these conditions, the syngas components of 1 kg/h of feedstock at 20\% moisture are represented as Eq. (1).

\[
\begin{align*}
C_{31.078}H_{43.495}O_{23.677}N_{0.069} + 31.078H_2O \rightarrow \\
29.946H_2 + 14.508CO + 0.255CH_4 + 8.796CO_2 + 6.109 \times 10^{-5} NH_3 + 21.964H_2O + 0.029N_2 + \text{char} (C_{2.866}H_{0.482}O_{0.343}N_{0.004})
\end{align*}
\]

Next, according to this whole reaction of Eq. (1), the cold gas efficiency which is defined by the ratio of lower heating value (LHV) of syngas [kJ/h] against LHV of feedstock [kJ/h] was 87.3\%. The carbon conversion ratio which is estimated by the converted carbon content in syngas [mol/h] against the initial carbon content in feedstock [mol/h] was 75.8\%. Also, based on the operating conditions for single step PSA (conventional case) and 2-step PSA, the $H_2$ productivity in each case are as follows: (1) in conventional case, the product rate of pure $H_2$ was 305.0 Nm$^3$/h with the total recovery efficiency of 39.1 %-LHV and the auxiliary power of 255.7 kW (PSA of 145.2 kW), and (2) in the case of 2-step PSA, the product rate was 301.6 Nm$^3$/h with the efficiency of 38.7 %-LHV and the auxiliary power of 226.5 kW (PSA of 114.3 kW). Note that the energy consumption for $H_2$ storage is not considered in both cases.

### Table 1 Chemical properties of Cedar

| Parameter               | Value |
|-------------------------|-------|
| Carbon [wt.%]           | 46.66 |
| Hydrogen [wt.%]         | 5.48  |
| Nitrogen [wt.%]         | 0.12  |
| Oxygen [wt.%]*          | 47.35 |
| Ash [wt.%]              | 0.39  |
| Volatiles [wt.%]        | 86.21 |
| Higher Heating value [kJ/kg] | 18,348 |

*Oxygen by difference

### Table 2 Chemical properties of Char

| Parameter               | Value |
|-------------------------|-------|
| Carbon [wt.%]           | 84.26 |
| Hydrogen [wt.%]         | 1.27  |
| Nitrogen [wt.%]         | 0.13  |
| Oxygen [wt.%]*          | 14.34 |
| Higher Heating value [kJ/kg] | 31,988 |

*Oxygen by difference

### Table 3 Parameters of process simulation\textsuperscript{6,11-13}

| Component                 | Value       |
|---------------------------|-------------|
| Gasifier (BT)             |             |
| - Plant Scale (Wet basis) | 15.0        |
| - Pyrolysis zone [deg.C]  | 550         |
| - Reforming zone [deg.C]  | 950         |
| - Circulation rate of heat carrier [kg/kg-feedstock] | 6.34 |
| - Steam/Carbon ratio [\(\times\)] | 1.0 |
| - Operating pressure [MPaA] | 0.106 |
| Reformer (HTS/LTS)        |             |
| - Operating temp. (HTS) [deg.C] | 350 |
| - Operating temp. (LTS) [deg.C] | 240 |
| PSA (Conventional)        |             |
| - Operating pressure [MPaG] | 0.8 |
| - $H_2$ recovery eff [%-LHV] | 67.5 |
| PSA (2-step)              |             |
| - Operating pressure [MPaG] | 0.4 |
| - $H_2$ recovery eff [%-LHV]* | 55.0 |
| - $CO_2$ recovery eff. [%]** | 62.6 |

* Purified $H_2$ [kJ/h]/Inlet $H_2$ of PSA [kJ/h]
** Captured C [mol/h]/Feedstock C [mol/h]

### Table 4 Adsorbent properties

| Specification | HAS-Clay | Zeolite A-5 |
|---------------|----------|-------------|
| Chemical composition | SiO$_2$:Al$_2$O$_3$:H$_2$O (Toda Kogyo Corp.) | MeO:Al$_2$:mSiO$_2$:nH$_2$O (Me: Cation) (Wako Pure Chemical Industries, Ltd.) |
| Particle size [mm] | >2.0 | 2.36-4.75 |
| Pore diameter [nm] | <0.02 | 0.5 |
The reason why the production rate of H2 is almost same in both cases is attributed to the heat volume of total supply of syngas and off-gas which are required in order to maintain the designed temperature in the gasifier. That is, in the conventional case, the total heat consumption of syngas and off-gas was 3,686 MJ/h, while that in 2-step PSA case was 3,704 MJ/h. Consequently, this indicates that the production of H2 fuel in our system can be achieved without any production loss, even if the recovery efficiency in PSA is decreased to some extent.

Additionally, in 2-step PSA, it implied that the adsorbed CO2 gas of 276.8 Nm3/h (0.44 Nm3/kg-Feedstock) can be obtained and the gas would be available for the agricultural facilities as a fertilizer. Note that CO2 can be sequestrated including the small amount of CH4 and CO from the 1st stage reactor. That is, it is assumed that CH4 and CO can be recovered due to a combustion process.

2.2 Estimation of H2S removal

Next, the performance influence due to contaminants in H2 fuel has been analyzed. In general, the Bio-H2 of product has the contaminants of H2S and/or HCl etc., which impacts the operating of application. For instance, in the case of H2S contamination, it was reported that the following matters in the operation of polymer electrolyte fuel cell (PEFC) occurred: On the performance test of PEFC with the membrane of Nafion 115 and the electrode catalyst of Pt-Ru/Pt, the voltage drops at 500 mA/cm2 were 8 mV of 0.5 ppm, 14 mV of 1.0 ppm, and 25 mV of 2.0 ppm, respectively \(^\text{14}\). Thus, the more suitable system which contributes to the LCO2 mitigation is necessary. From the viewpoint of LCA thinking, on the countermeasure, it would be better to have the some functions by which the benefit can be gained. In 2-step PSA, the CO2 gas can be captured at the 1st stage. If the contaminant of H2S can be removed from product fuel at the same stage, the any additional input energy might not be necessary.

Using the following experimental apparatus, the potential of H2S removal in 2-step PSA was estimated (see Fig. 2). The experimental conditions are as follows: (1) the sample gas which consists of H2S of 30 ppm and CO2 balance was used, and flow rate of 10 ml/min was fed, (2) the inner temperature and pressure in the reactor was 40 degC and 0.4 MPaG, respectively, and (3) HAS-Clay of 2 g was inserted in the reactor as an adsorbent. Note that N2 gas is for cleaning in the reactor and that the experimental conditions are still not optimized. That is, the volume of HAS-Clay, the inner temperature and pressure might not be suitable. However, from the viewpoint of LCA thinking, the lower temperature and the small pressure change would be better since the additional energy is not required.

Fig. 3 shows the capability of H2S removal in use of HAS-Clay. What is important about this experiment is that the pressure and temperature are the same as that of PSA operation. H2S removal rate in this result is defined by Eq. (2). The index of H2S recovery rate was measured every 20 min. in use of a gas chromatography (Shimadzu, GC-8A).

\[
\text{H2S removal efficiency} \% \quad = \quad 1 \quad - \quad \frac{\text{Permuted volume of H2S}}{\text{Inlet volume of H2S in sample gas}} \quad (2)
\]

This result implies that H2S besides CO2 can be adsorbed by HAS-Clay simultaneously. Probably, the rate would be 30 to 40%. Additionally, due to the combination of metal oxide (e.g. CuO or Fe2O3), H2S would be removed entirely. For instance, Liu, D. et al. reported that CuO-based adsorbents were able to remove H2S from a CO2 stream to less than 0.1 ppm at 40 degC (e.g. CuO of 177 mg/g) \(^\text{15}\). Consequently, the combination of physical adsorption due to HAS-Clay and chemical adsorption of CuO might be able to reach at the level of almost 0 ppm. Also, the pre-adsorption of HAS-Clay would expand the life-time of CuO. However, this time, since the optimal condition is still not obtained.
the LCCO₂ estimation was analyzed without any influence of contaminants.

2.3 H₂ storage and power supply due to EDLCs

Next, on performance of H₂ storage and usability of a FC-cell phone application (e.g., mobile device), the following systems are assumed on basis of basic performance tests.

First, in this study, Bio-H₂ storage was assumed to be executed by the metal hydride of LmNi₅₀Mn₃₀Al₂₀Pd₁₈ (Japan Metals & Chemicals Co., Ltd.). This metal hydride is commercial released product, whose adsorption ability has 157 NL/kg-metal. In general, metal hydride materials have many species, whose abilities have various properties from 0.5 to 10.0 wt.% of H₂. These properties are due to the combination of metals by which adsorption/desorption are affected. That is, since H₂ can be stored in metal hydrides under moderate temperature and pressure, these are the promising candidates due to their safety advantage with high volume storage capacity for on-board applications. However, the selection of appropriate metal should be carried out so as to meet the conditions (e.g., the duration time of application, or the weight etc.) of users. Note that the design of adequate storage will be an issue in the future.

While, in our previous study, the electrical consumption of mobile device was measured in consideration of operational duration of user behavior in each generation. For instance, the maximum consumption was 5.8 Wh/d in a holiday and 3.24 Wh/d at 20s, while the minimum was 2.4 Wh/d in a holiday at over 50s.

On the suitable weight of metal hydride, the advantage of operational duration per a charge and the weight of device etc. should be considered in comparison to conventional ones. Thus, our targets are to expand the duration from a day to approximately a week, and to keep the same weight. In the conventional device, the battery weight of Li-ion was approximately 50 g with 1,400 mAh. However, since our development is still at the initial stage, the weight of metal hydride was 45.76 g. In this case, the capability of storage was 7.12 NL on pure H₂. On the inlet status of storage vessel, H₂ was adsorbed at 0.4 MPaG and 25 to 30 degC. The storage efficiency against the inlet H₂ concentration was 99.9%, since the H₂ concentration through 2-step PSA was 99.96%. Also, the residual gases of H₂ and CH₄ can be recycled as off-gas fuel for the gasifier operation. While, the re-pressurizing of H₂ fuel at time of storage should be necessary due to the pressure drop in the PSA. Considering these points, in the case of 2-step PSA, the product rate was 302.0Nm³/h with the efficiency of 38.7 %-LHV and the auxiliary power of 239.5 kW (PSA of 127.2 kW).

Next, in the use phase of mobile device, the following problems have to be solved. In general, the power consumption of electronic devices rapidly changes due to communication and user interface devices that include wireless communication and LCD screen. Sometimes, the load waveforms include spikes for intermittent functions. FC device is known to have poor load-following performance due to large over-potential, and they can be easily corroded under load-varying conditions for electrochemical and mechanical reasons. Thus, a hybrid power source system with electric double-layer capacitors (EDLCs) which is one solution to compensate load following performance and improve FC lifetime was considered in this study. The combined EDLCs with the FC have been recently developed and already used in practical phase.

In our previous study, the performance of FC-EDLCs was investigated through device tests. The electrical demands including load waveforms on basis of the functions of a cell phone were measured. Here, the functions of a cell phone are internet browsing, messaging, game, phone call, music player and stand-by. On the specification of FC, the cell area of 1 cm² and the number of stack of 4 cells were assumed. The energy conversion efficiencies of FC operation (ηFC), EDLCs performance (ηEDLCs) and total efficiency (η = ηFC × ηEDLCs) were estimated on LHV basis in Table 5.

Compared to the conventional Li-ion battery, it is implied that the performance of FC-EDLCs for a cell phone is significantly improved due to these energy conversion efficiencies. That is, based on the energy demand of each function of a cell phone, the duration per a charge can be expanded up to 4.0 to 7.0 days. Also, the total weight of FC-EDLCs with H₂ storage would be approximately 70 g. However, the weight loss would be sufficiently possible through an optimized manufacture, since this level is at the initial stage.
3. Results and Discussion

3.1 Results of the LCCO\textsubscript{2} of Bio-H\textsubscript{2}

Next, based on the former discussions, the LCCO\textsubscript{2} of Bio-H\textsubscript{2} was evaluated. Here, the specific CO\textsubscript{2} emission of Bio-H\textsubscript{2} fuel \( FCO_2 \) \([\text{g-CO}_2/\text{MJ-H}_2]\) was defined in Eq. (3).

\[
FCO_2 = \frac{C_1 + C_2 + C_{aux} - \alpha C_{fs}}{Q_{H_2}}
\]

where, \( C_1, C_2, C_{aux}, \) and \( C_{fs} \) are CO\textsubscript{2} emission of a compressor at the first reactor on basis of electricity consumption \([\text{g-CO}_2/\text{h}]\), CO\textsubscript{2} emission of a compressor at the second reactor \([\text{g-CO}_2/\text{h}]\), auxiliary CO\textsubscript{2} emission of the storage compressor for H\textsubscript{2} and the feedstock pre-treatment \([\text{g-CO}_2/\text{h}]\), and the carbon content of feedstock expressed as CO\textsubscript{2} emission \([\text{g-CO}_2/\text{h}]\), respectively. In detail, \( C_{fs} \) is the sequestrated CO\textsubscript{2} emission. The coefficient \( \alpha \) \((0.0 \leq \alpha \leq 1.0)\) is the percentage of adsorbed CO\textsubscript{2} gas that is potential for being taken as a growth agent in the cultivation of agricultural products and is affected by CO\textsubscript{2} adsorption performance. In case of \( \alpha > 0 \), \( FCO_2 \) might be a negative emission.

In LCA methodology, the GHG emissions balance is discussed for the target system. In this case, the biomass feedstock has a carbon neutral impact. It is assumed that the carbon of biomass origin is sequestrated by 2-step PSA unit and that the adsorbed CO\textsubscript{2} is used for fertilizer in the greenhouse facility of vegetables or fruits cultivation. This means that CO\textsubscript{2} can be fixed through photosynthesis additionally. Consequently, the net balance is estimated as the emission that subtracted the available amount of CO\textsubscript{2} for agricultural product cultivation by the total indirect CO\textsubscript{2} emission on basis of LCA methodology, considering the carbon neutrality. The estimated emission is defined as negative emission in the case of the minus value in Eq. (3).

Here, \( \alpha \) was assumed to be 1.0 in this study\textsuperscript{10}. As a result, \( FCO_2 \) in a conventional PSA \((\alpha = 0)\) was 68.4 g-CO\textsubscript{2}/MJ-H\textsubscript{2}, and that in 2 step PSA \((\alpha = 1)\) was -101.3 g-CO\textsubscript{2}/MJ-H\textsubscript{2}. Note that the specific CO\textsubscript{2} emission of conventional electricity was 496 g-CO\textsubscript{2}/kWh as of 2014 in Tokyo, Japan\textsuperscript{20}. Using these emission results, the CO\textsubscript{2} emissions on basis of cell phone users’ behavior were estimated. Here, the users’ behavior is expressed by the used hours of each function which was obtained by the questionnaire. The indirect CO\textsubscript{2} emissions on application manufacturing and/or materials are affected by a period of use. In this study, that was assumed to be 3.7 years as the same in each generation on basis of the questionnaire. The energy demands for a period of use were between 3.50 and 5.10 Wh/d in weekday and between 2.68 and 5.83 Wh/d in holiday. In general, the younger generation was likely to use a cell phone frequently\textsuperscript{19}.

Fig. 4 shows the LCCO\textsubscript{2} emissions in each generation. Compared to a conventional cell phone with Li-ion battery, the effect of CO\textsubscript{2} capture would be beneficial in all generation. For instance, the H\textsubscript{2} fuel without CO\textsubscript{2} capture has no contribution to LCCO\textsubscript{2} abatement in under 20’s, even if biomass feedstock is used. However, in over 30’s, LCCO\textsubscript{2} abatement rate was 13.5% and this case was the most effective.

3.2 Discussions

This time, the performance of Bio-H\textsubscript{2} production in consideration of CO\textsubscript{2} capture was designed and the LCCO\textsubscript{2} emission was estimated for the FC-cell phone. In general, for a small device with FC which is operated by the eco-friendly H\textsubscript{2} fuel, the effect of CO\textsubscript{2} emission reduction would be small. Because the indirect CO\textsubscript{2} emissions attributed to
a device manufacture of parts and/or materials represent large percentage in comparison with the direct emission. Thus, if the large benefit such as negative emission is obtained to some extent, it would be difficult to contribute to the CO$_2$ abatement for a small device. That is, the negative emission is extremely significant.

On another system with CO$_2$ capture, Susmozas A. et al. estimated the CO$_2$ capture of 0.24 Nm$^3$/kg-Feedstock, which is equivalent to 141.2 g-CO$_2$/MJ. In this system, the production efficiency is around 20% $^5$). Based on this system performance, consequently, the LCCO$_2$ emission of H$_2$ was -24.2 g-CO$_2$/MJ, and the abatement rate against the emission of a conventional cell phone with Li-ion battery was approximately 11.0%. Note that the CO$_2$ emission on pre-treatment of feedstock was assumed to be the same as our estimation condition.

Referring to the results, Bio-H$_2$ with the negative LCCO$_2$ emission has sufficient possibility to contribute to the global warming protection. In this case, the most important points are to keep productivity of H$_2$ fuel and to reduce the energy consumption at the same time. Here, the Bio-H$_2$ production system which can contribute to CO$_2$ reduction was proposed. At the next step, on the usage of sequestrated CO$_2$ emission, the beneficial path would be considered.

4. Conclusions

The current study discussed the production of hydrogen fuel from biomass feedstock. In this system, it was described that not only CO$_2$ emission reduction but also H$_2$S removal were able to be achieved without any production loss. Also, the FC performance for a cell phone including H$_2$ storage capability was referred, and based on the estimation result, the CO$_2$ abatement benefit of a cell phone was discussed.

From the viewpoint of users’ preferences, the expansion of operational duration and/or the weight loss of a device should be most beneficial. Meanwhile, for the contribution of CO$_2$ mitigation the most important point is to achieve negative emissions. This aspect implies that the more benefit on eco-friendliness is obtained.

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