Steam based biomass gasification processes for syngas and hydrogen production

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Heating Applications of Bio-Pellet Made from Ecological-Hazard Plant in Small and Medium Enterprises to Enhance Utilization of Renewable Energy in the APEC Region

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- **Potential of Hydrogen**

- **FCEV fleet (Targets) announced by hydrogen initiatives (2020)**
  - **Europe:** ~350,000
  - **Japan:** 100,000
  - **Korea:** 50,000
  - **United States:** ~20,000

**Source:** Technology Roadmap Hydrogen and Fuel Cells
OECD/IEA, 2015 International Energy Agency
BLUE Tower II – Third pilot plant
(Shibukawa Technology Development Center of JBEC*)
*Japan Blue Energy Co. Ltd.
Our Strategy - R&D of Fuel Cell Drone by Bio-H₂ fuel supply -

1. Combination of Bio-H₂ production technology and a cell phone
2. Expansion of Bio-H₂ application.

**Prof. K. Dowaki** (Project leader): Gasification technology, Gas Cleaning Technology · LCA

**Prof. T. Gunji**: Removal technology of contaminants in Bio-syngas

**Prof. M. Hayase**: Small scale PEFC in consideration of CO and H₂S tolerance

**Dr. N. Katayama**: H₂ storage system (metal hydride) · power converter circuit

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**Biomass Feedstock**  →  **Bio-H₂** (Gasification)  →  **H₂ storage (cartridge)**  →  **Pure H₂**  →  **MH**  →  **Fuel Cell**  →  **Drone**

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Heating Applications of Bio-Pellet Made from Ecological-Hazard Plant in Small and Medium Enterprises to Enhance Utilization of Renewable Energy in the APEC Region

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Gasification Technology

Basic contents

- The gasifier type is the biomass pyrolysis system in a reductive atmosphere.
- The system has 2 furnaces of pyrolysis+reforming zone and pre-heater.
- H₂ concentration in syngas would be comparatively high (>40 vol.%).
- The plant capacity would be the range of 5 to around 70 t/day.

Our tasks in this study

- Confirmation of syngas yields and concentrations due to experiments.
- Confirmation of adsorption performance due to experiments. (CO₂ adsorption / H₂ purification)
- Process design of H₂ production system and estimations of production rate and auxiliary power
- Removal of impurities in syngas.
- Calculation of Eco-footprint of Bio- H₂ on basis of LCA concept.
In the previous LCA study on BT process design in use of the simulator of ASPEN-PLUS, we clarified that the highest impact energy consumption is due to the auxiliary power of PSA.

Reduction of energy consumption by our proposals will be achieved.

**Reduction of auxiliary in Bio-H₂ plant**

- In the previous LCA study on BT process design in use of the simulator of ASPEN-PLUS, we clarified that the highest impact energy consumption is due to the auxiliary power of PSA.

Reduction of energy consumption by our proposals will be achieved.

**Total: 280 kW**
Development of 2-step PSA

Performance test in use of “2-step PSA apparatus”

1. Confirmation of gaseous yields and concentrations
2. Preparation of bio-syngas
3. Execution of adsorption tests
   - Different species of absorbent (form and pore)
   - 2-step PSA controls (pressure and temperature)
   - Combination of absorbents

a. Power saving due to low pressure/low temperature controls and a reduction of amount of a working gas fluid (ex. 0.8MPaG->0.5MPaG, CO₂ removal)
b. Achievement of CO₂ adsorption and H₂ purification with higher efficiencies
Fabrication of apparatus (Gasification Process)

- Confirmation of the performance
- Data acquisition for the static modeling (AspenPlus)

<table>
<thead>
<tr>
<th>Proximate Analysis</th>
<th>Waste W</th>
<th>J. cedarc (Reference)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Moisture</td>
<td>6.02</td>
<td>wt.% 37.70</td>
</tr>
<tr>
<td>Ash</td>
<td>0.35</td>
<td>wt.% 2.20</td>
</tr>
<tr>
<td>Volatiles</td>
<td>82.92</td>
<td>wt.% 83.50</td>
</tr>
<tr>
<td>Fixed carbon</td>
<td>10.71</td>
<td>wt.% 14.30</td>
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</table>

<table>
<thead>
<tr>
<th>Ultimate analysis</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon</td>
</tr>
<tr>
<td>Hydrogen</td>
</tr>
<tr>
<td>Nitrogen</td>
</tr>
<tr>
<td>Oxygen</td>
</tr>
<tr>
<td>Chlorine</td>
</tr>
<tr>
<td>Sulfur</td>
</tr>
</tbody>
</table>

Measurement of gaseous yields

Note: This apparatus consists of pyrolysis zone and reforming zone.
Effects of pyrolysis and reforming temperatures

Performance (summary)

<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>550</td>
<td>550</td>
<td>1.02</td>
<td>0.2</td>
<td>0.02</td>
<td>71</td>
</tr>
<tr>
<td>550</td>
<td>700</td>
<td>1.10</td>
<td>0.2</td>
<td>0.04</td>
<td>68</td>
</tr>
<tr>
<td>900</td>
<td>950</td>
<td>1.19</td>
<td>0.2</td>
<td>0.008</td>
<td>73</td>
</tr>
</tbody>
</table>
**Fabrication of apparatus (2-step PSA)**

- Confirmation of the performance
- Data acquisition for the dynamic modeling (gPROMS)

- Adsorbent volume: 200 ml
- Opt. Temp.: RT
- Opt. P: 0.4 MPaG
- Feed gas: H₂ 55%, CO 20%, CH₄ 8%, CO₂ 17%
- Opt. time: 5 min

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**Measurement of PSA performance**

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Specification</td>
<td>SiO₂ · Al₂O₃ · H₂O</td>
<td>MeO · Al₂O₃ · mSiO₂ · nH₂O (Me: Cation)</td>
<td>Carbon</td>
<td>Carbon</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Chemical composition</td>
<td>&gt;2.0</td>
<td>2.36-4.75</td>
<td>2.36-4.75</td>
<td>2.36-4.75</td>
<td>2.39-4.75</td>
<td>4-5.5</td>
<td>2.36-4.75</td>
</tr>
<tr>
<td>Particle size [mm]</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pore diameter [nm]</td>
<td>&lt;0.02</td>
<td>0.3</td>
<td>0.4</td>
<td>0.5</td>
<td>0.9</td>
<td>0.5-10</td>
<td>0.5</td>
</tr>
</tbody>
</table>
Result of adsorption test (1)

HAS-clay

- Only CO$_2$ permeability was worse in comparison to the other gas components.
- That of H$_2$ was relatively higher.

Zeolite A-5

- The gaseous components percolated in the absorbent were H$_2$ and CH$_4$.
- H$_2$ permeability was highest among the candidates.
- The concentration of H$_2$ which passed through Zeolite A-5 was 90% or more.

Our proposals

- At the 1$^{\text{st}}$ step, HAS-clay is employed in order to adsorb CO$_2$ selectivity.
- At the 2$^{\text{nd}}$ step, Zeolite A-5 is employed in order to purify H$_2$. 
In order to make only CO$_2$ component separated from HAS-clay at 1$^{st}$ step, the breakthrough curve was measured.

**Result of adsorption test (2)**

In order to make only CO$_2$ component separated from HAS-clay at 1$^{st}$ step, the breakthrough curve was measured.

**Condition**

- Gas flow rate
  \[(\text{Syngas}(10\%)+\text{Ar}(90\%)) : 13.1 \text{ NL/min}\]
- Operating pressure
  \[0.4 \text{ MPaG}\]
- Operating temperature
  \[\text{Atmospheric (28 to 32 deg.C)}\]
- Amount of HAS-clay
  \[200 \text{ cc}\]

- H$_2$ and CO$_2$ are likely to be separated individually by the combination of HAS-clay at 1$^{st}$ step with Zeolite A-5 at 2$^{nd}$ step, and the gas flow control in consideration of CO$_2$ retention time.
Based on our experimental results, we designed the process of target system. We argued the optimal condition in consideration of LCA concept.

<Definition of Eco footprint>

\[ \eta_{\text{energy}} = \frac{Q_{H_2}}{W_1 + W_2 + H_1(T_1) + H_2(T_2) + Q_{\text{syn}} - Q_{\text{og}}} \]

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

The coefficient of \( \alpha \) can be set up by the test of CO\(_2\) assimilation in use of model plant (e.g. Arabidopsis thaliana).

- In the case of the combination of HAS-clay with Zeolite A-5,
  ✓ \( \eta_{\text{energy}} \): 82.8\%, \( FCO_2 \): -56.6 g-CO\(_2\)/MJ-H\(_2\) (164kW→112kW, Reduction by 31%)

- In the conventional case\(^{(1)}\),
  ✓ \( \eta_{\text{energy}} \): 79.0\%, \( FCO_2 \): 57.4 g-CO\(_2\)/MJ-H\(_2\)

\( Q_{H_2} \): LHV of H\(_2\) [MJ/h]
\( Q_{\text{syn}} \): LHV of syngas [MJ/h]
\( Q_{\text{og}} \): LHV of off gas [MJ/h]
\( W_1 \): first step of PSA [MJ/h]
\( W_2 \): second step of PSA [MJ/h]
\( H_1(T_1) \): sensible heat of first reactor [MJ/h]
\( H_2(T_2) \): sensible heat of first reactor [MJ/h]
\( C_1 \): CO\(_2\) emission of first reactor [g-CO\(_2\)/h]
\( C_2 \): CO\(_2\) emission of second reactor [g-CO\(_2\)/h]
\( C_{\text{aux}} \): CO\(_2\) emission of auxiliary [g-CO\(_2\)/h]
\( C_{fs} \): CO\(_2\) emission due to carbon neutral [g-CO\(_2\)/h]
\( \alpha \): the percentage of adsorbed CO\(_2\) gas [-]

\(^{(1)}\) Y. Watanabe et al. :\textit{J. Life Cycle Assessment, Japan, 9}(1), 20-36 (2013)
Dynamic Simulation of 2-step PSA

- 1st stage, CO$_2$ adsorption
- 2nd stage, H$_2$ purification
**Modeling**

**Mass balance**

\[
\varepsilon_{\text{bed}} \frac{\partial C_{g,i}}{\partial t} = \frac{\partial}{\partial z} \left( \varepsilon_{\text{bed}} D_{ax,i} \frac{\partial C_{g,i}}{\partial z} \right)
\]

Non constant

\[
-u \frac{\partial C_{g,i}}{\partial z} - \left(1 - \varepsilon_{\text{bed}}\right) a_p k_f \left(C_{g,i} - C_{s,i}\right)
\]

Diffusion
Convection
Adsorption

**Heat balance**

Consideration of heat transfer between surface of inner vessel and gas flow.

\[
\varepsilon_{\text{bed}} C_g \rho_g + \rho_{\text{bed}} C_p s \frac{\partial T}{\partial t} = -\frac{\partial}{\partial z} \left( K_L \frac{\partial T}{\partial z} \right) + \varepsilon_{\text{bed}} C_g C_p g \frac{\partial T}{\partial z} u
\]

Non constant

\[
+ \rho_{\text{bed}} \sum_{j=1}^{\text{Comp}} \Delta H_j \frac{\partial q_j}{\partial t} + \frac{2 h_{w}}{R_{\text{in}}} (T - T_{\text{wall}})
\]

Diffusion
Convection
Heat generation
Surface –gas flow

**Pressure loss (Blake-Kozeny)**

\[
-\frac{\partial P}{\partial z} = 180 \frac{\mu(1 - \varepsilon_{\text{bed}})^2 \varepsilon_{\text{bed}}^3}{D_p^2}
\]

**gPROMS** (finite element method)
**Simulation conditions**

- PSA Cycle: Compression->Refinery->Decompression->Purge
- Number of cycles (in this simulation): 3

Note: Simulation parameters (physical properties) are used by the reference data [1][2]

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Fig. PSA cycle

Results at 1st stage (HAS-Clay)

Confirmation of selective CO₂ adsorption
- Recovery efficiencies of H₂, CO and CH₄ are higher.
- Conc. of CO₂ is high in purge process.
  - CO₂ : 97.47% (adsorption efficiency)
- Temperatures in compression and refinery processes increase.
  - Physical adsorption

<table>
<thead>
<tr>
<th></th>
<th>H₂</th>
<th>CO</th>
<th>CH₄</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow rate 1st [%]</td>
<td>88.30%</td>
<td>88.42%</td>
<td>82.09%</td>
</tr>
</tbody>
</table>

![Graph showing composition over time](image-url)

![Graph showing inner temperature over time](image-url)
✓ Results at 2\textsuperscript{nd} stage (Zeolite A5)

- $\text{H}_2$ Conc. : 99.18%
- $\text{H}_2$ recovery eff. : 78.14%

- Advantage point is to improve the product quality (concentration) in comparison to the conventional PSA without CO\textsubscript{2} adsorption
- Reduction of auxiliary power: approx. 30\% abatement $^{[1]}$

<table>
<thead>
<tr>
<th></th>
<th>$\text{H}_2$</th>
<th>CO</th>
<th>CH\textsubscript{4}</th>
<th>CO\textsubscript{2}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow rate 2\textsuperscript{nd} [%]</td>
<td>99.18%</td>
<td>0.77%</td>
<td>0.04%</td>
<td>0.003%</td>
</tr>
</tbody>
</table>

Only $\text{H}_2$ gas can pass through. Sequestration of $\text{H}_2$ can be achieved.

Impurities problems on the operation by Bio-\( \text{H}_2 \)?

- Contaminations of bio-syngas: HCl, H\( \text{2} \)S, NH\( \text{3} \) etc.
- FC applications→Polymer Electrolyte Fuel Cell (PEFC) (present)

1. HCl (Hydrogen chloride)
   - Pt dissolution (Use of catalyst on FC electrodes)
2. H\( \text{2} \)S (Hydrogen sulfide)
   - Performance drop

Fig. Effect of current density on the performance deterioration during exposure to 20 ppm H\( \text{2} \)S/H\( \text{2} \). \( T_{\text{cell}} \) =70 °C, humidification temperatures of the anode and cathode: room temperature and 70 °C.

Where shall the impurities be removed?

- Forethought of operation profile in the plant
- Minimum consumption of adsorbents (Economic and Environment)

**Process design**

Diagram:
- Biomass → Pyrolyzer → Steam
- Dryer → Pre-heater → HC (Circulation) → Reformer → Syngas
- Hot Gas → Combustor → Hot Flue Gas
- Blower → HX-3
- HX-1: Heat exchanger
- Comp.: Compressor
- WGS: Water gas shift reactor at a high temperature (option)
- Conventional case
- Proposed case

Notation:
- HX: Heat exchanger
- HC: Heat carrier
- WGS: Water gas shift reactor at a high temperature (option)
- Comp.: Compressor

Process steps:
1. Bio-H₂ (FC application)
2. H₂S removal (Metal Oxide)
3. 1st stage (HAS-Clay)
4. 2nd stage (Zeolite A-5)

Water gas shift reactor at a high temperature (option):
- Conventional case
- Proposed case

H₂S / HCl removal:
- H₂S (HAS-Clay) / HCl (HAS-Clay+CaCO₃)

Bio-Pellet Made from Ecological-Hazard Plant in Small and Medium Enterprises to Enhance Utilization of Renewable Energy in the APEC Region

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**Estimation of breakthrough curve (Removal Tests)**

**Adsorbent candidates**

<table>
<thead>
<tr>
<th>Specification</th>
<th>HAS-Clay</th>
<th>Zinc oxide</th>
<th>Iron oxide</th>
<th>Calcium carbonate</th>
</tr>
</thead>
<tbody>
<tr>
<td>Target Impurities</td>
<td>H₂S/HCl</td>
<td>H₂S</td>
<td>H₂S</td>
<td>HCl</td>
</tr>
<tr>
<td>Chemical composition</td>
<td>SiO₂·Al₂O₃·H₂O</td>
<td>ZnO</td>
<td>Fe₂O₃</td>
<td>CaCO₃</td>
</tr>
<tr>
<td>Particle size [mm]</td>
<td>2.2-3.35 (H₂S)</td>
<td>2.2–3.35</td>
<td>2.2–3.35</td>
<td>12×10⁻³-15×10⁻³</td>
</tr>
<tr>
<td>Pore diameter [nm]</td>
<td>&lt;0.02</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Note: HAS-Clay means synthetic substances of hydroxyl aluminum silicate and clay.

(a) H₂S  
(b) HCl
Absorption performance of HAS-Clay (Results of H$_2$S (1))

**Condition**

1. H$_2$S: 30 ppmv, flow rate: 50 Nml/min., 40 deg.C and 0.4 MPaG,
   Sample weight: HAS-Clay (2 g), Space velocities (SV): 2,915 h$^{-1}$
2. H$_2$S: 100 ppmv, flow rate: 250 Nml/min., 40, 80, 120 deg.C and atmospheric pressure, Sample weight: ZnO (1.3 g) or Fe$_2$O$_3$ (0.95 g), Space velocities (SV): 8,784 h$^{-1}$ (constant)

(a) Physical Adsorption (HAS-Clay)

(b) Physical Adsorption (Zeolite A-5)

**Note:**

- In 2-step PSA (the combination of HAS-Clay and Zeolite A-5), almost H$_2$S can be eliminated.
- Prevention of H$_2$S+CO$_2$=COS+H$_2$O because of an absorption of CO$_2$ by HAS-Clay.
- Chemical adsorption is a complementary use.
Estimation of breakthrough curve (Results of H$_2$S(2))

(a) Chemical Adsorption (ZnO)

(b) Chemical Adsorption (Fe$_2$O$_3$)

Sulfur capture capacity (Definition)

\[ S_{\text{cap}} = \frac{t_{\text{BT}} \times \dot{V} \times C_{\text{PG}} \times A_{\text{as}}}{V_m \times W_{\text{sorbent}}} \times 100 \]

- \( t_{\text{BT}} \): Breakthrough time [min]
- \( \dot{V} \): Flow rate [L/min]
- \( C_{\text{PG}} \): Conc. of pollutant [ppmv]
- \( V_m \): Molar volume [L/mol]
- \( A_{\text{as}} \): Atomic weight (=32.07)
- \( W_{\text{sorbent}} \): Adsorbent weight [g]

Table Sulfur capture capacity

<table>
<thead>
<tr>
<th>Temp. [°C]</th>
<th>ZnO*</th>
<th>Fe$_2$O$_3$*</th>
</tr>
</thead>
<tbody>
<tr>
<td>40</td>
<td>0.24</td>
<td>0.54</td>
</tr>
<tr>
<td>80</td>
<td>0.31</td>
<td>1.65</td>
</tr>
<tr>
<td>120</td>
<td>0.28</td>
<td>1.69</td>
</tr>
</tbody>
</table>

*Unit: g-S/100 g-sorbent
Absorption performance of HAS-Clay (Results of HCl(1))

**Condition**

1. HCl: 1,160 ppmv, Flow rate: 200 Nml/min, 100-200 deg.C and atmospheric pressure
   Sample weight: HAS-Clay (0.1 g) and/or CaCO₃ (1.0 g)
   Space velocities (SV): 3,725-32,000 h⁻¹ (see Table)

2. Estimation of the effect of blend effect vs. mono-adsorbent.

\[
\text{CaCO}_3 + \text{HCl} \rightarrow \text{H}_2\text{O} \downarrow + \text{CO}_2 \downarrow \quad \text{(adsorption effect of HAS-Clay)}
\]

Note: In general, Ca based adsorption is worked at the temp. of >600 deg.C.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>#1</th>
<th>#2</th>
<th>#3</th>
</tr>
</thead>
<tbody>
<tr>
<td>CaCO₃ [g]</td>
<td>1.00</td>
<td>0.00</td>
<td>1.00</td>
</tr>
<tr>
<td>HAS-Clay [g]</td>
<td>0.00</td>
<td>0.10</td>
<td>0.10</td>
</tr>
<tr>
<td>Space Velocity [h⁻¹]</td>
<td>4,216</td>
<td>32,000</td>
<td>3,725</td>
</tr>
</tbody>
</table>

Based on the experimental results, the estimation of adsorption effect of blend case (Cl capture) was carried out.
Estimation of breakthrough curve (Results of HCl (2))

(a) CaCO$_3$

(b) HAS-Clay

(c) Blend

Table Chlorine capture capacity

<table>
<thead>
<tr>
<th>Temp. [°C]</th>
<th>CaCO$_3^*$</th>
<th>HAS-Clay$^*$</th>
<th>Blend$^*$</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.06</td>
<td>4.18</td>
<td>0.52</td>
</tr>
<tr>
<td>150</td>
<td>0.08</td>
<td>3.32</td>
<td>0.52</td>
</tr>
<tr>
<td>200</td>
<td>0.11</td>
<td>1.79</td>
<td>0.70</td>
</tr>
</tbody>
</table>

*Unit: g-Cl/100 g-sorbent
Impact analysis

Purpose
1. Due to reduction of amount of chemicals (adsorbent), the eco-burden in consideration of LCA is to be visualized.

Conditions
1. Functional unit: 1 Nm³ Bio-H₂ (4N)
2. Estimated Index: Abiotic metal depletion potential (ADP), global warming potential (GWP)
3. Software: SimaPro 8.2 software (Impact analysis: the Centrum voor Millieuwetecne Leiden (CML) method
4. Conventional case (H₂S): ZnO (Conv. ZnO) (Capture capacity: 2.81 g-S/100 g-sorbent)
5. Initial concentration (H₂S): 200 ppmv
6. Proposed case: 2-step PSA+ZnO (0.24 g-S/100 g-sorbent), PSA+Fe₂O₃ (0.54 g-S/100 g-sorbent) *Note: Temp. 40 deg.C
7. Conventional case (HCl): Na₂CO₃ (Conv. Na₂CO₃ ) (Capture capacity: 5.35 g-Cl/100 g-sorbent)
8. Proposed case (Blend case): HAS-Clay+CaCO₃ (0.70 g-Cl/100 g-sorbent)
9. HAS-Clay is assumed to be regenerable.
10. Calculation

Impact Potential = Inventory Value × Characterisation Factor
Estimation of GWP and ADP

(a) $\text{H}_2\text{S}$

(b) $\text{HCl}$

Advantages due to HAS-Clay use

- The eco-indexes of GWP and/or APD was remarkably improved in comparison to the conventional cases ($\text{H}_2\text{S}$).
- In the case of blend of HAS-Clay and $\text{CaCO}_3$, GWP and ADP in comparison to the conventional case were improved by 25.4% and 19.4%, respectively ($\text{HCl}$).
Conclusions

- In our group, Bio-H\textsubscript{2} production system using the gasification process of Blue Tower is developing.
- The design policy is based on LCA indexes to obtain the eco-benefit of biomass feedstock.
- Looking at the entire system, the key technology is the reduction of external energy consumption besides an assurance of steady operation.
- Also, the technology of impurities removal would be extremely important to match a fuel specification for FC application.
- In this case, the candidates have to be selected appropriately, since the adsorbents are associated with eco-burden.
Conclusions (continued)

- HAS-Clay which we concerned in this study has good potentials to reduce the auxiliary power and eliminate the impurities in product gas.
- The characteristic of HAS-Clay, especially, CO$_2$ adsorption, can promote the elimination of H$_2$S and HCl, too.
- Simultaneously, the consumption of metal oxide would be reduced due to these characteristics.
- Using the eco-index on basis of LCA (GWP and ADP), the obvious advantages can be obtained.
- In our future tasks, the adsorption performance of other impurities (e.g. NH$_3$) will be analyzed.
Thank you for your attention!