Optimization of Heat Transfer through Heat Carriers in Bio-H₂ Production Using CFD Simulation

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Hydrogen (H₂) is one of the most promising secondary energy resources expected to contribute to the prevention of global warming. Bio-H₂, which is derived from biomass feedstock, is more environmentally friendly than hydrogen synthesized from fossil fuels. In our indirect thermochemical processes with solid-gas reactions, the effective heat transfer, which is accomplished by circulation of alumina balls acting as heat carriers (H Cs), is a critical issue, and it is necessary to achieve circulation of the heat transfer medium that maximizes production efficiency.

In this study, the heat transfer performance of a small-scale indirect biomass gasification process to be promoted in the near future was investigated, focusing on heating of HCs using high-temperature gas to achieve optimum heat utilization in the preheating reactor. To continuously pyrolyze cedar feedstock in the subsequent pyrolysis reactor, HCs must be heated to the desired temperature in the preheater within a residence time. Even if high-temperature gas with sufficient calorific value flows into the preheater, if the heat transfer is not completed within the residence time, the HCs will not be heated to the target temperature and the sensible heat is discharged as tail gas. Therefore, we evaluated the operating conditions that promote the heat transfer during the residence time, focusing on the hot flue gas conditions.

Key Words
Bio-H₂, Indirect biomass gasification, CFD modeling

1. Introduction
Hydrogen (H₂) is considered a clean energy resource that can be used without any CO₂ emissions, and its use is expected to expand. However, fossil fuels are currently the major source of hydrogen, resulting in environmental problems. Therefore, alternative approaches for hydrogen production are needed to provide cleaner energy sources. Biomass feedstock is a renewable resource that does not increase greenhouse gas (GHG) emissions because of its carbon-neutral lifecycle.

We focused on the indirect biomass gasification process as a methodology to convert biomass to hydrogen. This process can easily convert solids into gas and prevents tar-related problems, such as clogged pipes. In our indirect thermochemical processes with solid-gas reactions, the
effective heat transfer that is carried out by the circulation of alumina balls acting as heat carriers (HCs) is a critical issue\(^7\). It is necessary to achieve the circulation of HCs that maximizes production efficiency, because the production rate is strongly affected by the heat exchange performance. Note that any improvement in production efficiency is directly related to the environmental impact caused by Bio-H\(_2\) production. Therefore, it is important to reduce any heat losses during heat transfer in each vessel.

In our process, alumina balls acting as HCs were used to exchange heat. During the process, the balls are heated to the designed temperature in the preheater, and are dropped into the pyrolyzer, in which heat energy is discharged to compensate for the pyrolysis reaction heat. They return to the preheater using a conveyor (see Fig. 1). Considering the heat transfer performance of the pyrolyzer, HCs must be heated to a target temperature in the preheater. Note that our process is characterized by its small scale, and we adopt small reactors for each vessel.

To achieve the heat transfer medium that maximizes production efficiency while preventing heat loss, the thermal profiles of the preheater were investigated, and suitable operating conditions were discussed. For the investigation, it was assumed that the heat carriers were packed in the preheater in a stationary state during their residence time. To reduce any heat loss during heat transfer, the HCs in the preheater must be heated to the target temperature within a specified residence time for continuous pyrolysis operation. Note that even if the hot flue gas at a sufficient volume is flown, the heat transfer would not be achieved within the residence time due to some heat losses. Therefore, we investigated the operating conditions which enable the heat transfer to be carried out for the residence time. Regarding the heat transfer, the hot flue gas temperature, the flow rate, the HCs’ circulation time, HCs’ circulating volume, and HC diameter have the significant effect to make the HCs reach the desired pre-heating temperature. Thus, in this study, the temperature of the hot flue gas and the flow rate were considered as the variables.

We assumed that the biomass feedstock consists of cedar wood chips, and a 34.5 kg-feedstock/h scale plant was evaluated. In this plant, it is designed to require 1,702 kJ/h/kg of heat to decompose the raw material in the pyrolizer, and the required heat can be achieved by heating HCs to 800 °C in the preheater, which are then supplied to the pyrolizer at 229 kg/h. Therefore, the HCs must be heated in the preheater from an initial temperature of 450 to 800 °C by heat exchange with the hot flue gas. On the other hand, because the preheating furnace is small (0.35 m in diameter and approximately 0.59 m in filling height) and can only hold 124 kg of HCs, the HCs must be fully heated within 1,953 s. Regarding the shape of the HCs, we assumed a spherical shape with a diameter of 10 mm.

As described in the previous section, because our process requires heating of the HCs and acceptable heat transfer in the preheater under the time constraint, it is important to apply computational fluid dynamics (CFD) simulations that can follow the time-series change of the temperature distribution to determine the optimum operating conditions of the plant. There have been many studies on particle-fluid heat transfer involving numerical studies and CFD simulations, but these studies were mostly aimed at obtaining the heat transfer parameters and analyzing the flow in the layer. In contrast, our study is novel in that the results of CFD simulations are applied to examine the operational conditions of the plant in terms of the relationship between the thermal profile and the time constraint, with the purpose of improving the heat utilization of our process. Two major methodologies for the numerical simulation of fixed-bed models such as the one envisioned in this study have been studied\(^8\): the first is to calculate the packed bed assuming that it is an effective porous medium. This method is computationally less expensive but requires an effective viscosity to determine average values of the dispersion, heat transfer, and bed velocity. The second method is to model each particle in the packed bed as a discrete. This model is well-defined and there is no need for effective parameter estimation, although modeling and gridding are more complicated, and thus increase the computational complexity. The latter method is used in this study. The CFD package "PHOENICS"\(^9\) was used for the analysis.
2. Process design and evaluation methods

2.1 Estimation using the transient heat conduction equation

Before the CFD simulation execution, the temperature of the inlet flue gas so that the temperature of HCs can be raised within the constraint time was estimated. The equation for transient heat conduction of a sphere with boundary condition of the second order was used to estimate the required gas temperature. Note that this estimation assumes fluid stasis, while the actual preheater involves fluid flow. Furthermore, a single HC particle is considered in this equation, while the actual preheater is filled with a large number of spheres.

The specific heat of the HC (Al₂O₃) at constant pressure (Cs) is 1,006 J/kg/K, the thermal conductivity (Ks) is 5.43 W/m/K, and density (ρ) is 3,650 kg/m³, respectively.

The temperature at position (r) [m] at time (t) [s] is represented as follows.

\[
\rho C_s \frac{\partial T}{\partial t} = \frac{K_s}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T}{\partial r} \right) \tag{1}
\]

The finite element method was applied to this equation. The time step (Δt) is 1.0 s (t=pΔt, p=1,2,...), the position from the center of the sphere is r (=nΔr, n=0,1,2,...,9), and the numerical solutions of temperature profile \( T_n^p \) is obtained on basis of Eq. (1). Here, the initial temperature condition at \( t = 0 \) \( (T_n^0) \) is 450 °C. The general equations for the process are expressed using the thermal diffusivity (α) as follows.

\[
\frac{(T_n^{p+1} - T_n^p)}{\Delta t} = \alpha \left[ \frac{T_{n+1}^p - 2T_n^p + T_{n-1}^p}{\Delta r^2} + \frac{T_{n+1}^p - 2T_n^p + T_{n-1}^p}{\Delta r^2} \right] \tag{2}
\]

\[
T_n^{p+1} = 6\theta r T_n^p + (1 - 6\theta r) T_n^p \tag{3}
\]

\[
T_n^{p+1} = \theta_n \left[ 1 + \frac{1}{n} \right] T_{n+1}^p + \theta_n \left[ 1 + \frac{1}{n} \right] T_{n-1}^p + (1 - 2\theta_n) T_n^p \tag{4}
\]

\[
\theta_n = \frac{\alpha \Delta t}{(\Delta r)^2} \tag{5}
\]

The estimation of transient heat transfer indicated that the inlet flue gas temperature should be set to 864 °C to heat the HCs from 450 to 800 °C within 1,953 s. Therefore, we decided to simulate the temperature rise of the HCs in the preheater using the hot flue gas temperature of 864 °C for the CFD simulation.

2.2 CFD simulation

We modeled a preheater with the shape shown in Fig. 2, simplified into a cylinder (see Fig. 3 a). The hot gas was assumed to flow from the bottom of the model. To reduce the calculation volume, we took advantage of the cylindrical symmetry and used an area equal to 1/36th of the cylinder for the calculations (see Fig. 3 b). A packed HC was based on a face-centered cubic structure, and spheres blocked by cylindrical wall were excluded. Here, we simulated the unsteady dynamic aspects on the heat transfer of HCs during duration time in the vessel. The calculation domain is assumed to be the cylindrical coordinates, divided into meshes with a radial angle of 0.9°, 2.5 mm in the radial direction, and 2.5 mm in the height, respectively. For instance, a calculation duration was approximately 40 hours. Note that our calculator is the personal computer of Intel (R) Xeon (R) E5-2630 (2.4GHz) CPU and 32 GB RAM.

The flow rate of the flue inlet gas in the entire preheater was designed by the following equations.

\[
m_f C_{f1} + m_s C_{s1} = m_f C_{f2} + m_s C_{s2} \tag{6}
\]

\[
m_f = \frac{m_s (T_{s2} - T_{s1})}{C_s (T_{f2} - T_{f1})} \tag{7}
\]

Here, the weight of packed HCs (mₐ) is 124 kg, and they must be heated from the initial temperature (Tₛ₁) of 450 °C to the target temperature (Tₛ₂) of 800 °C. Note that Tₛ₁ is the same as Tᵣ. The flue gas inlet temperature (Tᵣ) was designed to be 864 °C from the estimation in 2.1. The outlet temperature (Tₒ) was set to 500 °C with the approach temperature difference of 50 °C. Therefore, the amount
of gas flow into the vessel for the time constraint \( (\text{m}_f) \) is designed to be 109 kg. Note that the specific heat \( (C_f) \) was set to 1,102 J/kg/K averaged over the range of 100 to 1,000 °C \( ^{10} \). Based on \( C_f \), the sensible heat of inlet flue gas was designed to be 104 MJ in correspondence with \( m_f \).

Next, the parameters listed in Table 1 were used for our simulation model. Note that the HCs’ thermal conductivity and specific heat are based on experimental data using the transient hot wire method \(^{11-12} \), and these values include the effect of thermal radiation between particles \(^{10} \).

The thermal conductivity of HC at 550 °C \( (K_s) \) was estimated using the measured experimental data on the thermal conductivity of packed bed (see Table 2) \(^{11-12} \). In this experiment, we used the apparatus shown in Fig. 4. Here, \( K_s \) of spheres with diameters of 1 to 3 mm were estimated to be 1.21, 2.73, and 3.14, respectively. These values are expressed based on Eq. (8), which approximates the relationship between the diameter [mm] and thermal conductivity. The thermal conductivity for 10 mm-diameter HC was determined to be 5.43 W/m/K.

\[
K_s = 1.80 \ln d + 1.28 \quad (R^2=0.97) \quad (8)
\]

The specific heat of the HCs at constant pressure is obtained from the thermal conductivity of \( K_s \), and Eqs. (1) and (2). In this study, \( C_s \) was obtained from the estimated heat transfer under the initial temperature conditions of 525 to 575 °C for 7,200 s.

In addition, no-slip adiabatic walls were assumed as the boundary condition. The initial temperature is uniform at 450 °C, and the inner pressure is ambient pressure (1 bar). The \( \kappa-\varepsilon \) model \(^{10} \) was used for turbulence conditions in the CFD simulation.

In the result of each simulation run on basis of a cross section with an angle of 5°, the average temperature of the cross section \( \text{T}_{\text{ave}} \) (°C) is obtained. Moreover, the heat transfer efficiency in the preheater is evaluated by the following definition.

\[
\eta = \frac{m_C_s(T_{\text{ave}}-T_{s1})}{m_fC_fT_{f1}} \quad (9)
\]

### Table 1 Parameters in CFD simulation

| Computational domain |  |
|----------------------|----------------------|
| Radius of the cylinder [m] | 0.175 |
| Radius angle [''] | 10 |
| Height of cylinder [m] | 0.8 |
| Filling height [m] | 0.59 |

| Heat Carrier |  |
|----------------------|----------------------|
| Chemical formula of HC | Al₂O₃ |
| Particle size of HC [mm] | 10 |
| Density [kg/m³] | 3,650 |
| Porosity [%] | 40 |
| Thermal conductivity of HC \( K_s \) [W/m/K] | 5.43 |
| Specific heat of HC \( C_s \) [J/kg/K] | 1,006 |
| Initial temperature \( T_{s1} \) [°C] | 450 |

| Flue Gas |  |
|----------------------|----------------------|
| Gas inlet temperature \( T_{f1} \) [°C] | 864 |
| Flow rate of gas inlet [kg/s] | 1.55E-03 |
| Reynolds number [-] | 5,500 |

### Table 2 Parameters of the thermal conductivities

| HC diameter [mm] | 1.0 | 2.0 | 3.0 |
|------------------|-----|-----|-----|
| Measured [W/m/K] | 0.457 | 0.509 | 0.582 |
| Note: Packed bed (Fig. 4) |  |

| Estimated (550 °C) |  |
|------------------|----------------------|
| \( K_s \) [W/m/K] | 1.21 | 2.73 | 3.14 |

3. Results and Discussion

3.1 Results of CFD simulation

As a result of the simulation using the inlet flue gas at 864 °C with the designed 104 MJ sensible heat, it was found that all HCs could not be heated to the target temperature of 800 °C within the allotted time, and reached an average temperature of only 752 °C. The temperature...
distribution is shown in Fig. 5. The heat transfer efficiency in the preheater was \( \eta \) of 36%. That is, this means that the rest of 64% of the sensible heat was discharged. This indicates that although sufficient sensible heat was provided to raise the HCs temperature theoretically, the desired heat exchange was not completed within the residence time and discharged as tail gas.

3.2 Relationship between gas temperature, gas flow rate and heat transfer

The operating conditions of gas flow rate and gas temperature were examined due to the actual gas aspects, by which the more efficient heat exchange within the residence time would be achieved. Here, on the following two different conditions, the performances were analyzed in use our simulation model (see Table 3).

First, to satisfy with the required heat exchange, the effect of flow rate gain was investigated. That is, we evaluated how large benefit can be obtained within the residence time by increasing the gas flow rate. In this case (Case A), the inlet gas flow increased by 30% which corresponds to 136 MJ was simulated. Note that the inlet temperature is 846 °C as the same as an original one.

Second, the case that the inlet flue gas temperature raises up to 1,000 °C in the same gas the sensible heat capacity as an original case was simulated (Case B). Note that the maximum operating temperature of preheater is 1,000 °C due to the specification. In this case, the inlet flue gas has the sensible heat capacity of 104 MJ.

According to the result of Case A, it was found that the heat transfer would be improved, and that the HCs temperature raised up to 802 °C within the residence time (see Fig. 6). In this case, the heat transfer efficiency in the preheater was \( \eta \) of 33%. Therefore, feeding the heat capacity of the inlet flue gas whose temperature is 864 °C, the 1.3 times gas volume should be necessary to complete the heat transfer theoretically. In addition, the additional sensible heat input to the preheater would be a significant impact on the production efficiency of Bio-H₂. Likewise, the heat transfer efficiency in the preheater was 51%. The result of Case B indicated that the desired heat exchange was efficiently achieved for the residence time.

According to these results, it was found that the higher gas flow rate ensured to raise the temperature of the HCs up to the target level. While it should be noted that the additional sensible heat input to the preheater has a significant impact on the production efficiency of Bio-H₂. Likewise, it was found that higher temperature of inlet gas resulted in the more effective heat transfer, that is, the heat exchange to raise the temperature of the HCs for the residence time without any additional sensible heat supply.

4. Conclusions

In our biomass indirect thermochemical process, which uses a solid-gas reaction to produce Bio-H₂, the effective heat transfer condition carried out by the circulation of alumina balls acting as HCs is an important issue. That is, it is necessary to operate the circulation of the heat transfer medium more effectively so as to maximize the production efficiency.

The purpose of this study was to find out the suitable condition by simulating the heat utilization performance of HCs heated by a hot flue gas in the preheater, that is, to determine the operating conditions under the most effectively transferred to the HCs from the sensible heat of the hot flue gas for the designed residence time.

According to our analyzed results, it was found that the higher gas flow rate ensured to raise the temperature of the HCs up to the target level. For instance, feeding the air flow rate of 1.3 times gas volume was necessary to complete the heat transfer efficiently.

\[
\text{Table 3 Parameters in each case}
\]

|                        | Original | Case A | Case B |
|------------------------|----------|--------|--------|
| Gas temp. [°C]         | 864      | 864    | 1,000  |
| Inlet flue gas [kg/s]  | 1.55E-03 | 2.01E-03 | 1.34E-03 |
| Sensible heat [MJ]     | 104      | 136    | 104    |
| Reynolds number [-]    | 5,500    | 4,700  | 7,100  |

Fig. 6 Temperature distribution
the heat capacity of the inlet flue gas whose temperature is 864 °C, the heat exchange will be completed when 1.3 times the amount of heat calculated from the steady state heat balance is applied. Moreover, in this condition, the heat transfer efficiency in the preheater was 33%. Likewise, it was found that higher temperature of inlet gas resulted in the more effective heat transfer, that is, the heat exchange to raise the temperature of the HCs within the residence time without any additional sensible heat supply. For instance, supplying the inlet flue gas whose temperature is 1,000 °C, all HCs would be heated over the target temperature, and their average temperature was 806 °C.

In our future tasks, the heat loss to the outside will be considered to validate our model. This means that our simulated results will be compared using the measurement data of temperature profiles in the demo-plant, and that we will improve our model.

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