Development of a Design for a Carbon Capture and Storage Facility by Utilizing a Process Simulation Technology

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A pilot plant for a Carbon Capture and Storage (CCS) facility was designed by utilizing a reaction rate-based process simulation technology. The targeted energy consumption was achieved in the pilot plant tests, but the carbon capture performance was much higher than the simulation results. Based on the data obtained in the tests conducted at the pilot plant, the simulation model was improved to the point where the results matched the test data. In the future, this simulation model will be further improved and utilized in developing designs for demonstration and commercial plants.

1. Introduction

In 2015, COP21 was held and the Paris Agreement was adopted. An agreement was reached for a global framework for mitigating climate change, which aims to keep the rise in global temperature below 2°C compared to those days before the Industrial Revolution. This is called the 2°C scenario (2DS), determining that the member nations establish greenhouse gases emission reduction targets on a voluntary basis and take measures to achieve them. Following energy saving and renewable energy, Carbon Capture and Storage (CCS) is positioned as the third most important technology that contributes to the achievement of 2DS[1]. With the need to capture and store no less than 94 Gt of CO_2 by no later than 2050, CCS must be disseminated[2].

The term CCS refers to technologies intended to control the emission of CO_2 into the atmosphere by capturing CO_2 emitted from large-scale emission sources, such as power-generating plants, ironworks, and cement plants, and then storing the captured CO_2 underground or in other appropriate locations. Among such CCS technologies, IHI focuses on two CO_2-capturing technologies, namely, oxy-fuel combustion[3] and chemical absorption[4].

Chemical absorption is a technology that separates and captures CO_2 in gases using amine aqueous solution or other alkaline aqueous solution through an absorption-regeneration reaction. **Figure 1** illustrates the principle of capturing CO_2 in chemical absorption. The absorber is fed with absorbing liquid from the top and with gas from the bottom so that the gases is forced to come into contact with the liquid in the absorber and only CO_2 is absorbed into the liquid. The liquid is then transferred to the regenerator through a heat exchanger; in the regenerator, a reboiler located at the bottom heats the liquid to regenerate the CO_2 from the liquid. The liquid is then returned to the absorber from the regenerator to be recycled. Packed towers, i.e., towers filled with packing
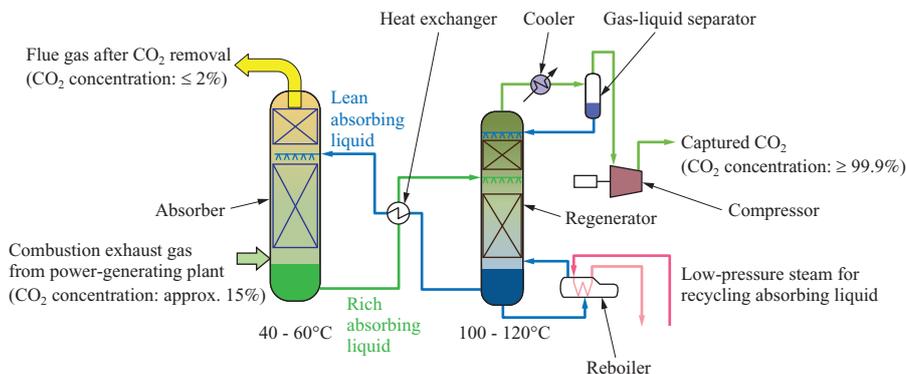
Fig. 1 Conceptual diagram for carbon capture using the chemical absorption method

that facilitates the contact of gas with liquid, are generally used as the absorber and regenerator.

This system requires a large amount of thermal energy to regenerate CO_2; the cost of this energy accounts for about half the total cost. For this reason, IHI is implementing development that focuses mainly on three elemental technologies (absorbing liquid, packing, and process), which have been identified as technologies for reducing the thermal energy required. With the objective of evaluating and validating these elemental technologies, demonstrating them in the future, and obtaining data for designing commercial plants, IHI designed and constructed a chemical absorption pilot plant (hereinafter referred to as the pilot plant) with a capacity of 20 t-CO_2/d (of a scale large enough to capture 20 metric tons of CO_2 per day) in IHI Aioi Works. This article reports one of our approaches to designing this pilot plant and using process simulations to study equipment specifications and predict its performance.

2. Specifications of the pilot plant

Tables 1 to **3** show the flue gas conditions, flue gas composition, and targets for carbon capture performance for the pilot plant. The flue gas composition was determined by imitating the flue gas from a coal fired power plant. 30 wt% MEA (Monoethanolamine) aqueous solution, which is generally used for chemical absorption, was used as the absorbing liquid.

3. Reaction models

The Aspen Rate-Based Distillation™ was used for process simulations. These simulations allow for calculations that take into account the reaction rate in a packed tower and enable precise analysis through general equilibrium-based calculations.

The reaction models defined by Aspen Plus®'s MEA rate-based CO_2 absorption model are expressed by formulae (1) through (9) below[5], [6].

$$2H_2O \Leftrightarrow H_3O^+ + OH^- \quad \text{...........................(1)}$$
$$CO_2 + 2H_2O \Leftrightarrow H_3O^+ + HCO_3^- \quad \text{.....................(2)}$$
$$HCO_3^- + H_2O \Leftrightarrow H_3O^+ + CO_3^{2-} \quad \text{...................(3)}$$
$$MEAH^+ + H_2O \Leftrightarrow MEA + H_3O^+ \quad \text{.....................(4)}$$
$$MEACOO^- + H_2O \Leftrightarrow MEA + HCO_3^- \quad \text{.............(5)}$$
$$CO_2 + OH^- \rightarrow HCO_3^- \quad \text{...........................(6)}$$
$$HCO_3^- \rightarrow CO_2 + OH^- \quad \text{...........................(7)}$$
$$MEA + CO_2 + H_2O \rightarrow MEACOO^- + H_3O^+ \quad \text{..........(8)}$$
$$MEACOO^- + H_3O^+ \rightarrow MEA + CO_2 + H_2O \quad \text{..........(9)}$$

Formulae (1) to (5) are equilibrium reaction models that incorporate Gibbs energy, from which an equilibrium constant is calculated. Formulae (6) to (9) represent reactions based on a reaction rate, which is calculated by formula (10).

$$r = k \exp\left(\frac{-E}{RT}\right) \prod_{i=1}^{N} C_i^{a_i} \quad \text{...........................(10)}$$

where k: frequency factor; T: temperature; E: activation energy; R: gas constant; C: concentration; and a: stoichiometric coefficient. The reaction rate parameters for k and E of reaction rate formulae (6) to (9)[5], [6] are respectively listed in **Table 4**.

4. Calculation models

The double boundary film theory[7] is generally used as a theory to explain the reaction through which gas is absorbed in liquid. **Figure 2** provides a conceptual diagram of the double boundary film theory.

Each of the gas and liquid phases forms a boundary film along the interface, and gas absorption into liquid is illustrated by mass (gas/liquid) transfer from one boundary film to another and an absorption reaction at the interface. Since the pilot plant used packed towers, the mass transfer coefficient calculation model uses the correlation formula[8] by Bravo et al., the holdup amount calculation model uses the correlation formula[9] by Bravo et al., and the heat transfer coefficient calculation model uses Chilton and Colburn's theory[10]. Using these correlation formulae, we calculated the material balance, heat balance, etc., in each section to evaluate the CO_2 absorption and regeneration performance in the absorber and regenerator, and studied the size best suited for achieving the targets for carbon capture performance as shown in **Table 3**.

Table 1 Flue gas conditions of the pilot plant

Item	Unit	Flue gas condition
Absorber flow rate of incoming gas (dry)	m³N-dry/h	3 218
Gas temperature	°C	40
Gas pressure at absorber inlet	kPaG	−7.87

Table 2 Flue gas composition

Item	Unit	Value
CO_2	dry %	14.6
O_2	dry %	4.8
N_2	dry %	80.5
SO_2	ppm (dry)	5.0
Smoke dust	mg/m³N	10.0

Table 3 Targets for carbon capture performance

Item	Unit	Performance target value
CO_2 regeneration energy	GJ/t-CO_2	4.0
CO_2 capture amount	t-CO_2/d	20
CO_2 capture rate	%	90

Table 4 Reaction rate parameters for the MEA model

Reaction rate formula	Reaction rate parameters	
	k (frequency factor) (−)	E (activation energy) (cal/mol)
(6)	1.33 e+17	13 249
(7)	6.63 e+16	26 656
(8)	3.02 e+14	9 855.8
(9)	1.28 e+25	18 361

(Note) G : Gas flow rate
L : Liquid flow rate
y : Gas molar fraction (bulk)
T : Temperature
H : Enthalpy
N : Mass transfer rate
q : Heat transfer rate
x : Liquid molar fraction
*1 : Heat and material balance calculations are performed within the area defined by the upper and lower broken lines (liquid/gas).
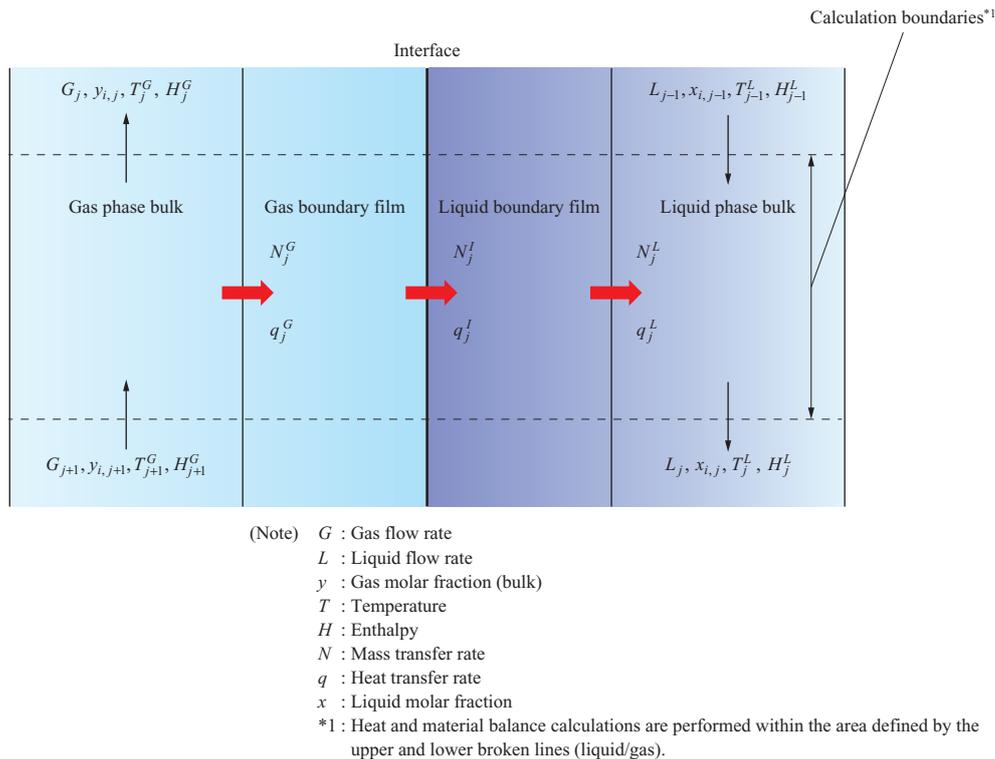
Fig. 2 Conceptual diagram for the double boundary film theory

5. Design of the absorber and regenerator by means of simulations

First, we conducted a study on the diameters of the absorber and regenerator. When countercurrent contact operation is performed on the gas and liquid in the absorber while the flow velocity of the gas is increased with the flow rate of the liquid remaining constant, gas pressure loss in the absorber increases. Once the gas flow velocity exceeds a certain value, there is a sharp increase in gas pressure loss and the liquid cannot flow down or flow up towards the top of the absorber. This phenomenon is called flooding and renders the absorber inoperable. Hence, the absorber must have a large enough diameter that eliminates the possibility of the gas flow velocity flooding during operation.

MellapakPlus[TM] 252Y (Sulzer Chemtech, Switzerland), which is a general commercially available packing material, was used as packing for the absorber and regenerator at the pilot plant. The flooding point of this packing is identified as the point at which 1 200 Pa/m of pressure loss is reached. In terms of fractional capacity, packed towers are designed with a tower diameter that forces the flooding limit, which represents the margin for flooding, to be 0.5 - 0.8.

Figure 3 shows the simulation evaluation results of the relationship between the absorber diameter and the flooding limit at a liquid-to-gas ratio of 6.0 l/m^3, which is generally utilized when MEA liquid is used. Given that the fractional capacity for an absorber is approximately 820 - 1 000 mm, we adopted JIS10K 850A as the diameter of the absorber at the pilot plant with reference to the applicable Japanese
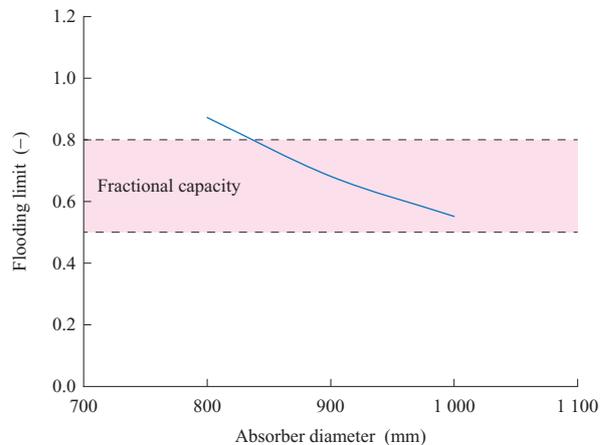
(Note) Liquid-to-gas ratio : 6.0 l/m^3
Absorber diameter : JIS10K 850A

Fig. 3 Fractional capacity of the absorber according to the results of the initial simulation model

Industrial Standard on piping (JIS G 3468) in order to ensure that the absorber had a smaller diameter than the maximum fractional capacity. In a regenerator, which includes a reboiler for heating liquid and regenerating CO_2, the optimum operating pressure varies depending on the type of absorbing liquid. Since any change in the operating pressure can cause a change in the gas flow velocity, the diameter of the regenerator must be determined while taking into account the flooding point, which depends on pressure. **Figure 4** shows the simulation results of the liquid-to-gas ratio vs. flooding limit at multiple operating pressures when the
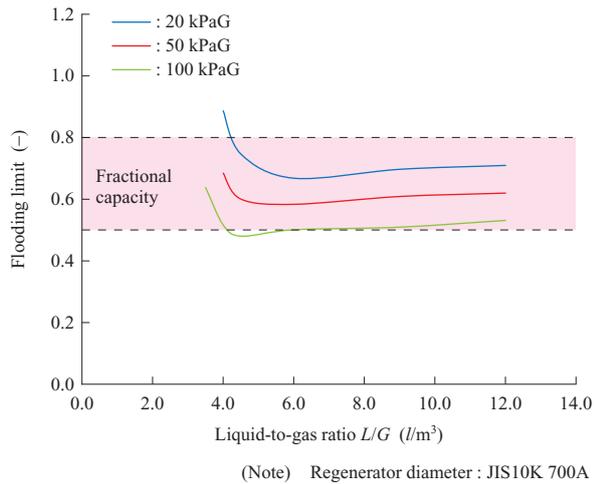
Fig. 4 Fractional capacity of the regenerator according to the results of the initial simulation model

regenerator diameter is equal to JIS10K 700A. When the liquid-to-gas ratio was varied within the expected operating pressure, ranging from 0 to 100 kPaG, most of the simulated flooding limits were within the fractional capacity for flooding limit (0.5 - 0.8). As such, we adopted JIS10K 700A as the diameter of the regenerator.

Next, we conducted a study of the packing height in absorber. The liquid-to-gas ratio must be reduced in order to reduce the volume of liquid circulating in the process and subsequently, the CO_2 regeneration energy. When the packing height in absorber is increased, the amount of CO_2 absorbed also increases, enabling the liquid-to-gas ratio to be reduced. From the perspective of cost, however, the packing height in absorber must be low. It is therefore necessary to determine the optimum liquid-to-gas ratio and the packing height in absorber while taking into account the CO_2 absorption rate of the absorbing liquid. **Figure 5** shows the simulation results of the liquid-to-gas ratio vs. CO_2 regeneration energy at different packing height in absorber.

The simulation results indicated that the packing height of 15 m and a liquid-to-gas ratio of 4.5 l/m^3 served as optimum operating conditions and that at a liquid-to-gas ratio of less than 4.0 l/m^3, the CO_2 regeneration energy was significantly higher.

6. Comparison with the performance evaluation test

The design and construction of the pilot plant were implemented based on the simulation results. **Figure 6** shows the pilot plant with a capacity of 20 t-CO_2/d. **Figure 7** shows the results of testing this pilot plant using MEA and the results of performance prediction based on the simulation.

The test results demonstrated that the lowest CO_2 regeneration energy, 3.8 GJ/t-CO_2, was achieved at a liquid-to-gas ratio of 4.0 l/m^3, and that performance of up to 4.0 GJ/ t-CO_2, which is the target value, was achieved. However,

Fig. 6 Photograph of the pilot plant

Fig. 5 Regeneration energy according to the results of the initial simulation model
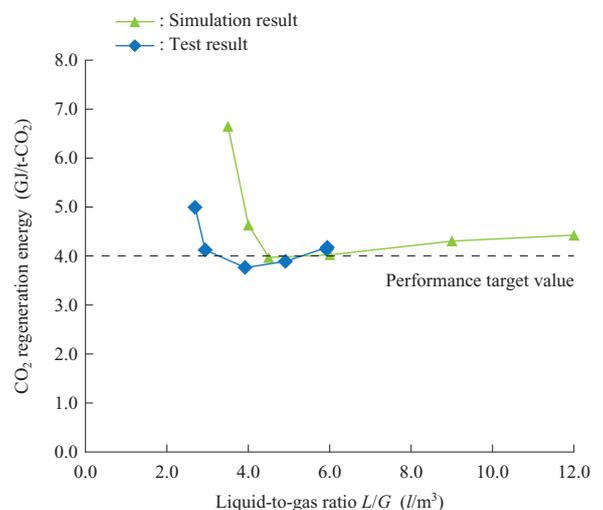
Fig. 7 Comparison between the test data and the results of the initial simulation model

after comparing the results shown in **Fig. 7**, various temperature profiles during the test, the results of analyzing CO_2 concentration in the absorbing liquid, and other test data with the relevant simulation data, it was revealed that the simulation model considerably deviated from the test results.

In order to render the simulation model consistent with the test results, we considered fitting the simulation model. Calculations in the simulation were performed on the assumption that the packing in the packing surface would have an ideal state, in other words, be perfectly wet. However, in reality, it is highly possible that the packing surface might have an unutilized portion because the liquid flow is drift or directed at a wall surface or for other reasons. In order to compensate for this loss, a parameter called Interfacial Area Factor (IAF) was adjusted. In addition, since the simulation result for the regeneration reaction in the regenerator was found to have a lower reaction rate compared with the test result, the reaction rate parameter of formula (9), which expresses a CO_2 regeneration reaction of MEA, was adjusted and subjected to fitting.

Figure 8 compares the performance of the fitted model, including the adjustments of the aforementioned two parameters, with the relevant test data. The fitting provided simulation results that are close to the test data. In addition, various temperature profiles, the CO_2 content in liquid, etc., were also reproduced well, proving that we have successfully created a simulation model that is capable of simulating the performance of the actual pilot plant.

7. Conclusion

We predicted the performance of a CCS facility and conducted a study of packed tower sizes using reaction-rate-based process simulations and used the obtained data to design a pilot plant. The original simulation model has different performance results from the test data on the pilot plant, but by performing fitting on the model, we successfully created a model that is consistent with the test data.

We are currently focusing on the development of elemental technologies (absorbing liquid, packing, and process) to improve CO_2 regeneration performance while obtaining various data using the pilot plant. Going forward, we will ① create a physical property model of the absorbing liquid developed by IHI, ② improve the mass transfer model to make it more suitable for the developed packing, and ③ implement process optimization, in order to develop an advanced simulation model in order to demonstrate the CCS facility, predict the performance of a commercial plant, and use the simulation model for designing.

Fig. 8 Comparison between the test data and the results of the modified simulation model

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