Hydrate-based carbon dioxide capture from simulated integrated gasification combined cycle gas

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Abstract
The equilibrium hydrate formation conditions for CO₂/H₂ gas mixtures with different CO₂ concentrations in 0.29 mol% TBAB aqueous solution are firstly measured. The results illustrate that the equilibrium hydrate formation pressure increases remarkably with the decrease of CO₂ concentration in the gas mixture. Based on the phase equilibrium data, a three stages hydrate CO₂ separation from integrated gasification combined cycle (IGCC) synthesis gas is investigated. Because the separation efficiency is quite low for the third hydrate separation, a hybrid CO₂ separation process of two hydrate stages in conjunction with one chemical absorption process (absorption with MEA) is proposed and studied. The experimental results show H₂ concentration in the final residual gas released from the three stages hydrate CO₂ separation process was approximately 95.0 mol% while that released from the hybrid CO₂ separation process was approximately 99.4 mol%. Thus, the hybrid process is possible to be a promising technology for the industrial application in the future.

Key words
integrated gasification combined cycle (IGCC); gas hydrate; CO₂ separation; H₂ purification; chemical absorption

1. Introduction
Carbon dioxide (CO₂) is considered as one main contributor to the greenhouse gases resulting in the global warming [1]. Near one third of all CO₂ emissions worldwide is produced by the fuel power plants [2]. Therefore, the prime target is to capture CO₂ from the fuel power plants to slow down the deterioration of the climate. Pre-combustion capture and post-combustion capture are two approaches extensively used for CO₂ capture from the fuel power plants [3]. Pre-combustion capture is capturing or removing CO₂ before the fuel being burnt while post-combustion capture is capturing or removing CO₂ from the flue gas [4]. Presently, the integrated gasification combined cycle (IGCC) is widely utilized in the coal-fired power plants. CO₂ capture from IGCC becomes one promising route of pre-combustion capture. In IGCC plant, coal is firstly gasified and then converted into a stream of hydrogen (H₂) and CO₂ [5]. Once CO₂ is removed efficiently, the resultant H₂ can be used as a clean fuel. Physical adsorption, chemical absorption, cryogenic fractionation and membrane process are the conventional technologies for capturing CO₂ from the fuel gas. However, the conventional technologies face some challenges, such as low capacity, technology feasibility, high cost and corrosion [6,7]. Therefore, novel and low cost technologies of carbon capture need to be developed.

Gas hydrate-based CO₂ capture technology is one of the novel technologies for capturing CO₂ from IGCC synthesis gas (mixture of CO₂/H₂). The gas hydrates are non-stoichiometric compounds formed by water molecules and small molecule gases such as CO₂, nitrogen (N₂), oxygen (O₂), H₂, methane (CH₄) [8]. The mechanism of hydrate-based CO₂ capture is selective partition of CO₂ component between hydrate phase and gaseous phase [9,10]. Because the equilibrium hydrate formation pressure of CO₂ is much lower than that of H₂ at the same temperature, it is expected that
CO₂ is preferentially encaged into the hydrate phase. The hydrates are then separated and subsequently decomposed to create CO₂-rich stream while the rest constitutes CO₂-lean one. Therefore, CO₂ is captured from IGCC synthesis gas.

IGCC synthesis gas mainly contains approximately 40.0/60.0 mol% CO₂/H₂ gas mixture, and the outlet pressure of the synthesis gas is generally 3−5 MPa [3]. In our previous work [10], the concentration of CO₂ can be reduced from 40.0 mol% in the feed gas to approximately 13.5 mol% in the residual gas after one stage hydrate-based CO₂ separation. Therefore, in order to separate CO₂ from CO₂/H₂ gas mixture completely, two hydrate stages or even three hydrate stages are required. However, the equilibrium hydrate formation pressure is higher with the decrease of CO₂ concentration in the gas mixture. Thus, higher driving forces are required for the gas mixtures containing lower concentration of CO₂ to form gas hydrate, that is, more serious conditions are required for the gas mixtures containing lower concentration of CO₂ to form gas hydrate. Therefore, a hybrid process based on the hydrate and chemical absorption process is developed. Moreover, no complete system configurations with key hybrid process parameters were presented in the papers [11,12].

In our previous work, 0.29 mol% TBAB solution was proved to be an optimal hydrate formation promoter for the hydrate-based CO₂ capture from either flue gases or IGCC synthesis gas [9]. Thus, 0.29 mol% TBAB solution is adopted in this work directly. In addition, the equilibrium hydrate formation conditions for CO₂/H₂/TBAB/H₂O systems with different CO₂ concentrations are measured firstly, and a hybrid process based on two hydrate stages coupled with a chemical absorption process is proposed for CO₂ capture. By the comparison of CO₂ capture efficiencies, the concentrations of H₂ in the residual gas, the energy consumptions between the third-stage hydrate-based CO₂ separation process and the hybrid process, and an optimal separation process are determined.

2. Experimental

2.1. Materials

A CO₂/H₂ gas mixture containing 39.6 mol% CO₂ was used in the work to simulate a pretreated fuel gas mixture. The gas mixture was supplied by Foshan Huate Gas Co., Ltd. Tetra-n-butyl ammonium bromide (TBAB) with 99.9% purity was supplied by Shanghai Sinopharm Chemical Reagent Co., Ltd., China. Monoethanolamine (MEA) with the purity of more than 99.0% was offered by Tianjin Fuyu Fine Chemicals Co., Ltd. The deionized water used with the resistivity of 18.25 mΩ·cm was produced by an ultra-pure water system supplied by Nanjing Ultrapure Water Technology Co., Ltd., China. The glass-balls with the diameter of 3 mm were supplied by Yunhui Business Co., Ltd., China.

2.2. Apparatus

The experimental apparatus in this work is shown in Figures 1 and 2. The apparatus in Figure 1 was the same as that in our previous work [9]. The crystallizer (CR) with inner volume of 336 mL and the supply vessel (SV) with the inner volume of 1350 mL were made of 316 stainless steels. They were immersed in a glycol-water bath. On the front and back of the CR, there are two circular viewing windows made of Plexiglas. The maximum working pressure of the CR was 25 MPa. A magnetic stirrer (500 r/min) was employed to mix the contents in the CR. The temperatures of the gas phase and the liquid phase in the CR were measured using two Pt1000 thermocouples (JM6081) with uncertainties of ±0.1 K. The pressures in the SV and CR were measured with two Setra smart pressure transducers (model 552, Boxborough, MA, USA) with the uncertainty of ±20 kPa. In addition, the composition of the gas phase in the CR was determined with a HP6890 gas
chromatograph (GC) connected on-line with CR and automated with a personal computer (PC). Figure 2 is one simulative MEA CO$_2$ separation system. The round bottom flask (a) (as an absorption bottle) with effective volume of 500 mL was added into adequate quantities of glass-balls. Then 300 mL MEA solution of 30 wt% was added into the round bottom flask (a) and the solution surface was ensured to be 2–3 mm higher than the top of the glass-ball stack. The gas mixture was introduced into MEA solution by a right-angle glass tube, of which one terminal was inserted into the glass-ball stack. The other acute-angle glass tube was used for collecting H$_2$, of which one terminal went through the rubber stopper. Another round bottom flask (b) was used as a stripper for regenerating MEA solution. The flask (b) was heated with a heater, and the released CO$_2$ and the solution steam were collected via an acute-angle tube, of which one terminal was connected to a coiled condenser. The final gas was collected with a sealed gaseous envelope, then, it was detected with GC.

![Experimental apparatus for MEA CO$_2$ separation. (a) Absorption bottle, (b) stripper for MEA regeneration](image)

2.3. Procedure

TBAB solution with a desired concentration was introduced into the evacuated high-pressure cell. The content was pressurized up to the desired pressure by supplying CO$_2$/H$_2$ gas mixture. In this work, we firstly measured the equilibrium hydrate formation pressure for different CO$_2$/H$_2$ gas mixtures (CO$_2$ concentrations were 39.8 mol%, 18.8 mol% and 10.0 mol%, respectively) in the presence of TBAB. The method of three-phase equilibrium measurement adopted in the work was as same as the “T-cycle method” used by Ohgaki et al. [13]. Each “T-cycle” experiment was repeated for at least three times.

After equilibrium hydrate formation experiments, the procedure for hydrate stage was similar to that in our previous work [9,14]. The crystallizer (CR) was cleaned again using de-ionized water and allowed to dry. Then TBAB solution of 180 mL was introduced into the CR. Subsequently, the CR is flushed with CO$_2$/H$_2$ gas mixture several times to remove gases, and then CO$_2$/H$_2$ gas mixture was charged into the CR until the desired pressure. Once the temperature was stabilized (typically within 1 min), the stirrer with a rate of 500 r/min was started, and the experimental time was recorded once every two second. The time when the stirrer was started was defined as $t_0$. The data of temperature and pressure were recorded during the experiment. As the gas in the crystallizer was consumed because of the hydrate formation, additional gas mixture in the SV was supplied and the pressure in the crystallizer was kept constant with a proportional integral derivative (PID) controller. The measurement of the compositions and the amount of the gas consumed ($n_H$) as well as the calculation of CO$_2$ separation efficiency were same as what mentioned in our previous work [10].

The hybrid process proposed in this work included a two-stage hydrate-based process and a chemical absorption process. The mechanism of the chemical absorption process was based on the following reaction [15]:

\[
\text{CO}_2 + 2\text{HOCH}_2\text{CH}_2\text{NH}_2 + \text{H}_2\text{O} \iff (\text{HOCH}_2\text{CH}_2\text{NH}_3)_2\text{CO}_3 \\
(\text{HOCH}_2\text{CH}_2\text{NH}_3)_2\text{CO}_3 + \text{CO}_2 + \text{H}_2\text{O} \iff 2(\text{HOCH}_2\text{CH}_2\text{NH}_3)\text{HCO}_3
\]

(1)

After two stages hydrate-based capture, the residue gas was introduced into a chemical absorption bottle, in which adequate amount of small glass-balls with the diameter of 3 mm and MEA solution of 30 wt% with desired volume had been added. CO$_2$ was absorbed completely by MEA solution and formed the compounds of (HOCH$_2$CH$_2$NH$_3$)$_2$CO$_3$ and (HOCH$_2$CH$_2$NH$_3$)HCO$_3$ while H$_2$ was not absorbed. Thus, CO$_2$ is separated from IGCC synthesis gas, and H$_2$ with high purity can be obtained. The pressure in the chemical absorption bottle was kept at 1.05 atm in order to ensure that the final gas can be collected easily. Then, the solution was introduced into a regeneration device, in which (HOCH$_2$CH$_2$NH$_3$)$_2$CO$_3$ and (HOCH$_2$CH$_2$NH$_3$)HCO$_3$ were heated to release CO$_2$ and regenerate MEA.

Based on the experiments, we further simulated CO$_2$ separation process in a 500 MW IGCC power plant with two CO$_2$
separation methods. One was the three stages hydrate-based capture process, and the other was the hydrate/chemical absorption hybrid process. The difference of the two processes was the chemical absorption stage in the hydrate/chemical absorption hybrid process which substituted the third hydrate stage in three stages hydrate-based capture process. The first two hydrate stages were the same for the two processes. Thus, we only need to compare the energy consumptions between the third hydrate stage of the hydrate process and the chemical absorption stage of the hybrid process. The energy consumption was calculated according to the formula listed in the book of Advanced Engineering Thermodynamics [16].

3. Results and discussion

3.1. Equilibrium hydrate formation pressure

We firstly measured the equilibrium hydrate formation pressure for CO$_2$/H$_2$ gas mixture with CO$_2$ concentration of 18.0 mol% in 0.29% TBAB solution. Compared with the results given by Li et al. [17], it is found that the results were in excellent agreement, as shown in Figure 3. This illustrates either the apparatus or the method adopted in this work is reliable.

![Figure 3. Equilibrium hydrate formation pressure for CO$_2$/H$_2$ gas mixtures in the presence of 0.29 mol% TBAB solution](image)

Figure 3 gives the measured equilibrium hydrate formation conditions for CO$_2$/H$_2$ gas mixtures with CO$_2$ concentrations of 10.0, 18.0 and 39.2 mol% in 0.29 mol% TBAB solution. The pressure ranged from 0.25 MPa to 6.23 MPa and the temperature ranged from 276.55 K to 284.55 K. The phase equilibrium data are summarized in Table 1. As shown in Figure 3, the equilibrium hydrate formation pressure decreased considerably with the increase of CO$_2$ in the gas mixture at the same temperature. For example, the equilibrium hydrate formation pressures were 3.45, 1.80 and 0.50 MPa at 280.15 K for CO$_2$/H$_2$ mixtures with CO$_2$ concentrations of 10.0, 18.0 and 39.2 mol%, respectively. In addition, the equilibrium temperatures at 4.0 MPa were 278.15, 279.75 and 282.65 K for CO$_2$/H$_2$ mixtures with CO$_2$ concentrations of 10.0, 18.0 and 39.2 mol%, respectively. The result illustrates that lower CO$_2$ concentration in CO$_2$/H$_2$ mixture results in higher equilibrium pressure at the fixed temperature or higher equilibrium temperature at the fixed pressure, and further results in forming the gas hydrate more difficulty.

Table 1. Equilibrium hydrate formation pressure for CO$_2$/H$_2$ gas mixtures with different concentrations in 0.29 mol% TBAB solution

<table>
<thead>
<tr>
<th>CO$_2$/H$_2$ gas mixtures</th>
<th>$T$/K</th>
<th>$P$/MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>10.0/90.0 mol%</td>
<td>276.55</td>
<td>2.33</td>
</tr>
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<td></td>
<td>277.15</td>
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</tr>
<tr>
<td></td>
<td>278.85</td>
<td>4.49</td>
</tr>
<tr>
<td>18.0/82.0 mol%</td>
<td>277.45</td>
<td>1.25</td>
</tr>
<tr>
<td></td>
<td>277.95</td>
<td>1.76</td>
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<td></td>
<td>279.15</td>
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<td></td>
<td>279.85</td>
<td>3.87</td>
</tr>
<tr>
<td></td>
<td>280.55</td>
<td>5.28</td>
</tr>
<tr>
<td>39.2/60.8 mol%</td>
<td>277.35</td>
<td>0.25</td>
</tr>
<tr>
<td></td>
<td>278.85</td>
<td>0.40</td>
</tr>
<tr>
<td></td>
<td>278.15</td>
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</tr>
<tr>
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<tr>
<td></td>
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<tr>
<td></td>
<td>282.85</td>
<td>4.05</td>
</tr>
<tr>
<td></td>
<td>284.55</td>
<td>6.23</td>
</tr>
</tbody>
</table>

3.2. Hydrate-based CO$_2$ separation

Figure 4 gives the gas uptake changes with time for three CO$_2$/H$_2$ gas mixtures with CO$_2$ concentrations of 10.0, 18.0 and 39.2 mol% at 4.0 MPa and 276.15 K. A typical example is shown in Figure 4. There was an inflexion in the curve for 10.0 mol% CO$_2$/H$_2$ gas mixture at the 625th second. The gas uptake increased slowly from 0 to 625 s while dramatically increased after the 625th second. At the 1830th second, the amount of gas uptake was 0.067 mol, and then the amount of gas uptake had little change from the 1830th second to the end of the experiment. Similar phenomenon was also shown for 18.0/88.0 mol% CO$_2$/H$_2$ gas mixture. The gas uptake had a slight increase from 0 to 162 s, then, it increased abruptly after the 162th second. At the 1945th second, the amount of gas uptake was 0.101 mol. Subsequently, the amount of gas uptake had little change. For 39.2/60.8 mol% CO$_2$/H$_2$ gas mixture, the gas uptake increased slowly from 0 to 66 s, and the amount of the uptake had a considerable increase from the 66th second. At the 1578th second, the amount of gas uptake was 0.115 mol, and subsequently it had little change. Similar to our previous work [10], the slow increase of the gas uptake are resulted from the dissolving of the gas mixture in TBAB solution while the abrupt increase of the gas uptake is attributed to the gas hydrate formation. Therefore, the time when the gas uptake turns to the increase abruptly is defined as the hydrate formation induction time [18]. Thus, as seen from Figure 4, the hydrate formation induction time for the three CO$_2$/H$_2$ gas mixtures at 4.0 MPa and 276.15 K were 625, 162 and 66 s, respectively. The result illustrates that as CO$_2$ concentration shifts to be lower, the hydrate formation
induction time increases observably. It is attributed to the fact that the gas mixture with lower CO\textsubscript{2} concentration has higher equilibrium hydrate formation pressure, and thus the corresponding driving force for the hydrate formation is lower, resulting in longer hydrate formation induction time [19]. However, for the hydrate-based CO\textsubscript{2} separation, longer induction time is unfavorable for the application in industry. Thus, the hydrate formation induction time for 10.0 mol\% CO\textsubscript{2}/H\textsubscript{2} gas mixture of 625 s, is too long to meet the requirement of industrial application for the hydrate-based CO\textsubscript{2} separation.

In addition, as seen from Figure 4, as CO\textsubscript{2} concentration shifted to be lower in the gas mixture, the gas uptake decreased remarkably at 4.0 MPa and 276.15 K. For example, for the three CO\textsubscript{2}/H\textsubscript{2} gas mixtures, the gas uptake obtained in this work were 0.067 mol, 0.101 mol, 0.115 mol when CO\textsubscript{2} concentrations were 10 mol\%, 18.0 mol\% and 39.2 mol\%, respectively. Although the gas uptakes obtained here were not high enough to meet the requirement of treating the large amount of IGCC synthesis gas in the industry, the shift tendency of the gas uptake with CO\textsubscript{2} concentration change in the gas mixture indicates that higher CO\textsubscript{2} concentration in the gas mixture is more helpful for the hydrate-based CO\textsubscript{2} separation from IGCC synthesis gas.

Table 2 shows the hydrate-based CO\textsubscript{2} separation efficiencies along with CO\textsubscript{2} concentrations in the residual gas phase and hydrate slurry phase for the three different CO\textsubscript{2}/H\textsubscript{2} gas mixtures at 4.0 MPa and 276.15 K. As seen from Table 2, because CO\textsubscript{2} concentration in the residual gas after the first and the second hydrate separation stages were 16.6 mol\% and 10.1 mol\%, the values were close to CO\textsubscript{2} concentration in the feed gas mixtures for the two stages (18.0 mol\% and 10.0 mol\%), respectively. Thus, a three stages hydrate-based CO\textsubscript{2} separation process was proposed. The process flowchart is shown in Figure 5. As seen from Table 2, CO\textsubscript{2} separation fraction and separation factor for CO\textsubscript{2}/H\textsubscript{2} gas mixture of 39.2/60.8 mol/mol were obviously larger than those for other two gas mixtures. As CO\textsubscript{2} concentration shifted to lower, CO\textsubscript{2} separation fraction and CO\textsubscript{2} separation factor shifted to lower. For CO\textsubscript{2}/H\textsubscript{2} gas mixtures with CO\textsubscript{2} concentrations of 39.2 mol\% and 18.0 mol\%, CO\textsubscript{2} separation fractions were 0.66 and 0.62, respectively, and CO\textsubscript{2} separation factors were 72.3 and 19.4, respectively. CO\textsubscript{2} separation fraction and separation factor for CO\textsubscript{2}/H\textsubscript{2} gas mixture with CO\textsubscript{2} concentration of 10.0 mol\% were only 0.45 and 10.3, respectively. The result illustrates that the hydrate-based CO\textsubscript{2} separation from CO\textsubscript{2}/H\textsubscript{2} gas mixtures with CO\textsubscript{2} concentrations of 39.2 mol\% and 18.0 mol\% are feasible. Nevertheless, that from CO\textsubscript{2}/H\textsubscript{2} gas mixtures with CO\textsubscript{2} concentrations of 10.0 mol\% is unfeasible because of the quite low CO\textsubscript{2} separation efficiency. Due to CO\textsubscript{2} concentration in the feed gas mixture of the third hydrate stage was lower than 10.0 mol\%, that separating CO\textsubscript{2} and purifying H\textsubscript{2} from IGCC synthesis gas via a whole three stages hydrate-based process is unfeasible. In other words, the continuous three stages hydrate-based CO\textsubscript{2} separation process (as shown in the flowchart in Figure 5) is unfeasible. Consequently, it is necessary to develop a hybrid process for separating CO\textsubscript{2} and purifying H\textsubscript{2} from IGCC synthesis gas.

### Table 2. Hydrate-based CO\textsubscript{2} separation efficiencies along with CO\textsubscript{2} concentrations in residual gas phases and hydrate slurry phases for three different CO\textsubscript{2}/H\textsubscript{2} gas mixtures at 4.0 MPa and 276.15 K in this work

<table>
<thead>
<tr>
<th>Feed gas</th>
<th>Gs uptake (mol)</th>
<th>Decomposed gas (mol)</th>
<th>CO\textsubscript{2} concentration in residual gas phase (mol%)</th>
<th>CO\textsubscript{2} concentration in hydrate slurry phase (mol%)</th>
<th>CO\textsubscript{2} separation fraction</th>
<th>CO\textsubscript{2} separation factor</th>
</tr>
</thead>
<tbody>
<tr>
<td>10.0/90.0 mol% CO\textsubscript{2}/H\textsubscript{2} gas mixture</td>
<td>0.067</td>
<td>0.0065</td>
<td>6.7</td>
<td>42.5</td>
<td>0.45</td>
<td>10.3</td>
</tr>
<tr>
<td>18.0/82.0 mol% CO\textsubscript{2}/H\textsubscript{2} gas mixture</td>
<td>0.10</td>
<td>0.016</td>
<td>10.1</td>
<td>68.5</td>
<td>0.62</td>
<td>19.4</td>
</tr>
<tr>
<td>39.2/60.8 mol% CO\textsubscript{2}/H\textsubscript{2} gas mixture</td>
<td>0.12</td>
<td>0.033</td>
<td>16.6</td>
<td>93.5</td>
<td>0.66</td>
<td>72.3</td>
</tr>
</tbody>
</table>

### 3.3. Chemical absorption of CO\textsubscript{2}

On one hand, the amounts of MEA used for absorbing CO\textsubscript{2} according to the Equation (1) depend on the amount of CO\textsubscript{2} in the residual gas mixture. On other hand, the target for adopting less MEA in the process is strictly managed owing to the environmental unfriendliness of MEA and the high energy consumption of the post-treatment for MEA. Therefore, one chemical absorption process is adopted to separate the residual gas released from the second hydrate stage. Figure 6 shows the flowchart of the hybrid CO\textsubscript{2} separation process. Compared with the flowchart shown in Figure 5, the
third stage of the hydrate-based separation process was substituted by a chemical absorption and desorption process in Figure 6. Besides, the residual gas mixture produced in the second hydrate stage was directly introduced into 30.0 wt% MEA solution in the absorption tank. Because excess MEA was added in the tank to absorb CO$_2$ according to the Equation (1), all CO$_2$ was considered to be adsorbed. In addition, in order to collect the gas mixture released from MEA solution easily at the outlet, the pressure difference of 0.5 atmosphere pressure was set between the inlet and outlet of the tank. The collected gas was almost pure H$_2$ with purity of approximately 99.4 mol%. However, H$_2$ concentration in the residual gas released from the third stage hydrate-based CO$_2$ separation process was approximately 95.2 mol%, as seen in Figure 7. Therefore, the concentration of H$_2$ released from the chemical absorption separation process was

**Figure 5.** Flowchart for three hydrate-stage CO$_2$ separation process

**Figure 6.** Flowchart for two hydrate-stage/chemical absorption hybrid CO$_2$ separation process

**Figure 7.** H$_2$ concentrations in the residual gas after third stage hydrate-based CO$_2$ separation and chemical absorption for feed gas coming from residual gas after second stage hydrate-based CO$_2$ separation in this work
4.2 mol% which was higher than that released from the third stage hydrate separation process. Thus, it illustrates that hybrid CO\textsubscript{2} separation process is feasible to obtain highly pure H\textsubscript{2}. However, the feasibility of hybrid CO\textsubscript{2} separation process for the industrial application still needs reasonable energy consumption estimation and the estimation will be made in our next work.

4. Conclusions

The equilibrium hydrate formation pressures for CO\textsubscript{2}/H\textsubscript{2} gas mixtures with CO\textsubscript{2} concentrations of 10.0, 18.0 and 39.2 mol% in the presence of 0.29 mol% TBAB solution are firstly measured. The results illustrate that the equilibrium hydrate formation pressure increases remarkably with the decrease of the concentration of CO\textsubscript{2} at the fixed temperature. Based on the above investigation, a hydrate-based CO\textsubscript{2} separation from IGCC synthesis gas is studied for its environment-friendly and economic features. In order to realize CO\textsubscript{2} separation from IGCC synthesis gas efficiently, at least three hydrate stages are required. However, with the decrease of CO\textsubscript{2} in CO\textsubscript{2}/H\textsubscript{2} gas mixture, CO\textsubscript{2} separation from the gas mixture becomes more difficult because of the increase of the corresponding equilibrium hydrate formation pressure. Especially for the third hydrate separation process, the equilibrium hydrate formation pressure shifts to high and the separation efficiency turns to quite low. Therefore, a hybrid CO\textsubscript{2} separation process with two stages hydrate separation process in conjunction with one chemical absorption process (absorption with MEA) is proposed and analyzed. The experimental results show H\textsubscript{2} concentration in the final residual gas released from the three stages hydrate CO\textsubscript{2} separation process is approximately 95.0 mol% while that released from the hybrid CO\textsubscript{2} separation process is approximately 99.4 mol%. In this work, the energy consumption estimation is not studied, and it will be analyzed in our next work.

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