Energy Consumption Analysis of Cryogenic-membrane Hybrid Process for CO2 Capture from CO2-EOR Extraction Gas

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ABSTRACT

CO2-EOR (CO2-enhanced oil recovery) is an effective method to increase oil recovery. With more and more exploitation of oilfields, a large amount of CO2 will spill out the surface, which can cause serious environmental problems. At present, high energy consumption, low CO2 purity and recovery are still the main challenges of single CO2 capture technology. Therefore, the combination of different CO2 capture technology is a reasonable choice to overcome these bottlenecks. In this work, a cryogenic-membrane hybrid process for the CO2 capture from extraction gas was proposed. In detail, the main parameters of cryogenic-membrane hybrid process (i.e., CO2 content in feed gas, compression pressure, membrane area, liquefaction temperature) which effect on CO2 properties were investigated. The simulation results indicated that the CO2 purity and recovery rate of cryogenic-membrane hybrid process can achieve 99% and 96%, respectively, and the energy consumption of the whole system was less than 850 MJ t⁻¹ CO2. Compared with membrane and cryogenic process, cryogenic-membrane hybrid process can save about 10% energy consumption. The results of this work indicate that cryogenic-membrane hybrid process has great potential for practical application.

Keywords: CO2 capture; Energy consumption; Hybrid process; Membrane process; Cryogenic process.

INTRODUCTION

Recently, environmental issues caused by large greenhouse gas emission has attracted more and more attention (Olajire, 2010; Cheng et al., 2019; Huang et al., 2019). CO2 as one of the greenhouse gases plays an important role in environmental issues (Song et al., 2019; Wu and Ku, 2019; Yang et al., 2019a; Zheng et al., 2019). China is the largest CO2 emission country in the world which greatly effect the balance of ecological of the world, therefore, it is urgent to develop an effective technology for mitigating large CO2 emission in China (Li et al., 2013, Jiang et al., 2019). CO2 capture and storage (CCS) is a prospective measurement to mitigate massive CO2 emission (Zhang et al., 2019). CO2-EOR as one of the CCS technologies, it can not only greatly improve oil recovery efficiency, but can sequester large amounts of CO2 underground (Tapia et al., 2016; Wang et al., 2018). However, with the pace of oil exploration accelerated, large amounts of injected CO2 is flow out the surface with the extraction gas. Fig. 1. demonstrated that the CO2 content in extraction gas can eventually reach about 90%. Considering the large-scale and high CO2 concentration in extraction gas, if the extraction gas is handled improperly, it would cause serious environmental problems. Therefore, it is essential for CO2 capture from extraction gas.

In recent decades, there have been developed several CO2 capture technologies, such as absorption, adsorption, membranes, cryogenic and hydrate etc. (Sreedhar et al., 2017; Vinoba et al., 2017; Yang et al., 2019). Monoethanolamine
(MEA) is the most mature technology used for CO\textsubscript{2} capture from extraction gas. However, it has some disadvantages including high energy consumption for solvent regeneration (about 3.5 MJ kg\textsuperscript{-1} CO\textsubscript{2}), equipment corrosion and environmentally unfriendly (White et al., 2003; Abu-Zahra et al., 2007; Oh et al., 2016; Liu et al., 2019b). Compared to MEA absorption, membranes is a novel technology for CO\textsubscript{2} capture from extraction gas. Despite its high energy efficiency and simplicity in equipment, it also has several drawbacks (Evangelos et al., 2017; Sreedhar et al., 2017). Membranes have strict temperature requirements, meanwhile, heavy hydrocarbons in extraction gas can cause irreversible damage to membranes (Zhang et al., 2013; Wang et al., 2017). Holmes and Ryan (1982) first proposed cryogenic distillation for natural gas purification. In recent years, it is also used for CO\textsubscript{2} capture from extraction gas. But, the cryogenic distillation has not been widely used in CO\textsubscript{2} capture from extraction gas because it needs high energy consumption which often accounts for about 50% of the total energy consumption (Li and Bai, 2012; Ebrahimzadeh et al., 2016). In order to overcome bottlenecks of these single CO\textsubscript{2} capture technologies, hybrid processes (cryogenic-hydrate, membrane-cryogenic, low temperature-membrane-cryogenic etc.) have attracted much more interests in these years (Song et al., 2018a).

**Hybrid cryogenic-hydrate process.** In 2011, Surovtseva et al. (2011) designed and proposed a hybrid cryogenic-hydrate process. The whole process can be divided into two parts: cryogenic condensation and hydrate precipitation. High pressure and low temperature were necessary for both cryogenic and hydrate stage. Cryogenic condensation was the first stage where the feed gas was cooled by four chiller and then refrigerated down to the temperature of −55°C. After first stage, the liquid CO\textsubscript{2} purity can achieve 96%. Then, hydrogen-enriched gas (non-condensable gas) passed through the second stage, named hydrate precipitation. In the second stage, the remaining CO\textsubscript{2} was captured in the form of hydrate-in-water slurry and CO\textsubscript{2} in hydrogen-enriched gas can reduce to 7–7.5%. Compared to single cryogenic or hydrate process, the low energy consumption and high CO\textsubscript{2} purity product of hybrid cryogenic-hydrate process had showed great potential (Sreenivasulu et al., 2015). Recently, Willson et al. (2012) proposed using hydrate unit, such as tetrabutylammonium chloride(TBAC), tetrabutylphosphonium chloride(TBPC) and tetrabutylphosphonium chloride(TBAB) etc. to improve hydrate stage and reduce high pressure.

**Hybrid membrane-cryogenic process.** In 2012, Belaissaoui et al. (2012a). looked forward to combining membrane and cryogenic process together. Meanwhile, Belaissaoui et al. (2012b). also validated the feasibility of the hybrid membrane-cryogenic process. The hybrid process was composed of membrane separation (first stage) and cryogenic condensation (second stage). Through simulation and analysis, the hybrid process can achieve 90% CO\textsubscript{2} purity and 85% CO\textsubscript{2} recovery (Belaissaoui et al., 2012b). Compared to conventional MEA absorption, the energy consumption was lower than 2 GJ t\textsuperscript{-1} CO\textsubscript{2} (Belaissaoui et al., 2012b). In 2014, Anantharaman et al. (2014) developed a hybrid membrane-liquefaction process for post-combustion CO\textsubscript{2} capture. The hybrid process contained two units: single membrane unit and low-temperature unit. The feed gas first passed through single membrane to capture 90% CO\textsubscript{2} and the permeate gas (CO\textsubscript{2}-rich gas) then compressed and cooled to a low temperature. Finally, the liquid CO\textsubscript{2} product was separated by separator. Compared with membrane process, the hybrid process consumed less compression power. According to techno-economic analysis, the hybrid process was 9% more cost-efficient than MEA absorption process (Belaissaoui et al., 2012b; Song et al., 2018a).

**Other hybrid process.** In 2014, Freeman et al. (2014) proposed a hybrid membrane-absorption process. In this hybrid process, the absorber initially removed half of the CO\textsubscript{2} in the feed gas and next the feed gas was purified by membrane. Therefore, the total CO\textsubscript{2} capture rate in the hybrid process can achieve 90%. In 2017, Song et al. (2017) improved and proposed a low temperature-membrane-cryogenic hybrid process. The feed gas was cooled to a low temperature which can improve hollow membrane performance, then the permeate gas (CO\textsubscript{2}-rich gas) was sent into cryogenic unit for next purification. The result showed that hybrid process CO\textsubscript{2} recovery can keep at over 90% and energy consumption can be reduced to 1.7 MJ kg\textsuperscript{-1} CO\textsubscript{2} (Song et al., 2017).

Although the above hybrid CO\textsubscript{2} capture processes have many advantages, there are also several drawbacks to handle (Scholz et al., 2013). The hybrid cryogenic-hydrate process, it only limits in laboratory scale test and the whole process has low CO\textsubscript{2} recovery (Surovtseva et al., 2011). Meanwhile, low temperature-membrane-cryogenic hybrid process is extremely sensitive to humidity and low temperature can also decrease membrane productivity (Hasse et al., 2014; Song et al., 2018a).

Therefore, it is vital to find a suitable hybrid process for CO\textsubscript{2} capture from extraction gas, a hybrid membrane-cryogenic was proposed in this work. The feed gas was initially treated by cryogenic unit, then the non-condensable gas was passed through for further purification, and the process diagram of hybrid membrane-cryogenic was shown in Fig. 4. The effect of several main parameters (CO\textsubscript{2} content in feed gas, compression pressure, membrane area, liquefaction temperature) was analyzed. Meanwhile, CO\textsubscript{2} capture performance (CO\textsubscript{2} purity, CO\textsubscript{2} recovery and energy consumption) compared to membrane and cryogenic process were also investigated. That will have great significance for hybrid CO\textsubscript{2} capture from extraction gas.

**PROCESS DESCRIPTION**

**The Properties of Extraction Gas**

Table 1 showed the main parameters and compositions of extraction gas from a single well of Shengli oilfield. The characteristics of extraction gas are illustrated: (1) Extraction gas has large flow rate. (2) Extraction gas has high partial pressure, high CO\textsubscript{2} concentration. (3) Extraction gas has high hydrocarbon content and water. For membrane process, high hydrocarbon and water will block the membrane and cause irreversible destruction. For cryogenic process, it will corrode pipes and equipment. Therefore, in order to protect equipment and ensure safe and reliable operation of the
Table 1. The properties of extraction gas.

<table>
<thead>
<tr>
<th>Main parameters</th>
<th>Numerical value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flow rate (kg h(^{-1}))</td>
<td>6000</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>25</td>
</tr>
<tr>
<td>Pressure (Mpa)</td>
<td>0.3</td>
</tr>
<tr>
<td>Extraction gas composition (wt %)</td>
<td></td>
</tr>
<tr>
<td>CO(_2)</td>
<td>90.15</td>
</tr>
<tr>
<td>CH(_4)</td>
<td>7.18</td>
</tr>
<tr>
<td>C(_2)H(_8)</td>
<td>1.15</td>
</tr>
<tr>
<td>C(_3)H(_8)</td>
<td>1.10</td>
</tr>
<tr>
<td>i-C(_4)</td>
<td>0.020</td>
</tr>
<tr>
<td>n-C(_4)</td>
<td>0.081</td>
</tr>
<tr>
<td>i-C(_5)</td>
<td>0.049</td>
</tr>
<tr>
<td>n-C(_5)</td>
<td>0.050</td>
</tr>
<tr>
<td>C(_6)+</td>
<td>0.1</td>
</tr>
<tr>
<td>N(_2)</td>
<td>0.12</td>
</tr>
</tbody>
</table>

device, the extraction gas must pass through pretreatment process for removing hydrocarbon and water. However, pretreatment process is not the scope of this paper.

Cryogenic-membrane Hybrid Process

Cryogenic process was first proposed by Holmes and Ryan (1982) for natural gas purification. In this paper, the whole process was modified for CO\(_2\) capture from extraction gas and was shown in Fig. 2. Feed gas is initially compressed and cooled by compression system, and then get into pre-cooler and liquefier for further cooling. Finally, liquefied gas is sent into distillation tower for CO\(_2\) and CH\(_4\) separation. The steam which pass through distillation tower is divided into two parts: top tower (including CO\(_2\) and CH\(_4\)) and bottom tower (CO\(_2\) product). Top tower product is also called non-condensable gas, it flows out condenser and then get into pre-cooler for releasing cold capacity. The bottom tower product is high purity CO\(_2\), which released by reboiler at the bottom of distillation tower.

Fig. 3 showed non-condensable gas flow rate and CO\(_2\) content in non-condensable gas varied with different CO\(_2\) content in feed gas which illustrated that non-condensable gas still had large flow rate (about 1400 kg h\(^{-1}\)) and high CO\(_2\) concentration (about 65%). In order to using non-condensable gas rationally and avoiding CO\(_2\) excessive discharge, it is necessary for CO\(_2\) purification from non-condensable gas. Therefore, Fig. 4 illustrated a proposed cryogenic-membrane hybrid process for CO\(_2\) capture from extraction gas. The cryogenic-membrane hybrid process contains cryogenic unit which we mentioned in Fig. 2 and membrane unit (one-stage membrane), the non-condensable gas from cryogenic unit is compressed first and then sent into membrane unit for further purification, which can obtain high purity CH\(_4\) (S13) and CO\(_2\) (S14). Table 2 shows main parameters of cryogenic-membrane hybrid process.

Membrane CO\(_2\) Capture Process

Three-stage membrane process can achieve the lower energy consumption about 1.2 MJ kg\(^{-1}\) CO\(_2\) (Scholes et al., 2014) compared with single-stage membrane. In the work, a typical three-stage membrane process were used to compare with the cryogenic-membrane hybrid process.

The three-stage membrane CO\(_2\) capture process was shown in Fig. 5 (Liu et al., 2019a). The feed gas initially passes through first membrane (M-I) for preliminary purification which the steam after M-I is divided into two parts: the permeate gas (high CO\(_2\) content stream) and the retentate gas (rich-CH\(_4\) stream). On the one hand, the permeate gas is compressed into the second membrane (M-II) for CO\(_2\) further purification. High CO\(_2\) product is assembled in the permeate side and retentate gas (including CH\(_4\) and few CO\(_2\)) returns to M-I. On the other hand, the retentate gas after M-I...
Fig. 3. Effect of CO₂ content on non-condensable gas flow rate and CO₂ content in non-condensable gas.

Fig. 4. Process flow diagram of cryogenic-membrane hybrid CO₂ capture process.

Table 2. Key nodes information of cryogenic-membrane hybrid CO₂ capture process.

<table>
<thead>
<tr>
<th>Properties</th>
<th>Compression system</th>
<th>Liquefier</th>
<th>Condenser</th>
<th>Reboiler</th>
<th>Membrane</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (°C)</td>
<td>S1</td>
<td>S2</td>
<td>S3</td>
<td>S4</td>
<td>S5</td>
</tr>
<tr>
<td></td>
<td>25</td>
<td>156.9</td>
<td>40</td>
<td>139.4</td>
<td>40</td>
</tr>
<tr>
<td>Pressure (MPa)</td>
<td>0.3</td>
<td>1.2</td>
<td>1.5</td>
<td>3.19</td>
<td>3.14</td>
</tr>
<tr>
<td>Mass flow (kg h⁻¹)</td>
<td>6000</td>
<td>6000</td>
<td>6000</td>
<td>6000</td>
<td>6000</td>
</tr>
<tr>
<td>CO₂ (wt %)</td>
<td>90</td>
<td>90</td>
<td>90</td>
<td>90</td>
<td>90</td>
</tr>
<tr>
<td>CH₄ (wt %)</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
</tbody>
</table>
Fig. 5. Process flow diagram of membrane CO2 capture process.

sent to the third membrane (M-Ⅲ) for CH4 purification which high CH4 product assembled in retentate side and the permeate side (including few CH4 and CO2) gas recycles to the first membrane (M-Ⅰ). In the whole process, the retentate gas of M-Ⅱ and the permeate gas of M-Ⅲ mix together in Mixer-2 and then recycle with feed gas in mixer-1 for next cycle.

PROCESS SIMULATION

Product Properties

There are two main parameters to estimate CO2 capture performance: CO2 purity and CO2 recovery. The calculation of CO2 purity and CO2 recovery is summarized as follows (Zhao et al., 2008):

\[
\text{CO2\ purity} = \frac{m_{\text{CO2}}}{m_{\text{product}}} \tag{1}
\]

where, \(m_{\text{CO2}}\) is the CO2 mass flow rate in the outlet of the capture process (kg h\(^{-1}\)) and \(m_{\text{product}}\) is the mass flow rate of product in the process outlet (kg h\(^{-1}\)).

\[
\text{CO2\ recovery} = \frac{V_{\text{out.CO2}} \times \gamma_{\text{CO2}}}{V_{\text{feed.CO2}} \times \chi_{\text{CO2}}} \tag{2}
\]

where, \(V_{\text{out.CO2}}\) is the CO2 volume flow rate in the outlet of the capture process (m\(^3\) h\(^{-1}\)), \(\gamma_{\text{CO2}}\) is the CO2 volume fraction in the outlet of the capture process, \(V_{\text{feed.CO2}}\) is the CO2 volume flow rate at the feed side (m\(^3\) h\(^{-1}\)) and \(\chi_{\text{CO2}}\) is the CO2 volume fraction at the feed side.

Energy Consumption

Energy consumption is also an important parameter for evaluating CO2 capture performance. The calculation of energy consumption is shown as follows (Song et al., 2012):

\[
E_{\text{hybrid,total}} = E_{\text{cryogenic}} + E_{\text{membrane}} \tag{3}
\]

where, \(E_{\text{hybrid,total}}\) is the total energy consumption of the hybrid process, \(E_{\text{cryogenic}}\) is the energy consumption of the cryogenic process and \(E_{\text{membrane}}\) is the energy consumption of the membrane unit in the hybrid process.

\[
E_{\text{cryogenic}} = E_{\text{compression system}} + E_{\text{phase change}} \tag{4}
\]

\[
E_{\text{phase change}} = m_{\text{CO2}} \times h_{\text{phase change}} \tag{5}
\]

\[
E_{\text{membrane}} = E_{\text{compressor}} + E_{\text{cooler}} \tag{6}
\]

where, \(m_{\text{CO2}}\) is the mass of the liquefaction CO2 and \(h_{\text{phase change}}\) is the heat of phase change for CO2.

Simulation Settings

The hybrid process simulation was based on Aspen HYSYS. The simulation data and key nodes information were shown in Tables 2 and 3. The following assumptions were used to simplify the calculation: (1) Peng-Robinson (PR) equation was selected as the thermodynamic calculation method; (2) In order to avoid the influence of heavy hydrocarbons, the extraction gas only contained two kind of gas: CO2 and CH4; (3) The extraction gas did not contain any form of water; (4) The adiabatic efficiency of compressor was 75%; (5) The whole system had no heat loss.

RESULTS AND DISCUSSION

Effect of CO2 Content on CO2 Capture Performance

Fig. 6. showed the effect of CO2 content in feed gas on (a) CO2 purity and (b) CO2 recovery rate of cryogenic process, hybrid process and membrane process. The basic
As shown in Fig. 6(a), when CO₂ content in feed gas was 80%, CO₂ purity was lower than 95% in cryogenic and membrane process. However, as the CO₂ content increased, the CO₂ purity increased accordingly. When the CO₂ content was above 90%, the CO₂ purity of cryogenic and membrane process can reach 99%. For the hybrid process, the CO₂ purity can be kept at 99% when CO₂ content in feed gas varied from 80% to 92%. Fig. 6(b) showed the effect of CO₂ content on CO₂ recovery of cryogenic process, hybrid process and membrane process which indicated that the CO₂ recovery rate of the hybrid process was higher than the cryogenic and membrane process. For hybrid process, CO₂ recovery rate can reach 96% when CO₂ content was more than 83%. Even for low CO₂ content (80%), CO₂ recovery rate of hybrid process can also achieve at 88%. Combining Figs. 6(a) and 6(b), hybrid process can reach higher CO₂ purity and CO₂ recovery compared to other two processes. That can be explained that non-condensable gas which contained about 60% CO₂ was further purified by membrane unit in the hybrid process. Therefore, high CO₂ purity and CO₂ recovery can be achieved.

Table 3. Simulation data and compositions of feed gas.

<table>
<thead>
<tr>
<th>Main parameters</th>
<th>Hybrid process</th>
<th>Membrane process</th>
<th>Cryogenic process</th>
</tr>
</thead>
<tbody>
<tr>
<td>Feed gas flow rate (kg h⁻¹)</td>
<td>6000</td>
<td>6000</td>
<td>6000</td>
</tr>
<tr>
<td>Feed gas temperature (℃)</td>
<td>25</td>
<td>25</td>
<td>25</td>
</tr>
<tr>
<td>Inlet pressure (MPa)</td>
<td>0.3</td>
<td>0.3</td>
<td>0.3</td>
</tr>
<tr>
<td>Feed gas composition (wt %)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CO₂</td>
<td>90</td>
<td>90</td>
<td>90</td>
</tr>
<tr>
<td>CH₄</td>
<td>10</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>Liquefaction temperature (℃)</td>
<td>-27</td>
<td>-</td>
<td>-27</td>
</tr>
<tr>
<td>CO₂/CH₄ selectively</td>
<td>42.75</td>
<td>42.75</td>
<td>-</td>
</tr>
</tbody>
</table>

The energy consumption of cryogenic, hybrid and membrane process under different CO₂ content in feed gas was shown in Fig. 7. It can be seen that energy consumption of membrane process and hybrid process both decreased as the CO₂ content in feed gas increased. However, with CO₂ content in feed gas increasing, the energy consumption in cryogenic process increased first and then decreased. For hybrid process, the energy consumption was far more lower than the cryogenic and membrane process under the same CO₂ content. Especially when the CO₂ content in feed gas was 84%, hybrid process can save 15% energy consumption at most compared with cryogenic and membrane process. For high CO₂ content in feed gas, the energy consumption of hybrid process was also far more less than cryogenic and membrane process. Thus, hybrid process can significantly reduce energy consumption when CO₂ content in extraction gas was above 80%.

**Effect of Compression Pressure on CO₂ Capture Performance**

Fig. 8 illustrate the effect of compression pressure on CO₂ purity (a) and CO₂ recovery (b) of cryogenic, hybrid and membrane process. The basic simulation conditions were as follows: feed gas flow rate was 6000 kg h⁻¹, feed gas inlet pressure was 0.3 MPa, CO₂ content in feed gas was 90%. Fig. 8(a) showed the effect of compression pressure on cryogenic, hybrid and membrane process. With the increase of compression pressure, the CO₂ purity of cryogenic and membrane process also increased. But, the increasing rate decreased gradually. When the compression pressure was above 80%, CO₂ purity was lower than 95% in cryogenic and membrane process. The basic simulation conditions were as follows: feed gas flow rate was 6000 kg h⁻¹, feed gas inlet pressure was 0.3 MPa, CO₂ content in feed gas was 90%.

Fig. 8(a) showed the effect of compression pressure on cryogenic, hybrid and membrane process. With the increase of compression pressure, the CO₂ purity of cryogenic and membrane process also increased. But, the increasing rate decreased gradually. When the compression pressure was 4.0 MPa, the CO₂ purity of cryogenic and membrane process reached the highest 94%. Meanwhile, as shown in Fig. 8(b), the CO₂ recovery rate of cryogenic and membrane process increased accordingly with the compression pressure increasing. The highest CO₂ recovery of cryogenic and membrane process was 92% and 94%, respectively. This proved that the high compression pressure enhanced the purification efficiency of the cryogenic process and membrane permeation of the membrane process. Compared with the cryogenic and membrane process, the hybrid process can reach higher CO₂ purity (about 96%) and higher CO₂ recovery (about 98%) even in a low compression condition. When the compression was 3.1 Mpa, the CO₂ recovery rate in the hybrid process was 98%. However, as the compression pressure was higher than 3.1 Mpa, the influence of the compression pressure on hybrid process became weakly.

The influence of compression pressure on energy consumption of cryogenic process, hybrid process and membrane process was shown in Fig. 9. which indicated the hybrid process can save 120 MJ t⁻¹ CO₂ at most compared with the cryogenic and membrane process. For cryogenic and hybrid process, energy consumption decreased firstly (from 2.4 MPa to 3.1 MPa) and then increased. However, energy consumption of the membrane process increased steadily with the increase of compression pressure. It was because high compression pressure can obtain better CO₂ purity and recovery. But, compression pressure was not the higher the better, over-high compression pressure can lead to an increasing cooling load and CH₄ leaking out in the permeate side. When compression pressure increased higher than 3.1 MPa, the energy consumption of these three processes grew rapidly. Therefore, for the hybrid process, the reasonable compression pressure was 3.1 MPa.

**Effect of Membrane Area on CO₂ Capture Performance**

Fig. 10. showed the influence of membrane area on CO₂ purity (a) and CO₂ recovery (b) of membrane process and hybrid process. The basic simulation conditions were as follows: feed gas flow rate was 6000 kg h⁻¹, feed gas inlet pressure was 0.3 MPa, CO₂ content in feed gas was 90%, compression pressure was 3.1 MPa.
Fig. 6. Effect of CO$_2$ content in feed gas on (a) CO$_2$ purity and (b) CO$_2$ recovery of cryogenic process, hybrid process and membrane process.

Fig. 7. Effect of CO$_2$ content in feed gas on energy consumption of cryogenic process, hybrid process and membrane process.
Fig. 8. Effect of compression pressure on (a) CO$_2$ purity and (b) CO$_2$ recovery of cryogenic process, hybrid process and membrane process.

Fig. 9. Effect of compression pressure on energy consumption of cryogenic process, hybrid process and membrane process.
The comparison of CO₂ purity between membrane process and hybrid process under different membrane area was shown in Fig. 10(a) which illustrated that the CO₂ purity of membrane process decreased from 98% to 84% with the membrane area increased from 1300 m² to 2000 m². However, CO₂ purity of hybrid process improved significantly and can keep at a high level (about 99%) even in the small membrane area (1300 m²). Fig. 10(b) showed the effect of membrane area on CO₂ recovery of membrane process and hybrid process. As the increasing of membrane area, the CO₂ recovery of membrane process and hybrid process increased accordingly. But, CO₂ recovery of hybrid process was higher than the membrane process. When membrane area was 1300 m², CO₂ recovery of hybrid process was 40% higher than membrane process. Combing Figs. 10(a) and 10(b), CO₂ purity and recovery of membrane process related inversely with the membrane area increasing. Increasing the membrane area can get better CO₂ recovery, but lower CO₂ purity. For hybrid process, CO₂ recovery rate increased from 90% to 97% as the membrane area increased from 1300 m² to 2000 m². When the membrane area increased over 1700 m², CO₂ recovery rate kept around 97%.

Fig. 11. showed the influence of membrane area on energy consumption of membrane process and hybrid process. For membrane process, energy consumption initially decreased for the membrane area increasing from 1300 m² to 1700 m² and then increased for the membrane area increasing from 1700 m² to 2000 m². The lowest energy consumption of membrane process was 1035 MJ t⁻¹ CO₂ when the membrane area was set as 1700 m². For hybrid process, it had the same energy consumption trend as membrane process. However, the hybrid process can save about 10% energy consumption compared to the membrane process. When the membrane area changed from 1300 to 1700 m², the energy consumption decreased from 1000 to 990 MJ t⁻¹ CO₂. That was because at the beginning the effective permeation area increased for the membrane area increasing, which can increase the CO₂ product purity and decrease the capture cost. But, larger

![Fig. 10. Effect of membrane area on (a) CO₂ purity and (b) CO₂ recovery of membrane process and hybrid process.](image-url)
membrane area was not always good for the hybrid system. With the membrane area increasing from 1800 to 2000 m², the energy consumption then increased from 995 MJ t⁻¹ CO₂ to 1005 MJ t⁻¹ CO₂. The reason was that over-large membrane area can cause CH₄ permeating the membrane, lower down the CO₂ product purity and increase the capture cost.

**Effect of Liquefaction Temperature on CO₂ Capture Performance**

The effect of liquefaction temperature on CO₂ purity (a) and CO₂ recovery (b) of cryogenic process and hybrid process was shown in Fig. 12. The basic simulation conditions were as follows: feed gas flow rate was 6000 kg h⁻¹, feed gas inlet pressure was 0.3 MPa, CO₂ content in feed gas was 90%, compression pressure was 3.1 MPa and membrane area was 1700 m².

Fig. 12(a) showed the effect of liquefaction temperature on CO₂ purity of cryogenic process and hybrid process. CO₂ purity of cryogenic process and hybrid process had the same increasing trend. For cryogenic process, CO₂ purity increased from 90% to 96% as the liquefaction temperature changed from –22°C to –40°C. For hybrid process, when liquefaction temperature decreased from –22°C to –40°C, CO₂ purity increased from 93% to 98%. Compared with cryogenic process, CO₂ purity of hybrid process was always higher than cryogenic process, even in a high liquefaction temperature (–22°C). That was because low temperature can improve CO₂/CH₄ selectively of membrane unit. The effect of liquefaction temperature on CO₂ recovery of the cryogenic process and hybrid process was shown in Fig. 12(b) which indicated that the growth rate for two processes slowed down gradually when liquefaction temperature decreased from –22°C to –40°C. As liquefaction temperature was –27°C, the highest CO₂ recovery for cryogenic process and hybrid process was 92% and 97%, respectively. When liquefaction temperature exceeded –27°C, the CO₂ recovery kept at the highest level (about 97%)

The comparison of energy consumption between cryogenic process and hybrid process under different liquefaction was shown in Fig. 13 which indicated that energy consumption of cryogenic and hybrid process initially decreased and then increased with the decreasing of liquefaction temperature. When liquefaction temperature decreased from –22°C to –27°C, the energy consumption for cryogenic and hybrid process decreased rapidly. Meanwhile, as the liquefaction temperature decreased from –28°C to –40°C, the energy consumption surged rapidly. Combing Fig. 12., the reason was that when the liquefaction temperature arrived at a certain range, the CO₂ product and purity increased accordingly and energy consumption decreased rapidly, but as the liquefaction temperature exceeded –27°C, the CO₂ product and purity unchanged, therefore, it caused huge extra energy consumption. When liquefication temperature was –27°C, cryogenic-membrane hybrid process had the lowest energy consumption and around 14% energy consumption was saved compared to cryogenic process.

**Technical Evaluation**

In order to estimate technical feasibility of cryogenic-membrane hybrid process, the researches of Song et al. (2017) for low temperature-membrane-cryogenic for CO₂ capture process from flue gas and membrane-cryogenic process for biogas upgrading (Song et al., 2018b) were used to compare with this work. Table 4 depicted the main parameters and capture performance of three different hybrid processes which can prove technical feasibility of this research. (1) CO₂ content in extraction gas was 90% which was far more than flue gas and biogas. But, according to Belaissoua et al. (2012b), when CO₂ content was 90%, cryogenic unit can reach a high CO₂ purity, high CO₂ recovery and lowest energy consumption. Therefore, cryogenic unit as first step to capture CO₂ from extraction gas was reasonable. (2) In this research, –27°C was the operating temperature of membrane unit which was proved by the experiment of Song et al. (2017) that indicates low temperature (about –30°C) that indicates low temperature can effectively improve membrane capture performance. (3) Compared with low
Fig. 12. Effect of liquefaction temperature on (a) CO₂ purity and (b) CO₂ recovery of cryogenic process and hybrid process.

Fig. 13. Effect of liquefaction temperature on energy consumption of cryogenic process and hybrid process.
temperature-membrane-cryogenic hybrid process (Song et al., 2017) and membrane-cryogenic hybrid process (Song et al., 2018b), the cryogenic-membrane hybrid process in this research can achieve better capture performance.

Through the comparison of the work of Song et al. (2017, 2018b), the membrane-cryogenic hybrid process for CO2 capture from extraction gas in this research can significantly reduce energy consumption and increase CO2 product properties.

CONCLUSIONS

A cryogenic-membrane hybrid process for CO2 capture from extraction gas was developed. In order to enhance CO2 capture performance, membrane unit was placed behind the cryogenic process to purify the non-condensable gas. The CO2 capture performance of cryogenic-membrane hybrid process had been studied by changing several main parameters (CO2 content in feed gas, compression pressure, membrane area and liquefaction temperature). Meanwhile, the CO2 capture performance of cryogenic process, cryogenic-membrane hybrid process and membrane process were conducted, and the technical feasibility of cryogenic-membrane hybrid process was also evaluated. The results illustrated that hybrid process can effectively improve CO2 purity and recovery rate, and can save about 10% energy consumption compared to cryogenic and membrane process. For high CO2 concentration extraction gas (above 90%), The CO2 purity and recovery of hybrid process can reach 99% and 96%, respectively. The energy consumption can reach below 850 MJ t–1CO2.

This work proposed a method for CO2 capture from extraction gas and analyzed some important parameters. However, the membrane materials and environmental influence is still a serious issue for cryogenic-membrane process application. In the future work, these parameters will also be undertook.

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REFERENCES


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