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,

Fig. 1. CO₂ content in extraction gas of Shengli oilfield.

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

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(MEA) is the most mature technology used for CO₂ capture from extraction gas. However, it has some disadvantages including high energy consumption for solvent regeneration (about 3.5 MJ kg⁻¹ CO₂), 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 CO₂ 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 CO₂ capture from extraction gas. But, the cryogenic distillation has not been widely used in CO₂ 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 CO₂ 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 CO₂ 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 CO₂ was captured in the form of hydrate-in-water slurry and CO₂ in hydrogen-enriched gas can reduce to 7–7.5%. Compared to single cryogenic or hydrate process, the low energy consumption and high CO₂ 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% CO₂ purity and 85% CO₂ recovery (Belaissaoui *et al.*, 2012b). Compared to conventional MEA absorption, the energy consumption was lower than 2 GJ t⁻¹ CO₂ (Belaissaoui *et al.*, 2012b). In 2014, Anantharaman *et al.* (2014) developed a hybrid membrane-liquefaction process for post-combustion CO₂

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% CO₂ and the permeate gas (CO₂-rich gas) then compressed and cooled to a low temperature. Finally, the liquid CO₂ 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 CO₂ in the feed gas and next the feed gas was purified by membrane. Therefore, the total CO₂ 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 (CO₂-rich gas) was sent into cryogenic unit for next purification. The result showed that hybrid process CO₂ recovery can keep at over 90% and energy consumption can be reduced to 1.7 MJ kg⁻¹ CO₂ (Song *et al.*, 2017)

Although the above hybrid CO₂ 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 CO₂ 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 CO₂ 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 (CO₂ content in feed gas, compression pressure, membrane area, liquefaction temperature) was analyzed. Meanwhile, CO₂ capture performance (CO₂ purity, CO₂ recovery and energy consumption) compared to membrane and cryogenic process were also investigated. That will have great significance for hybrid CO₂ 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 CO₂ 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.

Main parameters	Numerical value
Flow rate (kg h ⁻¹)	6000
Temperature (°C)	25
Pressure (Mpa)	0.3
Extraction gas composition (wt %)	
CO ₂	90.15
CH ₄	7.18
C ₂ H ₆	1.15
C ₃ H ₈	1.10
i-C ₄	0.020
n-C ₄	0.081
i-C ₅	0.049
n-C ₅	0.050
C ₆₊	0.1
N ₂	0.12

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 CO₂ 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 CO₂ and CH₄ separation. The steam which pass through distillation tower is divided into two parts: top tower (including CO₂ and CH₄) and bottom tower (CO₂) 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 CO₂, which released by reboiler at the bottom of distillation tower.

Fig. 3 showed non-condensable gas flow rate and CO₂ content in non-condensable gas varied with different CO₂ content in feed gas which illustrated that non-condensable gas still had large flow rate (about 1400 kg h⁻¹) and high CO₂ concentration (about 65%). In order to using non-condensable gas rationally and avoiding CO₂ excessive discharge, it is necessary for CO₂ purification from non-condensable gas. Therefore, Fig. 4 illustrated a proposed cryogenic-membrane hybrid process for CO₂ 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 CH₄ (S13) and CO₂ (S14). Table 2 shows main parameters of cryogenic-membrane hybrid process.

Membrane CO₂ Capture Process

Three-stage membrane process can achieve the lower energy consumption about 1.2 MJ kg⁻¹ CO₂ (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 CO₂ 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 CO₂ content stream) and the retentate gas (rich-CH₄ stream). On the one hand, the permeate gas is compressed into the second membrane (M-II) for CO₂ further purification. High CO₂ product is assembled in the permeate side and retentate gas (including CH₄ and few CO₂) returns to M-I. On the other hand, the retentate gas after M-I is

Fig. 2. Process flow diagram of cryogenic CO₂ capture process.

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.

Properties	Compression system					Liquefier		Condenser	Reboiler	Membrane		
	S1	S2	S3	S4	S5	S6	S7	S8	S9	S12	S13	S14
Temperature (°C)	25	156.9	40	139.4	40	35	-21	-27	-4.3	-27	-27	-27
Pressure (MPa)	0.3	1.2	1.5	3.19	3.14	3.12	3.1	3.1	3.1	3.1	3.1	0.1
Mass flow (kg h ⁻¹)	6000	6000	6000	6000	6000	6000	6000	883	5117	883	174	709
Composition (wt %)												
CO ₂	90	90	90	90	90	90	90	67	98	67	4.1	99.2
CH ₄	10	10	10	10	10	10	10	33	2	33	95.9	0.8

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):

$$CO_{2,purity} = \frac{m_{CO_2}}{m_{product}} \quad (1)$$

where, m_{CO_2} is the CO₂ mass flow rate in the outlet of the capture process (kg h⁻¹) and $m_{product}$ is the mass flow rate of product in the process outlet (kg h⁻¹).

$$CO_{2,recovery} = \frac{V_{out,CO_2} \times \gamma_{CO_2}}{V_{feed,CO_2} \times \chi_{CO_2}} \quad (2)$$

where, V_{out,CO_2} is the CO₂ volume flow rate in the outlet of the capture process (m³ h⁻¹), γ_{CO_2} is the CO₂ volume fraction in the outlet of the capture process, V_{feed,CO_2} is the CO₂ volume flow rate at the feed side (m³ h⁻¹) and χ_{CO_2} 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):

$$E_{hybrid,total} = E_{cryogenic} + E_{membrane} \quad (3)$$

here, $E_{hybrid,total}$ is the total energy consumption of the hybrid process, $E_{cryogenic}$ is the energy consumption of the cryogenic process and $E_{membrane}$ is the energy consumption of the membrane unit in the hybrid process.

$$E_{cryogenic} = E_{compression\ system} + E_{phase\ change} \quad (4)$$

$$E_{phase\ change} = m_{CO_2} \times h_{phase\ change} \quad (5)$$

$$E_{membrane} = E_{compressor} + E_{cooler} \quad (6)$$

where, m_{CO_2} is the mass of the liquefaction CO₂ and $h_{phase\ change}$ 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

Table 3. Simulation data and compositions of feed gas.

Main parameters	Hybrid process	Membrane process	Cryogenic process
Feed gas flow rate (kg h ⁻¹)	6000	6000	6000
Feed gas temperature (°C)	25	25	25
Inlet pressure (MPa)	0.3	0.3	0.3
Feed gas composition (wt %)			
CO ₂	90	90	90
CH ₄	10	10	10
Liquefaction temperature (°C)	-27	-	-27
CO ₂ /CH ₄ selectively	42.75	42.75	-

simulation conditions were as follows: feed gas flow rate was 6000 kg h⁻¹, feed gas inlet pressure was 0.3 MPa.

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.

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. illustrated 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 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 CO₂ content in feed gas on (a) CO₂ purity and (b) CO₂ recovery of cryogenic process, hybrid process and membrane process.

Fig. 7. Effect of CO₂ content in feed gas on energy consumption of cryogenic process, hybrid process and membrane process.

Fig. 8. Effect of compression pressure on (a) CO₂ purity and (b) CO₂ 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. Combining 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.

Fig. 11. Effect of membrane area on energy consumption of membrane process and hybrid process.

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. Combining 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 liquefaction 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 Belaisaoui *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) 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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