Experimental Study on Postcombustion Systems Including a Hollow Fiber Membrane and a Packed Column

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Cite This: *ACS Omega* 2020, 5, 17692–17702

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ABSTRACT: In this work, a comprehensive lab-scale carbon capture installation was established to study the separation performances of CO₂/N₂ systems for the postcombustion technology. Four kinds of mono-/two-stage carbon capture methods containing membrane separation and chemical absorption processes were investigated. The result shows that the CO₂ capture performance of the one-stage membrane separation method (*Memb*) exhibits a profitable CO₂ removal efficiency but defective CO₂ concentration, while the one-stage chemical absorption method (*Chem*) indicates both CO₂ removal efficiency and CO₂ purity of more than 95.0% but suffers a regeneration heat of at least 2.7 MJ/t CO₂. The CO₂ purity of the two-stage membrane separation method (*Memb–Memb*) is 46.2% higher than the *Memb* method because of the additional membrane pretreatment. Two-stage methods have a superior gas recovery efficiency of over 99.0%, which is dramatically higher than the homogeneous *Memb* method. In addition, the investigation on the hybrid chemical absorption–membrane separation method (*Memb–Chem*) provides an alternative approach to reduce the mass transfer and solve the problems caused by an unequal mass flow distribution.

1. INTRODUCTION

Climate change has become a major threat to humankind survival in recent years.¹ With the rapid growth of industry and population, the contradiction between greenhouse gas emissions and economic development becomes a severe topic.² This challenge is particularly prominent for developing countries, such as China, the biggest contributor to global carbon emission.³ For a long time, coal has occupied the main status of China's energy resource structure and produced a substantial amount of CO₂. In order to ensure the supply of sustainable energy, China has been committed to reducing the carbon emission from coal combustion from power plants.^{4,5}

The CO₂ capture process in coal-fired power plants mainly corresponds to postcombustion capture.^{6,7} Compared with other fuels, the flue gas generated from coal combustion has the characteristics of an enormous gas flow rate, a low CO₂ partial pressure, and a high discharge temperature, and the working condition is obviously different from other chemical processes, such as ammonia synthesis and natural gas purification.⁸ At present, the development of other carbon captures technologies, such as adsorption, absorption, and membrane separation, has made great progress in lab and industry, which provides more options for satisfying the carbon capture requirements in the postcombustion process.^{9,10}

Membrane separation is now considered as one of the most prospective carbon capture methods because of the low

pollution and energy consumption.^{11,12} However, in research and application processes, the separation performance and operation stability are more sensitive to the change of the working conditions, especially during the start-up and shut-down procedures.¹³ At present, few of the CO₂ separation membranes can perfectly meet the demands of permeability, selectivity, and manufacturing cost. Besides that, the chemical absorption method has been widely implemented in the carbon capture process according to the reaction between CO₂ and absorbents^{14,15} (e.g., mono-/di-/tri-ethanolamine and ionic liquids). It demonstrated some advantages about reliability and suitability, but the defects including superfluous energy consumption restricted its development. It was reported that every increase of 1.0 MJ/kg CO₂ in regeneration energy will cause the decline of power generation efficiency by 2.0%.¹⁶

As shown above, it is difficult for the onefold method to completely meet the demands of carbon removal efficiency and economy in the postcombustion process, thus some concept of

Received: May 14, 2020

Accepted: June 25, 2020

Published: July 7, 2020

Figure 1. Schematic diagram of carbon capture installation containing membrane and chemical processes.

novel technical routes such as multistage hybrid methods were proposed as a practically promising way.¹⁷ However, the coupling characteristics of each unit in system integration are hard to meet the requirement because of the strict requirements of the hybrid process on design parameters, and the combination performance between different carbon removal units is vulnerably challenged by some factors, such as differences in mass-transfer rates. Hence, most of the research only stays in the stage of theoretical research and numerical simulation so far.

Therefore, in this work, a comprehensive lab-scale carbon capture installation was established to study the properties of one-/two-stage carbon capture methods containing membrane separation and chemical absorption processes for the postcombustion technology. Four kinds of configurations were proposed, and the operation coefficients, such as the feed gas pressure and amine solution flow rate, were investigated to determine the overall CO₂ removal performances. Eventually, a comparison of the critical parameters was proposed to evaluate the technical properties of different carbon capture methods.

2. EXPERIMENTAL INSTALLATION

The experimental carbon capture system is schematically shown in Figure 1. There are mainly five subsystems in the overall installation.

- **Gas supply system:** Carbon dioxide 99.9% (v/v) and nitrogen 99.9% (v/v) cylinders, provided by Shanghai Dahua, are applied to generate a simulated gas mixture. Three-stage gas filters, EVOLUTION-X, provided by Parker, are equipped to purify the feed gas mixture and ensure the safety of the follow-up equipment. The temperature of the inlet gas is controlled between 303 and 323 K by the heating belt and the temperature controller and remains consistent with that of the membrane module.
- **Membrane separation system:** Two kinds of CO₂-selected polyimide (PI) membrane modules (Table 1) are used in the membrane separation system. Pressure-regulating valves are adopted at the retentate and permeate sides to adjust the pressure. Needle valves are

Table 1. Parameters of Separation Membrane Modules

items	1# membrane	2# membrane
supplier	Shandong High-tech	Chengdu Asia
type	SE2050-P2-HO-BO	Generon 210
material	PI	PI
endurance temperature	93.3 °C	55 °C
weight	92 kg	1.81 kg
external diameter	8.23 cm	5.40 cm
length	181 cm	69 cm

Figure 2. Stability test of the membrane module.

Table 2. Packing Specifications

packing type	material	packing height	packing size	bulk density	specific surface area	porosity
rasching ring	glass	1500 mm	diameter 7 mm, length 22 mm	0.3 g/cm ³	198 m ⁻¹	0.757

Figure 3. CO₂ absorption performance of the packed column.

arranged on the stainless-steel pipes to control gas flow directions and transform the arrangements of different carbon removal methods.

Before the experiment begins, the separation performance and stability were carefully verified; part of the results can be found in Figure 2, which depicts that the membrane can quickly reach the performance balance and dynamic stability within 2–3 min and stably maintained during the subsequent operation.

- Chemical absorption system: The main body of the self-designed absorption column is manufactured by borosilicate glass, and two packing regions connected with clamps are axially random-packed with glass spring rings (Φ 6 mm $\times$ 14 mm). The length of the absorption column is 2300 mm long and the internal packing diameter is 100 mm; packing specifications are displayed in Table 2.

Five sampling holes are evenly arranged in every 300 mm at its axial distribution for detecting the gas component and an air insulating layer is covered outside the packing regions to lessen

the thermal loss; the absorption performance was verified and is shown in Figure 3. The total mass-transfer coefficient $K_G a_e$ of the gas phase volume is adopted to characterize the mass-transfer performance of the packed column. The total volume mass-transfer coefficient of the packed tower was measured by a differential method. In the packed tower with a counter-current contact, the height of the micro unit dz is taken as the material balance, and the specific calculation method is the same as that of Naami et al.¹⁸

- The desorption column is equipped with a heat-resistant fiberglass steam jacket, in which the moving of the high-temperature steam can reduce the temperature difference of the desorption column to ensure the operation safety. A reboiler provided by Wenzhou Dashun is applied to provide the driving force for the desorption process by introducing the superheated vapor into the desorption column. Magnetic-driven circulating pumps, MP-30R, provided by Shanghai Xinxishan are adopted to confirm the amine solution supply of the overall system. According to the change in the working

conditions, the amine flow s is controlled between 15 and 35 L/h, and the CO₂ loadings of lean and rich solutions are controlled around 0.2–0.3 mol CO₂/mol amine and 0.4 mol CO₂/mol amine, respectively.

- Two types of infrared gas analyzers, Gasboard-3100 (0–35 vol % CO₂, ± 1 FS) provided by Wuhan Sifang and XH-3010E1 (0–100 vol % CO₂, ± 2 FS) provided by Beijing Huayun, are adopted for measuring the composition of gas flow. During the working course, the filtered gas was flown into gas analyzers, the output signal of which was transported to a computer for online-monitoring.
- Exhaust gas evacuation system: A gas terminal box, 142526-188, provided by Anhui Guorui is installed at the end of gas pipes to collect and purify the purge gas. Functions, such as the stabilizing pressure, resisting fire, and filtering wastes, could guarantee the operation safety of the whole system. After being treated, the filtered gas was pumped out to the atmosphere by an air extractor, MTS-YIDA-01010 provided by Mantingshu.

3. PROCESS DESCRIPTION

For the sake of investigating the characteristics of various carbon capture methods for postcombustion, our work has been divided into four parts: a one-stage chemical absorption method (*Chem*), a one-stage membrane separation method (*Memb*), a two-stage membrane separation method (*Memb–Memb*), and a hybrid chemical absorption–membrane separation method (*Chem–Memb*), which are shown in Figure 4.

For one-stage methods, the main facilities are separately membrane modules and packed columns. During the *Memb* process (Figure 4a), the feed gas entered a hollow fiber membrane module, and the fast gas (rich in CO₂) and slow gas (rich in N₂) were, respectively, enriched at the permeate side and retentate sides, and the gas components were measured by infrared analyzers. In the *Chem* process (Figure 4b), the feed gas was delivered into the absorption column from the bottom entrance, and the lean solution was delivered into the column by a sprayer installed above the packing areas. After the gas–liquid phase reaction, the exhaust gas was purged out to the pipes, while the rich solution was pumped to the desorption column for the regeneration process.

With regard to two-stage methods such as *Memb–Memb* (Figure 4c), two hollow fiber membrane modules were serially arranged to form its configuration. The gas mixture was delivered into 1# module to get the initial-stage carbon removal, then the permeate gas was delivered into 2# module by the differential pressure, and the retentate gas was collected by pipes in order to heighten the gas recovery performance of the whole system. Meanwhile, the *Chem–Memb* (Figure 4d) configuration was formed by combining the membrane and chemical processes. The retentate gas from the membrane module was gathered to recover N₂, while the permeate gas was fed into the absorption column for further treatment and then the N₂-rich gas mixture was purged out from the top exit.

In general, this installation was established mainly to investigate the properties of one-/two-stage carbon capture methods containing membrane separation and chemical absorption processes for the postcombustion technology. The operating variables are listed in Table 3 including the parameters such as the gas flow rate, temperature, and so forth.

Figure 4. Process diagram of four kinds of carbon removal methods containing membrane separation and chemical absorption. (a) One-stage membrane separation method (*Memb*). (b) One-stage chemical absorption method (*Chem*). (c) Two-stage membrane separation method (*Memb–Memb*). (d) Hybrid membrane separation-chemical absorption method (*Memb–Chem*).

Table 3. Operating Variables Used in the Experimental Work

parameter	unit	value
feed gas flow	L/min	30–60
feed gas CO ₂ concentration	%	15
feed gas pressure	MPa	0.3–1.7
membrane module temperature	K	293–318
absorbent temperature	K	293–328
absorbent flow rate	L/min	15–35
steam generator output	K	428
superheated steam temperature	MPa	0.45

4. ANALYTICAL METHODS

In this study, the CO₂ removal efficiency γ (%) is adopted to estimate the system carbon removal performance (eq 1).¹⁹

$$\gamma = \frac{V_{\text{CO}_2, \text{in}} - V_{\text{CO}_2, \text{out}}}{V_{\text{CO}_2, \text{in}}} \times 100\% \quad (1)$$

where $V_{\text{CO}_2,\text{in}}$ and $V_{\text{CO}_2,\text{out}}$ are the inlet and outlet CO_2 volume flow rates, respectively.

The gas recovery efficiency ψ (%) refers to the ratio between the amounts of feed and recovered gases (eq 2).²⁰

$$\psi = \frac{V_{\text{rec}}}{V_{\text{feed}}} \times 100\% \quad (2)$$

where V_{feed} and V_{rec} are the inlet and recovery gas flow rates, respectively.

CO_2 regeneration efficiency λ (%) is defined to describe the regeneration performance of the aqueous amine solution in the desorption process (eq 3).

$$\lambda = \frac{n_{\text{out}}}{n_{\text{in}}} = \frac{Q_{\text{out}} \times \omega_{\text{out}}}{Q_{\text{in}} \times \omega_{\text{in}}} \times 100\% \quad (3)$$

where n_{in} , Q_{in} , and ω_{in} are the CO_2 amount, volume flow rate, and CO_2 concentration of the rich solution, respectively. The n_{out} , Q_{out} , and ω_{out} are the parameters for the lean solution.

During the chemical process, the lean solution converts to rich solution by absorbing CO_2 . The regeneration heat, consisting of sensible heat, vaporization heat, and desorption heat, can be calculated by the energy balance in the desorption column (eqs 4–7).^{21,22}

$$Q_{\text{re}} = Q_{\text{se}} + Q_{\text{va}} + Q_{\text{de}} \quad (4)$$

$$Q_{\text{se}} = q_m C_p (T_o - T_r) \quad (5)$$

$$Q_{\text{va}} = \kappa r c_{\text{CO}_2} \quad (6)$$

$$Q_{\text{de}} = c_{\text{CO}_2} \Delta q_r \quad (7)$$

where Q_{re} , Q_{se} , Q_{va} , and Q_{de} are, respectively, the regeneration heat, sensible heat, vaporization heat, and desorption heat. q_m is the mass flow of the rich liquid, kg/h; C_p is the specific heat of the absorption liquid, kJ/(kg • K); T_o and T_r are, respectively, the temperature of the desorption tower and rich liquid, K; c_{CO_2} is the CO_2 molar flow rate in the desorption process, mol/h; Δq is the heat of reaction between CO_2 and amine, kJ/mol; κ is the reflux ratio of the condensate in the desorption tower; and r is the heat of evaporation of the amine solution, kJ/mol.

5. RESULTS AND DISCUSSION

5.1. One-Stage Membrane Separation Method (Memb). Figure 5 shows the effect of gas temperature on the carbon removal performance of the Memb method.

It can be indicated in Figure 5a that CO_2 concentrations of 1# membrane declined from 55.3 to 42.6%, but the CO_2 removal efficiency increased from 58.1 to 84.8% as the temperature increased from 298 to 318 K. The kinetic diameter of CO_2 is 0.33 nm, which is less than 0.36 nm of N_2 ; while the narrower the size distribution of free volume holes in membranes, the greater the difference between the diffusion rates of CO_2 and N_2 , so the diffusion of CO_2 occurs prior to N_2 in membranes.

The promotion of CO_2 removal efficiency is because the rising of temperature can lead to a higher permeability but a lower solubility of the PI membrane, proving that the condensable gas CO_2 is more susceptible to temperature variation than the permanent gas N_2 .²³ Meanwhile, Figure 5b shows that the increase in the temperature can decrease the gas recovery efficiency from 89.4 to 78.3% but promote the N_2

Figure 5. Effect of the feed gas temperature on the comprehensive carbon removal performance of the Memb method. (a) Effect of feed gas temperature on CO_2 capture performance. (b) Effect of feed gas temperature on gas recovery performance.

concentration from 90.4 to 94.6%. This is because the temperature growth increases the amount of permeate N_2 , but weakens the gas recovery process, and the N_2 permeability growth rate is drastically lower than CO_2 for the CO_2 -selected membrane. Thus, the proportion of N_2 collected at the retentate side promotes the increase of the N_2 concentration. According to the solubility–diffusivity mechanism, the gas permeation behavior is determined by the diffusion effect, the temperature growth promotes the diffusion rate in the main body of the membrane, resulting in a promotion of the CO_2 permeate rate.

Figure 6 shows the effect of feed gas pressure on the separation performance of two kinds of membranes. It can be seen from Figure 6a,b that the CO_2 concentration increases but the gas recovery efficiency decreases as the pressure rises. Because CO_2 is a condensable gas subjected to the plasticizing swelling effect, the CO_2 permeability of the PI membrane increases with pressure enhancement. However, for the CO_2/N_2 gas mixture, the rising of the feed gas pressure can enlarge the competitive adsorption coupling effect between gas molecules.²⁴ The CO_2 solubility and intermolecular adsorption competition reach a relatively balanced state when the feed gas pressure remains at an overly high level, which is the reason that the CO_2 concentration of the permeate gas cannot show significant promotion when the pressure is above 0.7 MPa.

Figure 6. Effect of feed gas pressure on the separation performance of two kinds of membranes. (a) Effect of feed gas pressure on CO_2 concentration. (b) Effect of feed gas pressure on gas recovery. (b) Effect of feed gas pressure on gas recovery.

Furthermore, the promotion of the feed gas pressure also increases the amount of permeate N_2 , especially under low-pressure conditions. At the same time, the gas recovery efficiency is negatively influenced when the increase in the rate of N_2 exceeds CO_2 .

It can be concluded from the overall view that the experimental results from Figure 6b also prove that 1# membrane has a higher gas recovery efficiency and CO_2 concentration yielded from the permeate side, which will optimally do good to the overall CO_2 capture performance when the 1# membrane is applied in two-stage methods. This is because the higher CO_2 concentration of the feed gas in second-stage treatment can available promote the CO_2 purity of the final gas product. Whereas the higher primary gas recovery rate also accelerates the overall gas recovery performance.

5.2. One-Stage Chemical Absorption (Chem). Figure 7 shows the effect of absorption temperature on the overall separation performance of the Chem method. It can be seen from Figure 7a that the CO_2 removal efficiency and N_2

Figure 7. Effect of absorption temperature on the separation performance of the Chem method. (a) Effect of absorption temperature CO_2/N_2 separation performance. (b) Effect of absorption temperature on regeneration performance.

concentration increase as the absorption temperature rises from 298 to 318 K. This is because the absorption between MEA and CO_2 is a mass-transfer process accompanied by chemical reactions. The carbamate produced by the absorption reaction has high viscosity but is sensitive to the temperature vibration, thus resulting in the decline of liquid-phase mass-transfer resistance promoting the absorption process.

In addition, increasing the absorption temperature can profitably raise the desorption temperature, which intensifies the rich solution desorption process and depresses the CO_2 loading of the lean solution. However, when the temperature rises above 318 K, the increasing curves of CO_2 removal efficiency and N_2 concentration show reducing trends. This is because the absorption reaction is exothermic; the superlative heightening of the absorption temperature will improve the Henry coefficients of the absorption column, which refers to the fact that the CO_2 solubility of the amine solution declines under exorbitant temperature conditions.²⁵

Moreover, as shown in Figure 7b, the regeneration heat decreases but the regeneration efficiency increases as the absorption temperature rises. Because the growth of carbon removal efficiency provides supplemental heat and initial temperature, the increase of the rich-lean solution loading difference can lead to a higher regeneration efficiency,

contributing to the desorption temperature increase and regeneration heat reduction at the same time.

Figure 8 shows the effect of solution flow on the overall carbon separation performance in the *Chem* process. From

Figure 8. Effect of solution flow on the overall carbon separation performance in the *Chem* process. (a) Effect of solution flow rate on CO₂ capture performance. (b) Effect of solution flow rate on regeneration performance.

Figure 8a, it can be seen that the CO₂ removal efficiency and N₂ concentration, respectively, rise up to 99.81 and 99.78% when the solution flow rate increases from 20 to 30 L/h.

This is because the solution flow growth enlarges the amine content in the liquid phase body and reduces the gas–liquid ratio during the absorption process, thus driving more MEA molecules into the liquid film and enhancing the absorption reaction. Meanwhile, the heightening of the solution flow rate can also increase the specific surface area as well as the liquid-phase mass-transfer parameters. However, when the solution flow rate rises above 30 L/h, the carbon removal performances weaken due to the diminishing of the rich solution temperature, which can result in a higher lean solution loading and a lower absorption rate. As shown in Figure 8b, the regeneration efficiency increases but the regeneration heat decreases with the rise of the solution flow, which is due to the temperature drop in the desorption column. Because the carbamate decomposition is an endothermic reaction, the temperature lessening will prevent CO₂ desorption from the rich solution and decrease the regeneration efficiency.²⁶ The loading

difference between the rich and lean solutions decreases as the solution flow rate increases, aggravating the desorption sensible heat.

5.3. Two-Stage Membrane Separation Method (*Memb–Memb*). Figure 9 shows the effect of feed gas

Figure 9. Effect of feed gas pressure on the carbon removal performance of the *Memb–Memb* method. (a) Effect of feed gas pressure on gas components. (b) Effect of feed gas pressure on separation properties.

pressure on the carbon removal performance of the *Memb–Memb* method. It can be seen that the concentrations of produced CO₂ and recovered N₂, respectively, increase from 69.5 to 88.4 and 86.7 to 91.7% with the growth of the feed gas pressure (Figure 9a). The CO₂ removal efficiency increases and the gas recovery efficiency remains more than 98.5% (Figure 9b). This is because the additional second-stage membrane module can further separate the feed gas flow and heighten the CO₂ concentration of the permeate gas. Meanwhile, the improvement of the feed gas pressure between both membranes increases the recovery N₂ amount, this is why a dramatical promotion for carbon removal performance appears but no significant growth of gas recovery efficiency emerges in the *Memb–Memb* system.

Figure 10a,b illustrates the effect of feed gas flow rate on the carbon removal performance of the *Memb–Memb* method.

With the increase of the feed gas flow rate, the gas concentrations and recovery efficiency rise simultaneously, but the CO₂ removal efficiency descends from 80.1 to 52.4%. The results indicate that the improvement of the feed gas flow rate

Figure 10. Effect of the feed gas flow rate on the carbon removal performance of the Memb–Memb method. (a) Effect of feed gas flow on gas components. (b) Effect of feed gas flow on separation properties.

can effectively guarantee the gas recovery performance and permeate gas CO₂ concentration but decline the CO₂ removal capability of the Memb–Memb process. The reason is that the increase of the feed gas flow rate can simultaneously promote the mass-transfer performance of both PI membrane modules, which profitably enhances the CO₂ concentration of the permeate gas but inhibits more retentate gas to be recovered.²⁷ The acceleration of gas flow cuts down the contact time between CO₂ and the membrane, which is the primary cause of carbon removal efficiency reduction. Thus, according to effective demand, it is a requisite to choose a reasonable flow rate for keeping the balance between the treated gas amount and the carbon removal efficiency.

5.4. Hybrid Membrane Separation–Chemical Absorption (Memb–Chem). The effect of feed gas pressure on the overall carbon capture performance of the Memb–Chem method is shown in Figure 11. It can be found from Figure 11a that the carbon capture efficiency increases from 41.5 to 70.7% and the CO₂ permeate rate rises from 10.3 to 19.3 mol/h with the enhancement of the feed gas pressure. This is mainly because the growth of feed gas pressure significantly promotes the CO₂ permeation performance of the PI membrane, thus more CO₂ can be introduced into the absorption column and increases the carbon removal efficiency of the overall system.

Furthermore, Figure 11b shows that the N₂ concentration increases as the feed gas pressure grows. The reason is that the

Figure 11. Effect of feed gas pressure on the overall carbon capture performance of the Memb–Chem method. (a) Effect of feed gas pressure on CO₂ separation performance. (b) Effect of feed gas pressure on gas recovery performance. (c) Effect of feed gas pressure on the regeneration heat of Memb–Chem process.

promotion of feed gas pressure reduces the superfluous CO₂ component purged into pipelines from the retentate side. In addition, it can also be obtained from Figure 11b that the amount of N₂ recovered from the absorption column increases from 6.2 to 12.1 mol/h, but the total recovered amount N₂ remains steady. Because the incremental permeate gas into the absorption process is accompanied by surplus N₂, most of them can be collected from the exhaust outlet of the

absorption column, so that the gas recovery performance of the overall system can be promoted. Figure 11c presents the effect of feed gas pressure on the regeneration heat of the *Memb–Chem* process. It can be indicated that when the feed gas pressure increases up to 0.7 MPa, the regeneration heat rises from 2.50 MJ/kg CO₂ to 3.33 MJ/kg CO₂, but the regeneration heat remains stable as the feed gas pressure grows more than 0.7 MPa. The explanation can be given as follows: the increase of the feed gas pressure can heighten the permeability of the PI membrane and drive more CO₂ into the absorption apparatus, which raises the total regeneration heat. However, when the pressure rises to a high level, the gas flow enlargement will lead to a superfluous CO₂ concentration in the gas–liquid interface and impel more MEA molecules to diffuse into the liquid film to participate in the absorption reaction.²⁸ Thus, the solution loading difference increases, and CO₂ regeneration heat can be profitably reduced.

Figure 12 shows the effect of the solution flow rate on the CO₂ capture performance of the *Memb–Chem* method. It can

Figure 12. Effect of solution flow rate on the CO₂ capture performance of the *Memb–Chem* method.

be seen that the overall CO₂ capture efficiency of the *Memb–Chem* system increases from 68.2 to 74.7% with the growth of the amine flow rate. The reason is that the increase of the amine solution flow forces more MEA molecules to reach the liquid film and promotes the absorption reaction of both liquid and gas phases. In addition, it can also be found from Figure 12 that the promotion of amine flow rate enhances the regeneration heat from 2.05 MJ/kg CO₂ to 3.92 MJ/kg CO₂, which is chiefly due to the drop of the solution temperature and growth of the CO₂-loading difference.

5.5. Comparison between Four Kinds of Carbon Capture Methods. The comparison of CO₂ capture performance on four kinds of methods is shown in Figure 13 indicating that the CO₂ purity of the *Memb–Memb* method is 46.22% higher than the *Memb* method; such sharp improvement is mainly due to the pretreatment of the additional membrane separation stage. Meanwhile, the two-stage methods, including *Memb–Memb* and *Memb–Chem*, have a superior gas recovery efficiency of over 99.0%. This is because the second-stage process remaining in two-stage methods can retrieve an extra N₂ component existing in the permeate gas, and the promotion of gas recovery performance is realized by accessing the recovery gas with a high N₂ concentration to mix with the primary-stage retentate gas. In addition, the CO₂ removal

Figure 13. Comprehensive comparison of CO₂ capture performance on four kinds of methods.

efficiency of two-stage methods is slightly lower than the *Memb* method; this is because the recovery gas contains 0.5–8.6% CO₂, which cannot be captured by a packed column or membrane module. Especially, the lessening of the CO₂ capture performance significantly occurs when the feed gas pressure is maintained at a lower value.

Moreover, the data in Figure 13 also show that the *Chem* method has the upper carbon capture performance compared with other methods, which possesses both the CO₂ removal efficiency and CO₂ purity of more than 95%. While the N₂ purification and recovery properties also keep in excellent standards. However, the regeneration heat of amine desorption heightens the energy consumption of the overall system. As shown in Table 4, regeneration heat requirement of various solvents ranges from 2.51 MJ/kg CO₂ to 16.9 MJ/kg CO₂ under different operating conditions.

In this study, the optimal reboiler regeneration heat of the *Chem* method attains 2.70 MJ/kg CO₂, which is 34.33% higher than the *Memb–Chem* method. This proves that the additional membrane process of the *Memb–Chem* method can effectively reduce the peroration loading of absorption devices, especially at a low amine solution flow rate. Besides that, the *Memb–Chem* method can reduce the application amount of amine solution, which will decline the negative effect caused by amine solvents. Furthermore, the hybrid method provides a coupled approach to overcome the challenge in the carbon capture method by combining membrane separation and absorption processes to balance the differences in mass transfer in these two processes as well as reduce the overall rate of mass transfer.

6. CONCLUSIONS

Four kinds of carbon capture configurations containing chemical absorption and membrane separation were adopted to investigate the carbon capture performances of the CO₂/N₂

Table 4. Regeneration Heat Requirement of Various Solvents

reference	solvents	regeneration heat (MJ/kg CO ₂)
Asif ²⁹	10 wt % NH ₃	6.7
Asif ²⁹	30 wt % AMP	9.3
Asif ²⁹	30 wt % AMP and 3 wt % NH ₃	6.7
Warudkar ³⁰	20 wt % MEA	6.8
Razavi ³¹	40 wt % MEA	3.0
Brüder ³²	MEA	6.5
Warudkar ³⁰	40 wt % DEA	3.2
Warudkar ³⁰	60 wt % DGA	3.1
Zhang ³³	NH ₃	5.8
Jilvero ³⁴	5 wt % NH ₃	2.5
Darde ³⁵	7.8 wt % NH ₃	2.5
Darde ³⁵	6 wt % NH ₃	4.2
Artanto ³⁶	25 wt % AMP + 5 wt % PZ	4.4
Adeosun ³⁷	3 wt % DEA + 27 wt % AMP	3.7
Adeosun ³⁷	DEA/MDEA	4.3
Adeosun ³⁷	MEA/AMP	7.4
Kaiqi ³⁸	NH ₃	1.9
Jilvero ³⁴	NH ₃	2.5
Cousins ³⁹	MEA	3.3
Chem (this study)	20 wt % MEA	2.7
Memb–Chem (this study)	20 wt % MEA	2.0

systems in a self-designed lab-scale instigation. A comparison of critical parameters was proposed to evaluate the technical properties of different carbon capture methods. The combination of membrane separation and absorption processes by a common solvent was systematically realized to solve the problem originating from the difference in mass transfer. The major conclusions are drawn as follows.

- The growth of the feed gas temperature can promote the diffusion effect in the membrane module, resulting in the promotion of CO₂ permeate rate for the *Memb* method. While the competitive adsorption coupling and plasticizing swelling effects provide a bidirectional influence under variable pressures.
- There exists a reasonable MEA solution temperature of 318 K according to the performance variation of absorption and desorption processes for the *Chem* Method. Increasing the amine flow rate makes a positive impact on the mass-transfer performance in the absorption column but causes a decline of the desorption temperature.
- The CO₂ purity of the *Memb–Memb* method is 46.22% higher than the *Memb* method; such sharp improvement is mainly due to the pretreatment of the additional membrane separation stage. The increase of the feed gas flow rate can simultaneously promote the mass-transfer performance of membrane modules because of the enhancement of permeate CO₂ concentration.
- The *Memb–Chem* method provides an alternative way to reduce the solution flow and regeneration heat of the *Chem* method. The investigation on the *Memb–Chem* method provides an alternative approach to reduce the mass transfer and solve the problems caused by the unequal mass flow distribution.

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Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

This work was funded by the National Key Research and Development Program of China (2017YFB0603403).

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