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Analysing the Effect of CO_2 Concentration in Flue Gas on CO_2 Capture Characteristics of Hollow Fibre Gas Membrane System

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Membrane technology has been established as an efficient technology for capturing CO2 from multicomponent flue gas mixtures. The primary focus of this study is to examine the potential influence of CO_2 concentrations in the feed flue gas stream on the CO_2 capture characteristics of hollow fibre membrane module systems, whose fibre materials are made of polyetherimide-polyimide. The experiments were conducted on pure gases (CO_2, N_2) and multicomponent gas mixtures (N_2 + CO_2 + O_2), containing fixed amount of O_2 (4.3 %vol) and variable %vol of CO_2 and N_2 respectively, at a fixed volumetric flow rate of 130 Nlh^{-1}, at a temperature of 30 °C and a constant pressure difference of 8.57 bar. The objective was to determine and predict how flux, permeance, CO_2/N_2 selectivity/separation factor, stage cut through the membrane, retention coefficient as well recovery to the permeate stream vary with different CO_2 feed compositions. The results demonstrated that CO_2 flux, permeance and percentage recovery kept increasing as the proportion of CO_2 in the feed gas stream increases. CO_2 permeance 6.29 GPU and recovery 96 % peaked at a feed concentration of 55 %vol CO_2, with a minimum retention coefficient of 4 %. In contrast, the highest CO_2 separation factor 3.3 occurred at a feed concentration of 25 %vol CO_2.

1. Introduction

Rapid industrialization since the mid-20th century has driven global warming due to increased CO_2 emissions. According to Nedoma and Netušil (2021), the last few decades have witnessed a surge in topics related to CO_2 capture technologies in Europe and across the globe due to the high dependence on energy from fossil fuels. However, not only has CO_2 emission been on a steep rise but also a massive advancement in carbon capture and utilization technologies. Varieties of CO_2 capture technologies developed over the years are distinct, each with its pros and cons, however, energy utilization and consumption is a stand-out factor considered across all engineering fields (Habib et al., 2020). Membrane technology stands out among these methods due to its high energy efficiency, compact design, and scalability (Hu et al., 2022) and relatively lower cost (Adhikari et al., 2023). Zuberi et al. (2024) also suggested that CO_2 capture and utilization technologies will be substantial in achieving Vision 2050 of zero CO_2 emission.
Jasim et al. (2023) modeled the optimal operating conditions for the separation of CO_2 from CH_4/CO_2 mixture, at 2 to 10 mol% CO_2 feed concentrations, and observed that the mass transfer coefficient, flux and gas diffusivity through the membrane surface increase with increase in the CO_2 feed gas concentration. Xu et al. (2022) revealed that the purification of biogas improves with an increase in CO_2 concentration. Higher CO_2 feed concentrations enhance the efficiency of CO_2 capture by creating a larger concentration gradient. Similarly, Yan et al. (2022) found that during the upgrading of biogas, certain solvents perform better when the concentration of CO_2 in the biogas mixture is high. According to Michailos and Gibbins (2022), the efficiency of carbon capture from flue gases largely depends on the concentration of CO_2 in the flue gas feed. Wang and Song (2020) also noted that a higher percentage of CO_2 by volume in the feed flue gas positively impacts the efficiency of carbon capture technology being used. Aydani et al. (2020) explained that the effectiveness of capturing CO_2 from

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binary mixtures (such as CO_2/CH_4 and CO_2/N_2) increases with a higher concentration of CO_2 in the feed gas, as the increased CO_2 permeance implies the depletion of other gas components in the mixture. Fernández-Barquín et al. (2017) studied CO_2/N_2 separation using a two-stage membrane process with feed CO_2 concentrations of 5% to 65%. Their findings showed that membrane performance improves with higher concentrations of the target gas. Higher CO_2 concentrations in the feed gas enhance the efficiency of carbon capture technologies, reducing energy demands and solvent use in absorption processes. However, proper operational measures are essential to tackle challenges such as corrosion and system degradation (Xu et al., 2022). A review of the literature highlights a lack of studies on the impact of CO_2 concentrations variations on carbon capture technologies, particularly in membrane systems. Most research has concentrated on binary gas separations, such as CH_4/CO_2 or CO_2/N_2, using absorption or adsorption methods. This study aims to address this gap by experimentally analyzing how different CO_2 concentrations impact the efficiency and mass transfer characteristics of a hollow fiber membrane system, under fixed temperature and pressure conditions, with a particular emphasis on flue gas systems.

2. Methods

2.1. Experimental set-up

The experiments were conducted to evaluate the impact of varying CO_2 concentrations in flue gas streams on the capture of CO_2. This was achieved using a laboratory membrane unit, the Ralex GSU-LAB 200, manufactured by MemBrain. The full characteristics of the membrane system are detailed in Table 1, while Figure 1 provides a schematic representation of the process. Initially, tests were performed using pure gases (100 %vol CO_2 and N_2) at the same volumetric flow rate of 130 Nlh^{-1}, with variable feed-side pressure ranges and a constant permeate pressure of 1.43 bar. Following this, six different model flue gas mixtures with varying compositions, as outlined in Table 2, were passed through the membrane gas analyzer/separator under steady-state conditions. The complete experimental configuration and analysis were carried out at a constant volumetric flow rate of 130 Nlh^{-1}, at an average process temperature of 30 °C, with feed and permeate pressures maintained at 10 bar and 1.43 bar, respectively.

Table 1: Hollow fibre membrane module (P2-1.2) system properties

Fibre material	Polyetherimide
Quantity of fibre	2900 pieces
external fibre diameter	0.3 mm
Active length of fibre	0.3 m
Fibre wall thickness	0.00006 m
Mass transfer surface area	0.82 m^2

Table 2: The compositions of the model feed gas mixtures used in this study

Model gas mixture %composition ($N_2 + CO_2 + O_2$)	N_2 (%)	CO_2 (%)	O_2 (%)
90.7-5-4.3	90.7	5.0	4.3
80.7-15-4.3	80.7	15.0	4.3
70.7-25-4.3	70.7	25.0	4.3
60.7-35-4.3	60.7	35.0	4.3
50.7-45-4.3	50.7	45.0	4.3
40.7-55-4.3	40.7	55.0	4.3

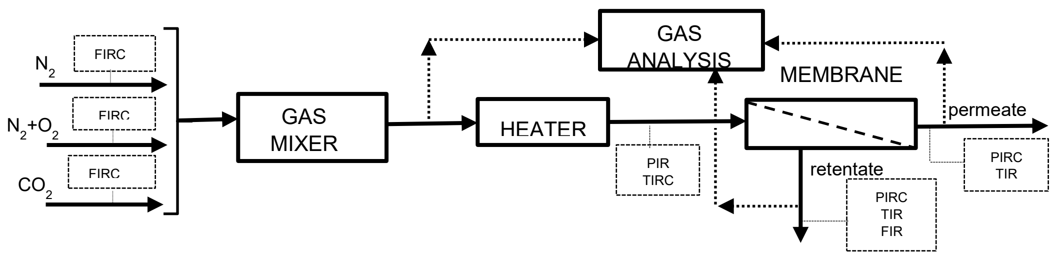

Figure 1: Block diagram illustrating CO_2 membrane separation process from the flue gas stream

2.2. Data analysis

Experimental data were analyzed to calculate key gas flow properties, including permeate flux (molm^{-2}s^{-1}), Permeability P_i (barrer), Permeance ($\frac{P_i}{l}$) (GPU), Selectivity α_i, Stage cut Θ using standard equations from literature (Kratky et al., 2020). The calculations assumed ideal gas behavior, incompressible, laminar, Newtonian flow under steady-state conditions. Parameters such as separation factors (Sf*), CO_2 recovery in the permeate stream R and CO_2 retention coefficient Rc are also determined using the equations below, to ascertain their interactions with different CO_2 feed concentrations in the multicomponent gas mixtures.

$$\mathrm{R} = \frac{n_i^p}{n_i^f} \cdot 100\% \tag{1}$$

$$\mathrm{Rc} = \frac{n_i^f - n_i^p}{n_i^f} \tag{2}$$

$$\mathrm{Sf^*} = \frac{c_i^p}{c_j^p} \Big/ \frac{c_i^f}{c_j^f} = \frac{n_i^p}{n_j^p} \Big/ \frac{n_i^f}{n_j^f} \tag{3}$$

where *n* (mols^{-1}) represents gas concentrations, superscripts p and f refer to permeate and feed sides respectively while the subscripts i and j represents CO_2 and any other gas respectively.

3. Results and Discussion

3.1 Pure gases

Figures 2a and 2b illustrate the relationship between the permeance of individual pure gases through the hollow fiber membrane and partial pressure differences at a constant temperature under steady-state conditions. The membrane exhibited high CO_2 permeance, emphasizing its potential for efficient and cost-effective CO_2 capture. CO_2 permeates easily through the membrane, while N_2 remains largely in the retentate stream, highlighting the module's selectivity for CO_2 over N_2.

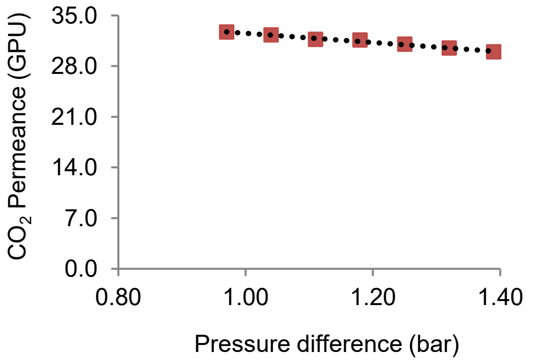

Figure 2a: CO_2 permeance against partial pressure difference

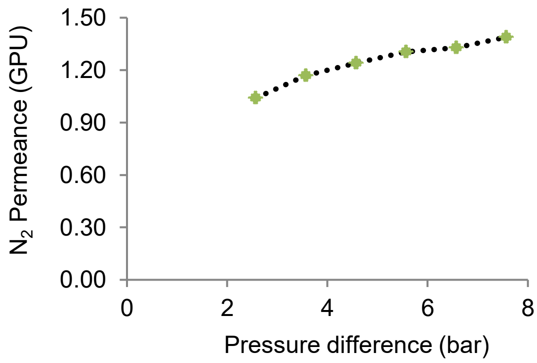

Figure 2b: N_2 permeance against partial pressure difference

The data also revealed that CO_2 permeance decreases as the partial pressure difference increases, while N_2 permeance shows a slight increase. This behavior differs from the typical trend of permeate flux, which generally rises with increasing partial pressure difference. The inverse relationship between CO_2 permeance and partial pressure difference arises because permeance depends directly on permeate flux but inversely on the pressure difference. The decrease in gas flux efficiency at high pressures can be attributed to plasticization, as condensable gases like CO_2 can induce this effect in polyimide membranes. Plasticization causes swelling in the polymer matrix, altering free volume and inter-chain spacing. This increases polymer segment mobility, reducing the size-sieving ability of polyimide membranes and ultimately decreasing selectivity (Zhang et al., 2019). Similar observations were made by Thundyil et al. (1999), who noted a decline in pure CO_2 permeability with rising pressure differentials, and by Jasim et al. (2023), who reported reduced CO_2 diffusivity (a property linked to permeability) as pressure differences increased.

3.2 Effect of different CO_2 concentrations on the CO_2 capture from the model flue gas

This section delves into the effect of increasing CO_2 concentrations (% by volume) in the model gas on the mechanisms of mass transfer involving CO_2 capture from flue gas stream.

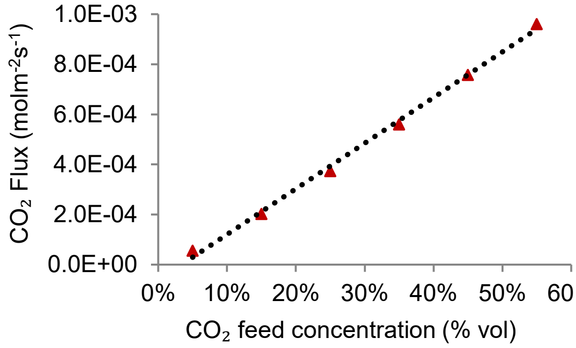

Figure 3a: Effect of different CO_2 concentrations on CO_2 permeate flux

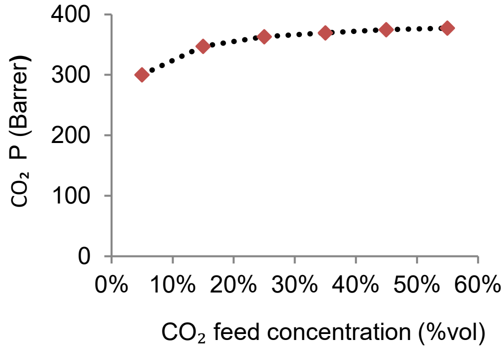

Figure 3b: Effect of different CO_2 concentrations on CO_2 permeability

Figures 3a and 3b illustrate the impact of increasing CO_2 concentrations on CO_2 flux and permeability in the hollow fiber membrane system. The data show a direct relationship between CO_2 concentration, permeate flux and CO_2 permeability. Permeate flux exhibits a sharp, continuous increase with rising CO_2 concentrations across all levels. However, CO_2 permeability shows the most significant increase between 5 %vol and 25 %vol CO_2 concentration, with the rate of increase slowing beyond this range. Figure 3b highlights the steepest rise in permeability between 5 %vol and 15 %vol, followed by a slower increase from 15 %vol to 25 %vol, and a more pronounced decline in the rate of increase above 25 %vol. This trend suggests that very high CO_2 concentrations may negatively affect CO_2 selectivity and separation factor from the gas mixture. Figure 3a illustrates that permeate flux increases directly with an increase in CO_2 concentrations because higher CO_2 concentrations result in higher concentration or partial pressure difference (driving force) across the two sides of the membrane (Jasim et al., 2023). Xu et al. (2022) and Jasim et al. (2023) reported a similar trend in the permeate flux with CO_2 concentrations.Figures 4a and 4b illustrate the effects of feed CO_2 concentration on CO_2 recovery, retention coefficient, stage cut, and selectivity in a membrane system. CO_2 recovery refers to the proportion of CO_2 in the feed gas that reaches the permeate stream, while the retention coefficient measures the amount of CO_2 retained in the retentate stream. The data show that as feed CO_2 concentration increases, CO_2 recovery rises due to the membrane's high CO_2 permeability, while the retention coefficient declines. This occurs because higher CO_2 concentrations drive more CO_2 into the permeate stream, leaving the retentate with reduced CO_2 content, indicating higher purity in the permeate stream. These findings align with Fernández-Barquín et al. (2017), who linked higher CO_2 recovery to the increased driving force at elevated feed CO_2 levels. Figure 4b highlights that stage cut (fraction of feed gas passing through the membrane) increases with rising feed CO_2 concentrations. However, CO_2 selectivity peaked at 15 %vol feed CO_2 concentration and then declined as feed concentration increased further, suggesting that excessively high CO_2 levels may negatively impact selectivity. This trend, consistent with Aydani et al. (2020), implies a trade-off between stage cut and selectivity, as selectivity decreases with increasing stage cut. This observation also aligns with Seghman et al. (2022).

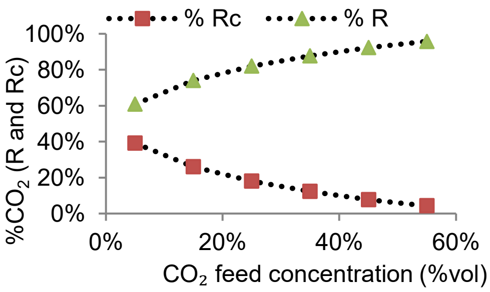

Figure 4a: Effect of CO_2 feed concentrations on CO_2 recovery (R) and retention coefficient (Rc)

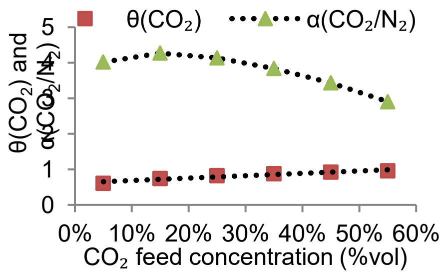

Figure 4b: Effect of CO_2 feed concentrations on Stage cut $\theta(CO_2)$and selectivity $\alpha(CO_2/N_2)$

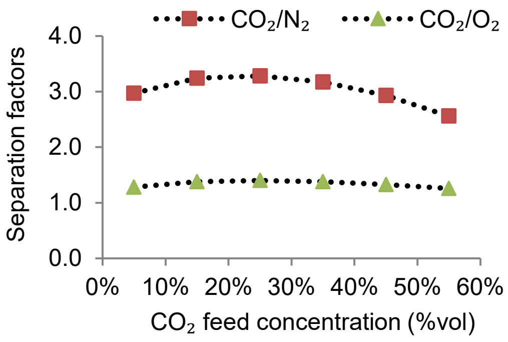

Figure 5a: Effect of different CO2 concentrations on separation factors

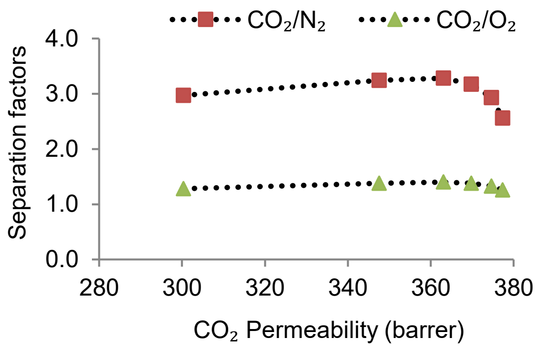

Figure 5b: Relationship between separation factor and permeability at different CO2 concentrations.

Figures 5a and 5b delineate the correlation between CO_2 separation factors about the other two gases (N_2 and O_2) and CO_2 concentrations and permeability. The results indicated that the separation factor Sf*(CO_2/N_2) is far greater than the separation factor Sf*(CO_2/O_2) across all CO_2 concentrations, emphasizing the membrane's superior ability to separate CO_2 from N_2 due to the substantial difference in their diffusivities. This affirms the efficacy of the membrane module used in this study for the separation of CO_2 and N_2 Mixtures. A further perusal through Figure 5a also revealed maximum separation factor Sf*(CO_2/N_2) of 3.3 was recorded at 25 %vol feed CO_2 concentration; further increase in CO_2 concentrations results in a decrease in the separation factors. Figure 5b easily portrayed that the increase in CO_2 separation factors CO_2 with permeability was short-lived as maximum separation factor Sf*(CO_2/N_2) was obtained at CO_2 permeability of 363 barrer, after which a downtrend sets in. This agrees with our earlier assumption that very high CO_2 concentrations in the feed gas may be detrimental to CO_2 selectivity and separation factor. This pattern aligns with the understanding that membranes with high permeability often exhibit reduced selectivity, a trend supported by Jong Hak Kim (2022). Kratky et al. (2021) observed that the separation factor decreases as CO_2 permeability and feed flux increases.

Table 3: The overview of the mass transfer properties of CO_2 in the model gas mixtures used

cCO_2F	P_{CO_2} (GPU)	% R	% Rc	θ(CO_2)	α(CO_2/N_2)	Sf*(CO_2/N_2)	Sf*(CO_2/O_2)
5%	5.01	61%	39%	0.61	4.02	2.97	1.28
15%	5.79	74%	26%	0.74	4.27	3.24	1.38
25%	6.05	82%	18%	0.82	4.14	3.30	1.40
35%	6.16	88%	12%	0.88	3.84	3.17	1.38
45%	6.24	92%	8%	0.92	3.43	2.93	1.33
55%	6.29	96%	4%	0.96	2.90	2.56	1.26
100%	30.0						

3. Conclusions

This study highlights the effectiveness of the selected hollow fiber membrane for separating CO_2/N_2 mixtures, with CO_2 predominantly diffusing to the permeate stream and N_2 remaining in the retentate. CO_2 concentrations in flue gases typically range from 8–15 %vol but can exceed 80 %vol in oxy-fuel combustion due to the absence of nitrogen. Higher CO_2 concentrations in the feed gas enhance the separation process by increasing the driving force, resulting in improved recovery rates and membrane permeability. This also lowers operational costs and energy requirements as nitrogen composition in the feed gas decreases. The study observed that CO_2 flux, stage cut, and recovery increased with higher CO_2 concentrations, while the retention coefficient declined due to elevated partial pressure differences. The peak of the separation factors Sf*(CO_2/N_2) and Sf*(CO_2/O_2) are 3.3 and 1.40 respectively at 25 %vol CO_2 feed concentration, indicating optimal selectivity at moderate concentrations. However, at very high CO_2 feed concentrations, plasticization effects could occur, thereby reducing the membrane's selectivity despite an overall increase in permeability. These findings demonstrate that the improved performance of the membrane at high CO_2 %vol feed concentrations is primarily due to enhanced permeability, though selectivity may be compromised at extreme levels of CO_2 in the multicomponent feed gas.

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