Refereed Proceedings

Separations Technology VI: New Perspectives on Very Large-Scale Operations

Engineering Conferences International *Year* 2004

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ECONOMIC EVALUATION OF MEMBRANE SYSTEMS FOR LARGE SCALE CAPTURE AND STORAGE OF CO_2 MIXTURES

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ABSTRACT

The capture and storage of CO_2 (CCS) as a greenhouse mitigation option is becoming an increasingly important priority for Australian industry. Membrane based CO_2 removal systems can provide a cost effective, low maintenance approach for removing CO_2 from gas streams. This study examines the effect of membrane characteristics and operating parameters on CCS costs using economic models developed by UNSW for any source-sink combination. The total sequestration cost per tonne of CO_2 avoided for separation, transport and storage are compared for the separation of CO_2 from coal fired power plants and natural gas processing. A cost benefit analysis indicates that sequestration of gases of high purities are dominated by compression costs which can be off-set by utilising membranes of higher selectivity coupled with higher permeability to reduce the required transmembrane pressure.

INTRODUCTION

CO_2 capture and storage (CCS) is an important short-term lever for addressing climate change. Over the course of the century, CCS could account for 30% or more of all climate mitigation beyond "business as usual" technology improvements (1).To meet reduction targets, CCS technologies are likely to be deployed on a large scale around the globe.

Early studies of CO_2 capture with membranes indicated that the cost was 30% higher in cost than amine chemical absorption (1, 2). The limitations of the studied membranes was due to the high cost of compressing low pressure flue gas and the low purity of permeate which results in the need for multi-stage processing to achieve the most economic arrangement for CCS. The objective of this study is to examine the effect of the membrane characteristics of permeability and selectivity and operating parameters such as trans-membrane pressure on the total cost of CCS.

METHOD

The economic analysis for this paper examines the cost of CO_2 capture and storage of mixtures for both a 500 MW coal-fired power plant and a 35 MMSCFD natural gas processing facility. The analysis for the coal-fired power plant assumes that the flue gas is from a typical 500 MW Australian pulverised black-coal power plant. The analysis for the natural processing facility assumes a typical offshore Australian facility, with processing conditions that meet the Australian pipeline specifications (3). It is assumed that the feed gas for both processes is dehydrated and all contaminant gases such as NO_x, SO_x and H_2S have been removed. The conditions for the geological reservoir are the same as those in Ho et al (4). A summary of the input parameters is given in Table 1. The specifications of the two polymeric membranes considered in this study are shown in Table 2. For convenience, the power plant flue gas is referred to as the CO_2/N_2 system and the natural gas facility is referred to as the CO_2/CH_4 system.

In this study, the effect of removal efficiencies and the purity of product using gas separation membranes for CO_2 recovery and hence the subsequent effect on storage of a mixed gas product is evaluated. Both 1-stage and 2-stage membrane systems were investigated. To simulate a 2-stage system, the output from the first stage is taken as the input for the second stage simulation. The membrane was modelled using a modified cross-flow model with no recycle stream as described by Shindo et al (5). This cross-flow model enables a multi-component gas mixture to be examined and provides a reasonable first approximation of real systems which have many membranes modules operating in series and parallel in each stage.

One of the consequences of using gas separation membranes is that the permeate or retentate streams contains other component gases as well as the desired gas because membranes are not perfect separators. To increase the concentration of CO_2 in the gas stream to a level which is economically viable for storage, the permeate stream from the first membrane, which is enriched in CO_2, is recompressed and passed through a second membrane (6). This configuration is referred to as a two-stage cascade membrane system (TCSM) as shown in Figure 1.

The second 2-stage membrane system examined in this study was the two-stage series membrane system (TSMS), as shown in Figure 2. In this process set-up, the retentate stream from the first membrane is passed through a second membrane, removing further CO_2 (7). This configuration was used for the recovery of CO_2 from natural gas streams to achieve the desired pipeline specifications of less than 2% CO_2.

In this study, it was assumed that the power requirement needed for the CO_2 separation process and compression stages is provided from a supplementary power supply. A standard assumption made purely for the purposes of this study is that the supplementary energy will come from a new natural gas combined cycle power plant ('NGCC'). The CO_2 emission from the NGCC power plant is assumed to be 0.4 kg CO2 per kWh15 (4). Due to the lower concentrations of CO_2 in NGCC flue gases, we assume that such CO_2 emissions are vented to the atmosphere and not captured. Therefore, they contribute to the total CO2 emissions of the system. The net tonnes of CO_2 avoided is the difference between the tonnes of CO2 stored and the tonnes of CO_2 emitted after capture. The percent CO_2 avoided is calculated as:

$$\%\ CO_2\ \text{avoided} = \frac{CO_2\text{captured - }CO_2\text{emitted from supplementary power}}{CO_2\text{original emission from source}} \qquad (1)$$

In assessing the economic feasibility of membranes for the purposes of CCS, the economic indicator of \$/tonne CO_2 avoided or cost of CO_2 avoidance is widely used. The \$/tonne CO_2 avoided describes the total capital and operational investment needed for the purposes of CCS and is calculated as:

$$\text{Cost of } CO_2 \text{avoided} = \frac{\sum_{i=1}^{n} \frac{K_i + O_i}{(1+d)^i}}{\sum_{i=1}^{n} \frac{(CO_2 \text{ avoided})_i}{(1+d)^i}} \qquad (2)$$

where K_i and O_i are the real capital and operating costs (US$ million) in the *i*th year of operation, *d* is the discount rate (% pa) and CO_2 avoided is the annual amount of CO_2 avoided in million tonnes. The economic assumptions used in this study are listed in Table 3.

RESULTS AND DISCUSSION

Relative cost breakdown

For low pressure feed gas mixtures such as the flue gas from a power plant, results in Figure 3 shows that the largest contributor to capital costs for the membrane system is the cost of the compressor required to elevate the pressure of the inlet feed gas to a suitable level before separation, coupled with the cost of the expanders needed to de-pressurise the retentate waste gas stream. The compressor and expanders account for approximately 80% of the total investment, while the membrane and membrane housing account for less than 15% for both a 1-stage and 2-stage membrane system (8).

For the natural gas facility, the largest cost item is for the membrane and associated housing, which accounts for 62% to 85% of the total capital cost as shown in Figure 4. This is because there is no high compression cost associated with the natural gas feed, which is already available at a high pressure. For a 1-stage membrane system there is no compression cost, however for a 2-stage membrane system such as a TCMS configuration an intermediate compressor is used. From the cost analysis, the cost of the compressor required to recompress the CO_2 enriched stream accounts for approximately 30% of the total cost. This is significantly less than for the low-pressure flue gas system.

The effect of permeability and thickness

The CO_2 permeability will influence the rate at which CO_2 is removed from the feed gas by the membrane. Higher CO_2 permeabilities will reduce the size of the membrane needed for separation. Figure 5 shows that increasing the CO_2 permeability by factors of 10 from 1 Barrer to 1000 Barrer results in significant reductions in the cost of capture for both the power plant flue gas and natural gas processing feed gas. This cost reduction is most noticeable between CO_2 permeabilities of 1 and 100. This reduction occurs due to the reduced area of membrane needed for CO_2 separation. Figure 5 also shows the effect on cost by changing the permeance; the ratio of the permeability divided by the membrane thickness. For the same CO_2 permeability, if the membrane thickness is halved from 0.1 □m to 0.05 □m, the cost of capture for both CO_2 feed gases also reduces. This reduction is most noticeable between permeabilities of 1 and 100 and is also a result of the reduced area of membrane needed. However, as the permeability increases to values greater than 100, the economic gain in reducing the membrane area is not as significant. This is due to the small cost contribution of membranes to the overall capital investment as shown in Figures 3 for the power plant flue gas system, and the low membrane cost assumed in Table 3.

The capture cost for the natural gas feed gas is substantially less than for the power plant flue gas at the same CO_2 permeability values. This is because the natural gas feed is already provided at a high pressure eliminating the need for a feed gas compressor. The economic analysis assumes that compressors are more expensive than membranes. The capital expenditure for the natural gas processing facility is therefore less than for the power plant flue gas and hence the capture cost is also less.

The effect of selectivity

The CO_2 selectivity of a membrane represents the ratio of the CO_2 permeability over the permeability of other component gases in the mixture. Figure 6 shows the changes that occur in the concentration of CO_2 in the permeate stream as a function of the CO_2 selectivity for both the power plant flue gas and natural gas streams. Figure 7 shows the effect on the capture cost for the two CO_2 feed gases with changes to the CO_2 selectivity from a value of 20 to 100. As shown in Table 2, the CO_2/N_2 selectivity of the PPO membrane used for the flue gas separation is 20 (1), and the CO_2/CH_4 selectivity of the cellulose acetate membrane considered for natural gas processing is 20 (7). For the power plant flue gas, the CO_2 recovery rate is fixed at 70%, while for the natural gas processing stream the level of CO_2 recovery is varied to achieve pipeline specifications.

From the results, for the flue gas stream, increasing the CO_2 selectivity from 20 to 50 results in an increase in permeate CO_2 concentration from approximately 50% to 65%. Further increases in selectivity from 100 and 200 can achieve CO_2 concentrations of 75% or 80% respectively, as shown in Figure 6. Meanwhile, the results in Figure 7 show that the CO_2 storage cost reduces with the improved selectivity due to the increased CO_2 purity in the permeate stream. It has been shown by Allinson et al (4) that it is more cost effective and requires less power to compress high CO_2 purity streams than streams with low CO_2 content. However, improving the CO_2 selectivity does not have significant impact on the capture cost of the flue gas stream. The capture cost actually increases slightly with improved CO_2 selectivity. This occurs because for this analysis, the amount of CO_2 removed and CO_2 permeability are fixed. According to Fick's law as shown in Equation 3, by increasing the CO_2 selectivity, the mole fraction of CO_2 in the permeate increases and the mole fraction of CO_2 retentate is reduced. Consequently the driving force across the membrane is also reduced and to obtain the same effective CO_2 recovery; that is the same number of moles of CO_2 removed, the membrane area increases. This increase in membrane area increases the total capital cost.

$$n_{CO_2} = \frac{\overline{Q}_{CO_2}}{t} \times A_{membrane}\left(P_{feed}x_{CO_2} - P_{permeate}y_{CO_2}\right) \qquad (2)$$

where n_{CO2} is the number of moles of CO_2 removed, $A_{membrane}$ is the membrane area (m2), Q is the CO_2 permeability, t is the thickness, P_{feed} and $P_{permeate}$ is the pressure in the feed and permeate streams respectively, and x_{CO2} and y_{CO2} is the mole fraction of CO_2 in the retentate and permeate respectively.

For the natural gas feed, improving the CO_2 selectivity from 20 to 60 results in an increase in

CO_2 permeate concentration from approximately 60% to 77% as shown in Figure 6. The methane gas in the permeate stream is considered as lost product. Therefore any improvements in CO_2 selectivity for a natural gas processing facility have the twin benefits of reducing the required membrane area resulting in less capital expenditure, as well as reducing the quantity of methane product lost. Figure 8 shows the changes in capture cost and quantity of methane product loss per annum at two CO_2 selectivity values. For a feed gas with a 10% mole fraction of CO_2, improving the CO_2 selectivity from 20 to 60 almost halves the quantity of methane product that is lost. Improving the selectivity results in better production rates at all CO_2 feed concentrations, from 10% to 50%. Improving the selectivity also decreases the capture cost even though the area of the membrane required increases. This is because the gains from the reduced methane loss compensates for the increase in capital cost. Improving the CO_2 selectivity for the natural gas feed will also reduce the storage cost due to the increase in CO_2 purity in the permeate stream (Figure 8). The storage cost for the natural gas facility is also less than the storage cost for the power plant flue gas because the facility is already located offshore. This has the benefit of reduced capital expenditure due to fewer infrastructure cost items such as pipelines.

Combined effects of permeability and selectivity

The results above indicate that improving the CO_2 permeability will reduce the capture cost, while improving the CO_2 selectivity will reduce the storage cost but may increase the corresponding capture cost for both CO_2 feed gases.

The cost analysis in Figure 9 examines the combined effect of improving both the CO_2 permeability and selectivity for CO_2/N_2 separation. This analysis considers doubling the permeability and selectivity of the PPO membrane listed in Table 2. By doubling the permeability from 75 Barrer to 150 Barrer, and doubling the CO_2/N_2 selectivity from 20 to 40, the storage cost reduces compared to the base case but the capture cost remains unchanged. The storage cost reduces due to the higher concentration of CO_2 in the permeate stream by utilising the higher CO_2 selectivity. However the capture cost remains unchanged because even though the improved permeability reduces the required membrane area, the improved selectivity increases the required membrane area. Thus there is no net change in the membrane area. Improving the permeability and selectivity in isolation only generates total CCS savings of 1-2%.

The effect of pressure ratio

The pressure ratio is the ratio of the permeate pressure to the feed pressure for the gas separation membrane stage. Keeping all other membrane characteristics constant, a high-pressure ratio will result in a lower membrane area for a fixed flux across the membrane. From the cost analysis shown in Figure 4, the biggest contributing factor in the both the capital and operating cost is the energy required to compress the inlet feed gas to a suitable pressure. The analysis in the previous sections show that improvements in the membrane properties can provide cost benefits, however if the feed pressure remains unchanged, improving the permeability and selectivity of the membranes considered in Table 2 only generate savings of less than 10%. By utilising advantages of improved selectivity and permeability in the relation to pressure ratio, the cost of mitigation may considerably be reduced. Figure 10 shows the capture, storage and total CCS cost for CO_2 recovery from power plant flue gas for two scenarios. The first considers CO_2 recovered at various recovery rates of 60 to 90% or CO_2 avoided rates of 35% to 55% using the PPO membrane characteristics listed in Table 2. The second case examines a membrane system at the same CO_2 recovery rates but the permeability and selectivity values have been improved. The analysis has been set so that the mole fraction of CO_2 in the permeate stream is the similar to that as the first case, but the feed pressure has been reduced. This is achieved because at each CO_2 recovery rate, the amount of CO_2 is fixed and according to Fick's Law as shown in Equation 3, increasing the CO_2 permeability while maintaining similar membrane area results in a reduction in the driving force across the membrane. Due to the improved selectivity, the CO_2 mole fraction in the permeate stream increases and thus, the feed pressure can be reduced. The storage cost is the same for both scenarios because the concentration and flowrate of the CO_2 in the permeate stream is the same. However, the capture cost and hence total CCS cost for the membrane system with the improved selectivity, permeability and pressure ratio is much lower than the standard case. This is because a lower feed pressure requires a smaller compressor, reducing both the capital and operational costs. By being able to operate at a lower trans-membrane pressure when membrane selectivity and permeability are improved, the CO_2 capture and total CCS costs may be reduced up to 15%.

CONCLUSION

The analyses shows that considerable CO_2 removal rates can be achieved with gas membrane separation systems, and the economic competitiveness of the systems in comparison to amine based systems depends on both the membrane characteristics and characteristics of the feed gas. For low-pressure systems such as power plant flue gas, utilisation of reduced trans-membrane pressure through improvements to the selectivity and permeability can improve the capture, storage and total CCS cost. Improvements in permeability and selectivity for membrane systems recovering CO_2 from natural gas processing can result in substantial cost benefits through reduced capital expenditure and reduction in methane losses.

ACKNOWLEDGEMENTS

Australian Cooperative Research Centre for Greenhouse Gas Technologies (CO2CRC) provided financial support for this research project.

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KEY WORDS

Greenhouse mitigation, carbon capture, geosequestration, CCS economics, membrane systems, CO_2 capture

Table 1 Summary of the input parameters for CCS studies

Feed gas condition		500 MW Power Plant Flue gas (CO_2/N_2)	Natural Gas Processing Feed gas (CO_2/CH_4)
Flowrate	m^3/s	500	11.5
Composition	% mol	CO_2 14	CO_2 10
		N_2 80	CH_4 90
		O_2 6	-
Temperature	^oC	93	45
Pressure	bar	1	55

Table 2 Properties of the polymeric membranes considered in this study

Membrane properties	500 MW Power Plant Flue gas (CO_2/N_2)	Natural Gas Processing Feed gas (CO_2/CH_4)
Membrane material	Polyphenyleneoxide (PPO) (1)	Cellulose Acetate (7)
CO_2 permeability (Barrer)	75	6
Selectivity	CO_2/N_2 20	CO_2/CH_4 20
	CO_2/O_2 4	-
Membrane temperature (^oC)	30	30
Membrane feed pressure (bar)	20	55

Table 3 Summary of the economic parameters used for CCS studies.

Discount rate	7 % pa
Cost of external power	20 $/MWh
Fixed annual operating cost	4% of total Capital Costs
Project life	20 years
Construction period	2 years
Membrane cost	80 $/m^2
Production cost for Natural Gas Methane	1.5 $/GJ

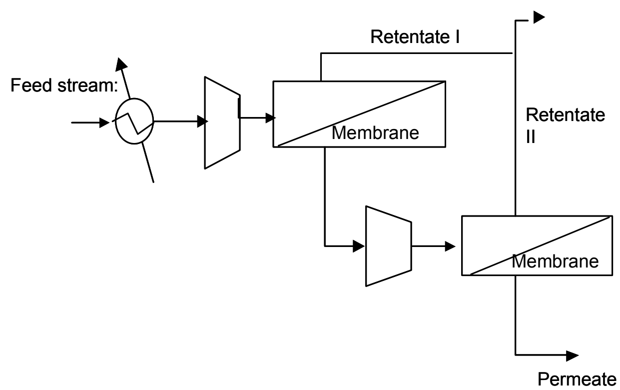

Figure 1 A two-stage cascade membrane system configuration (TCMS)

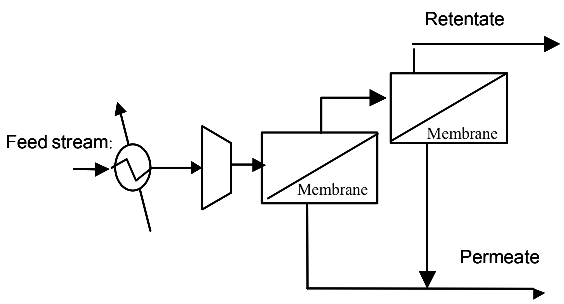

Figure 2 A two-stage series membrane system configuration (TSMS)

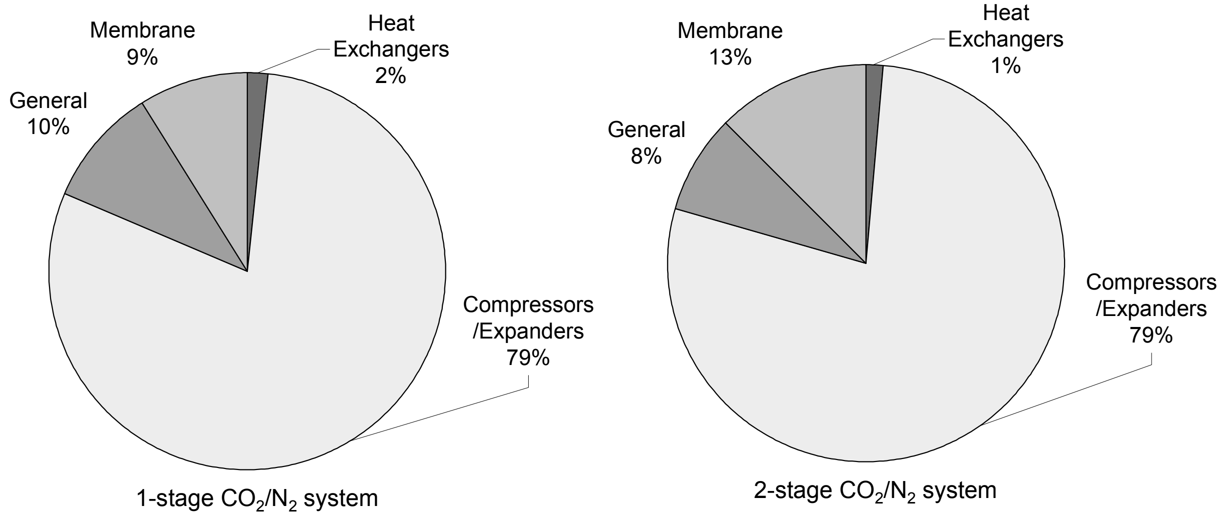

Figure 3 Relative capital expenditure cost breakdown for both a 1-stage and 2-stage CO_2/N_2 system

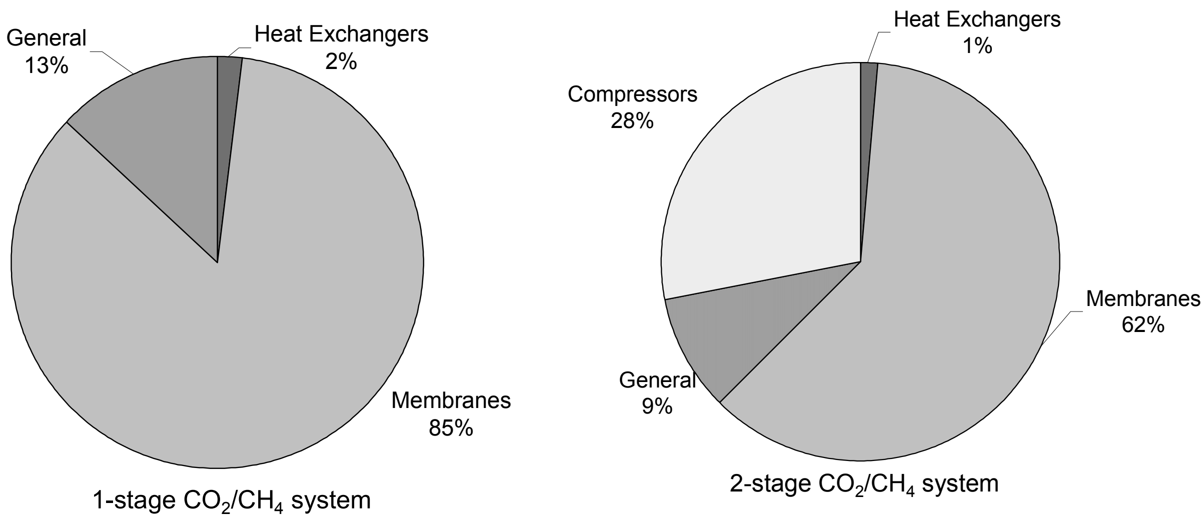

Figure 4 Relative capital expenditure cost breakdown for both a 1-stage and 2-stage CO_2/CH_4 system

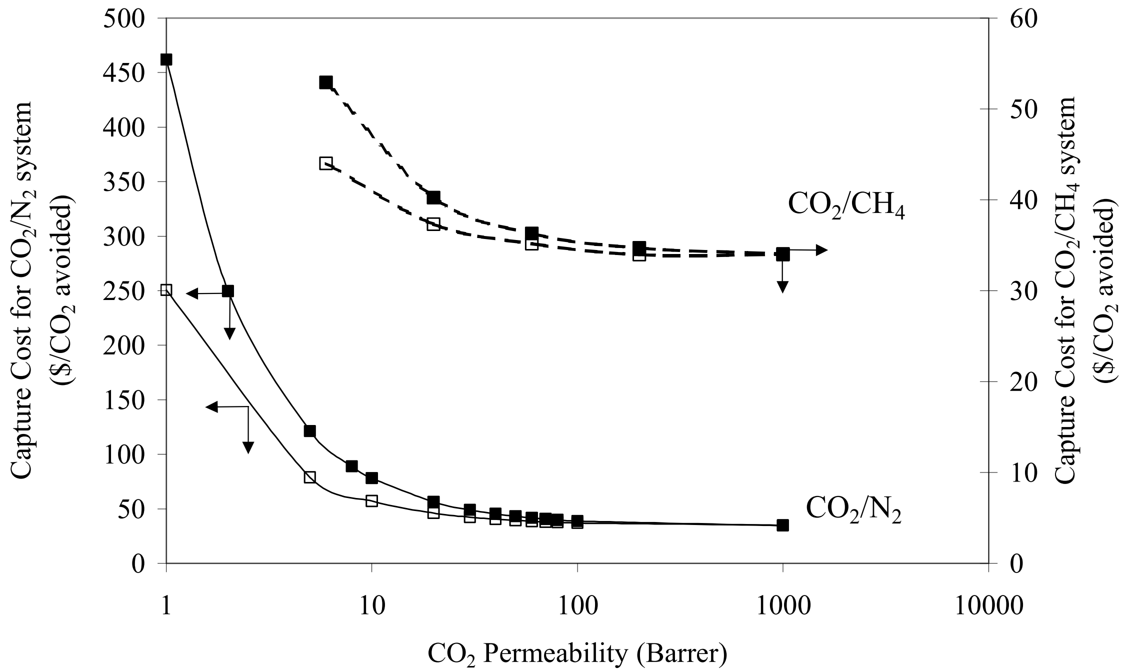

Figure 5 The effect on the capture cost for CO_2/N_2 and CO_2/CH_4 systems with changes to the CO_2 permeability at two thicknesses; ■ 0.1 μm and □ 0.05 μm

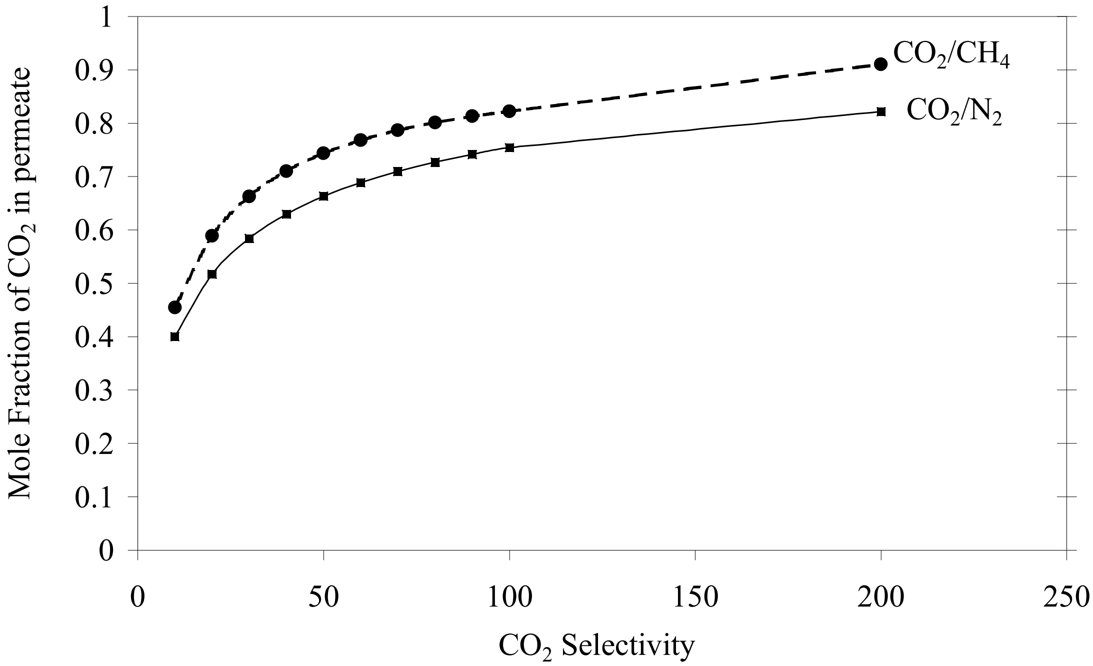

Figure 6 Effect of selectivity on CO_2 permeate purity

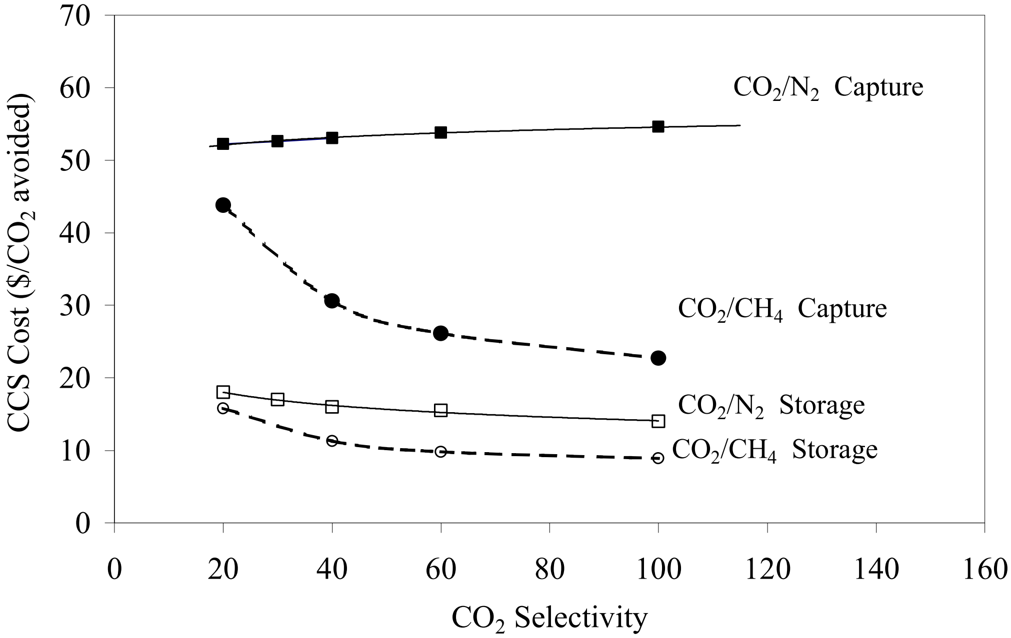

Figure 7 Selectivity effect on the capture and storage cost for CO_2/N_2 and CO_2/CH_4 systems

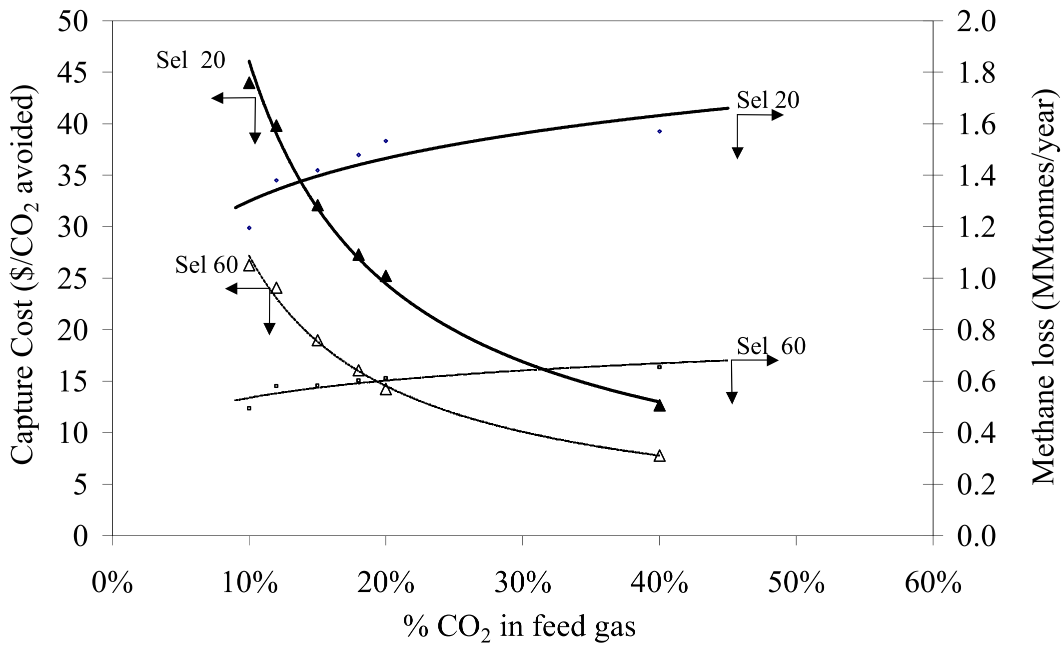

Figure 8 Selectivity effect on the capture cost and methane loss for the CO_2/CH_4 system

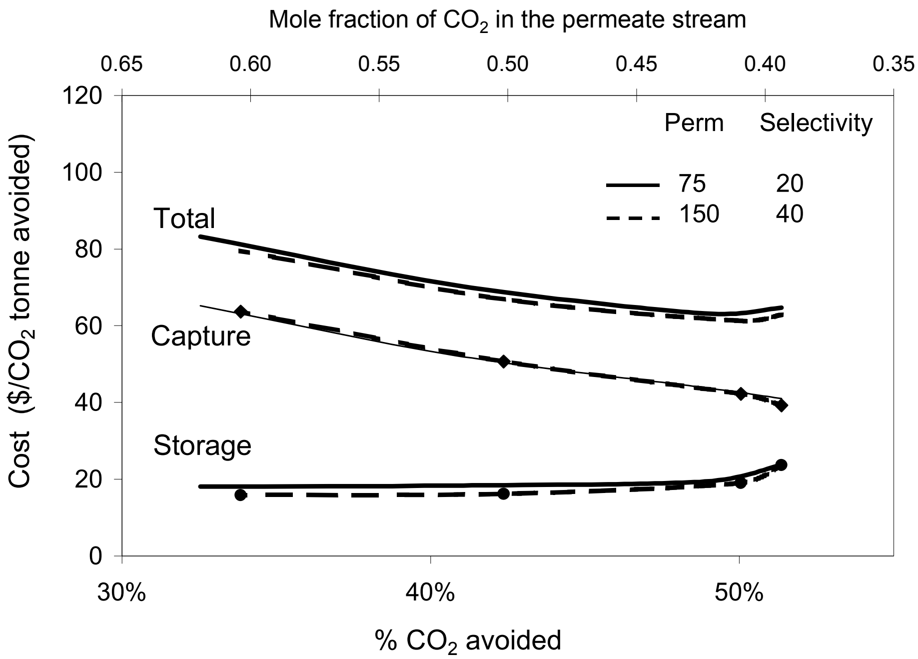

Figure 9 Combined effects of improved permeability and selectivity for CO_2/N_2 capture, storage and total CCS costs

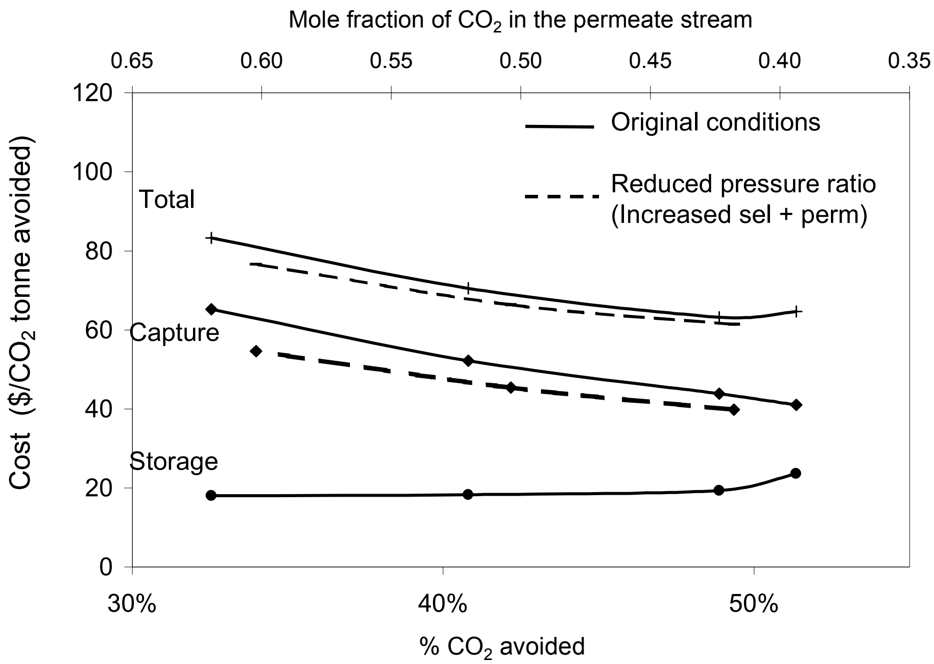

Figure 10 Effect of improved permeability, selectivity and reduced membrane pressure for CO_2/N_2 system