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A Technical Evaluation of Hybrid Membrane-Absorption Processes for Acid Gas Removal

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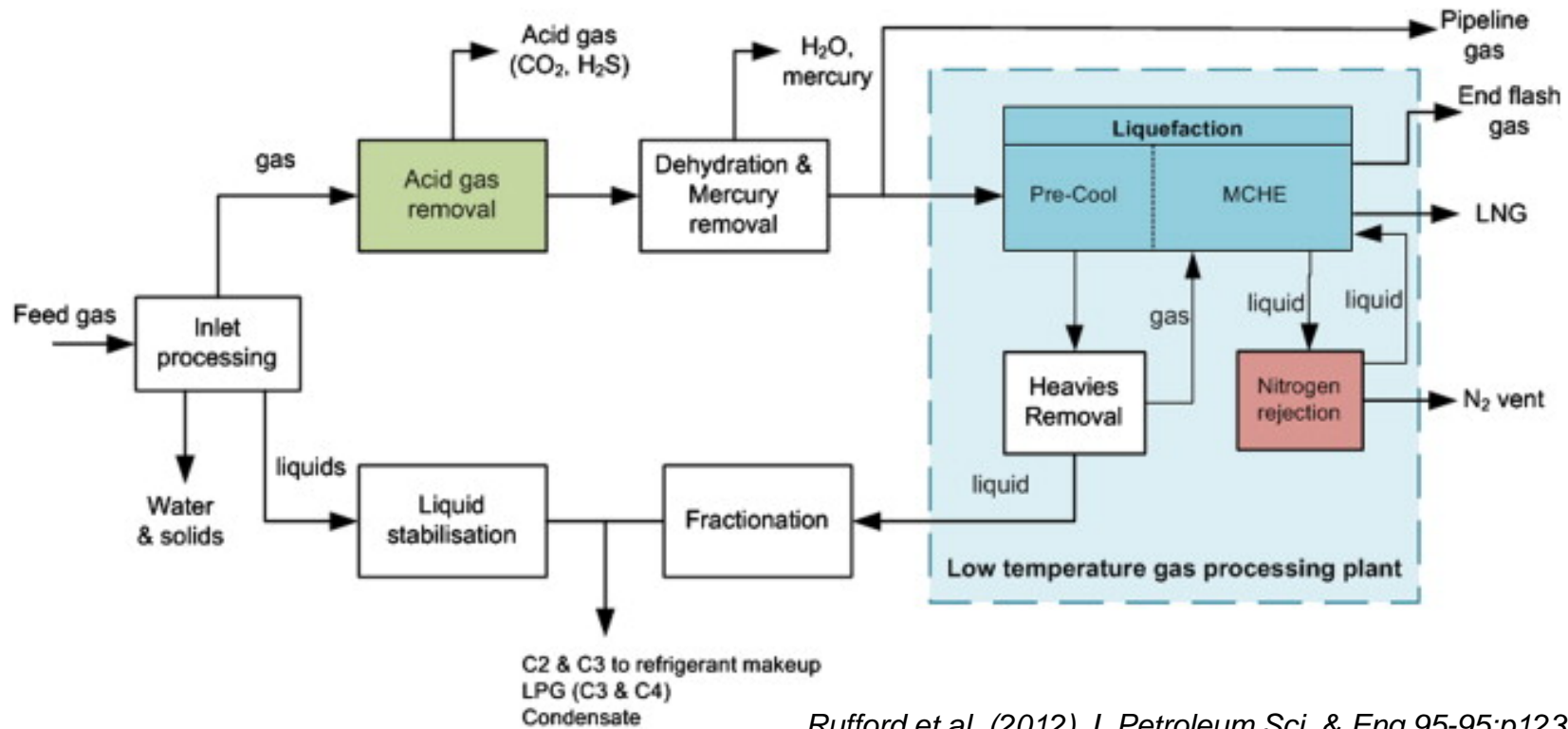
Highlights

Comparison of (1) conventional MDEA amine absorption and (2) hybrid membrane + amine process to treat sour gas for feed to an LNG plant.

Feeds with 10%mol CO₂ → no apparent benefits of a hybrid system.

Very sour feed gas with 50%mol CO₂ the hybrid system has potential for significant reductions in equipment weight, plant volume, investment costs, and operating costs.

Typical process flow scheme for LNG production



Rufford et al. (2012) *J. Petroleum Sci. & Eng* 95-95:p123-154

Challenges for gas processing in remote or stranded fields

Feed to LNG plant has tight specification, e.g.:

- $\text{CO}_2 \leq 50 \text{ppmv}$
- $\text{H}_2\text{S} \leq 4 \text{ppmv}$

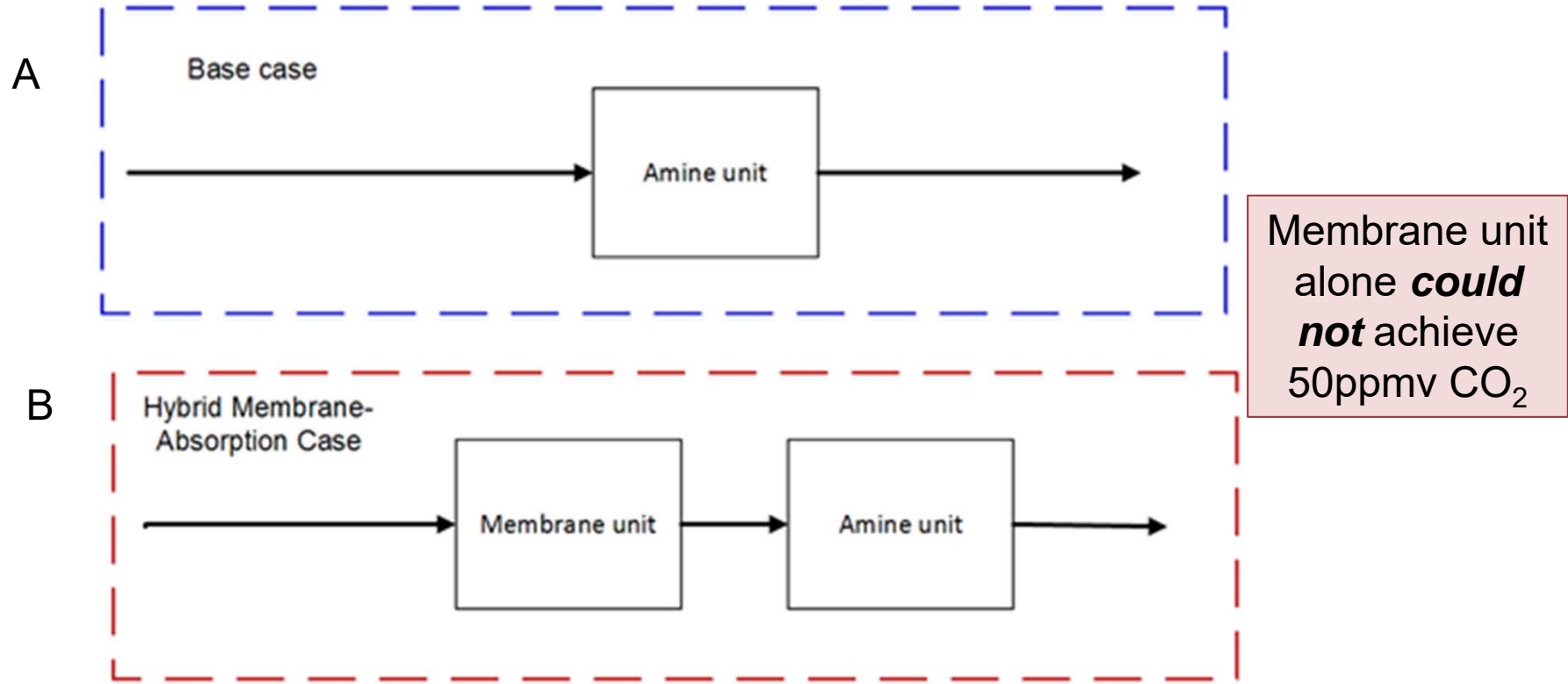
To meet these specifications amine plants require large columns, large solvent inventories, high demand for energy.

Floating LNG (FLNG) and micro-LNG plants may need significant reductions in weight, cost, and energy demand.


This project seeks to evaluate the potential of a hybrid membrane + absorption process to treat sour gas onboard FLNG plants.



Overview of the processes compared



Summary of methodology

Two feed cases developed based on
Roussanaly et al. (2014) 

Simulations in *Aspen Hysys V8.6*

Process metrics considered:

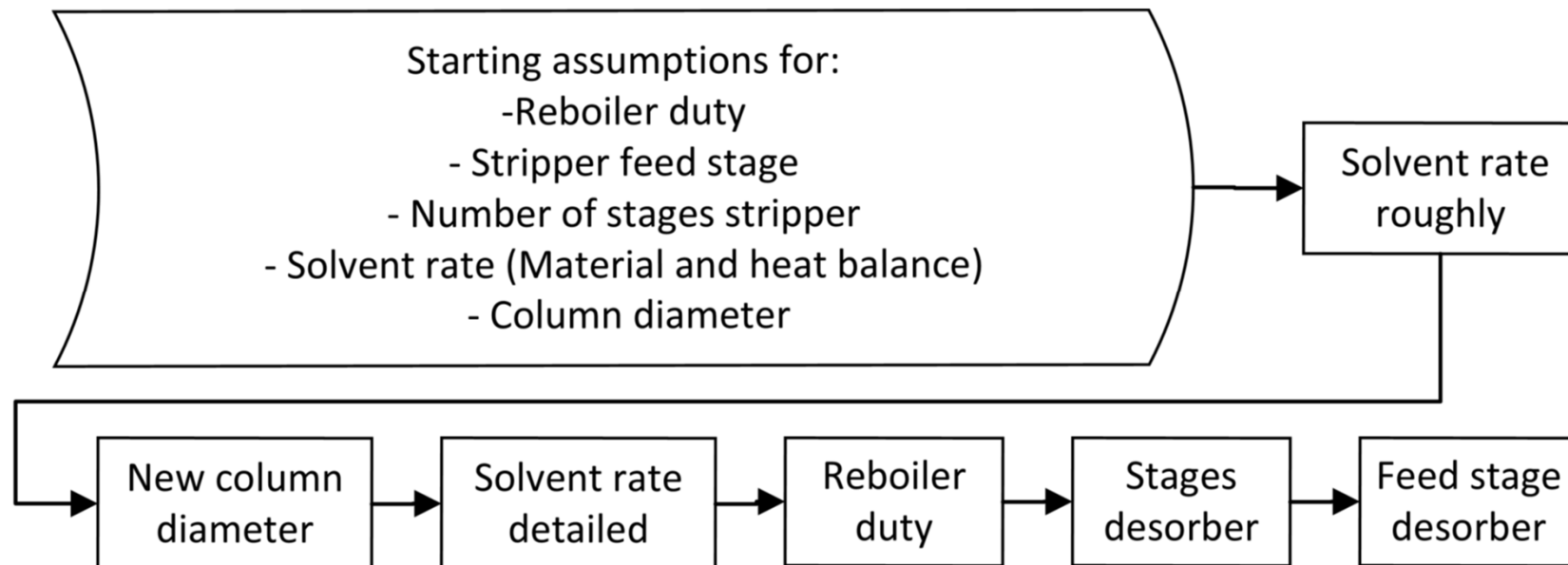
- Methane slip (MS)
- Relative energy demand (RED)
- Dry equipment weight
- Dry installed weight
- Total plant volume
- Equipment cost & installed cost
- Operating cost

Aspen Process Economic Analyzer

	Feed1	Feed2
	Feed compositions in mole %	
Methane	83	41
Ethane	5	4.5
Propane	2	3.5
CO ₂	10	50
H ₂ S	0	1
H ₂ O	0	0
N ₂	0	0
Temp., °C	40	40
Pressure, bar	70	70
Flow Nm ³ /h	590,000	590,000



General simulation strategy



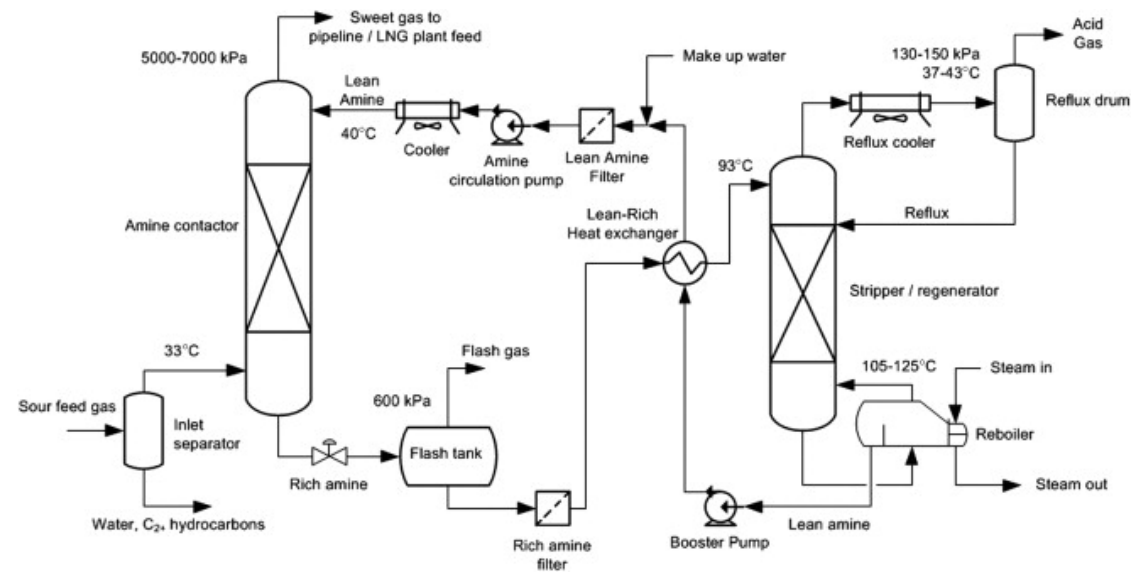
Summary of Base Case: 50% MDEA absorption

50% methyl-di-ethanolamine (MDEA) solvent + piperazine (PZ)

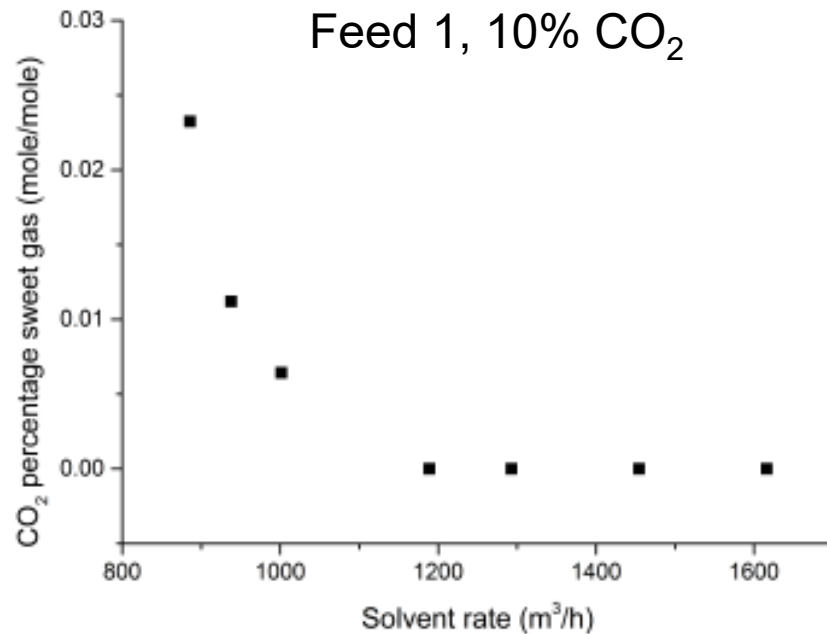
Contactor: P=70bar; Mellapak 250Y structured packing

Stage efficiencies: 0.15 CO₂; 0.8 H₂S

Stripper: 1.9 bar, 50°C



Solvent rate required to achieve 50 ppmv CO₂ spec



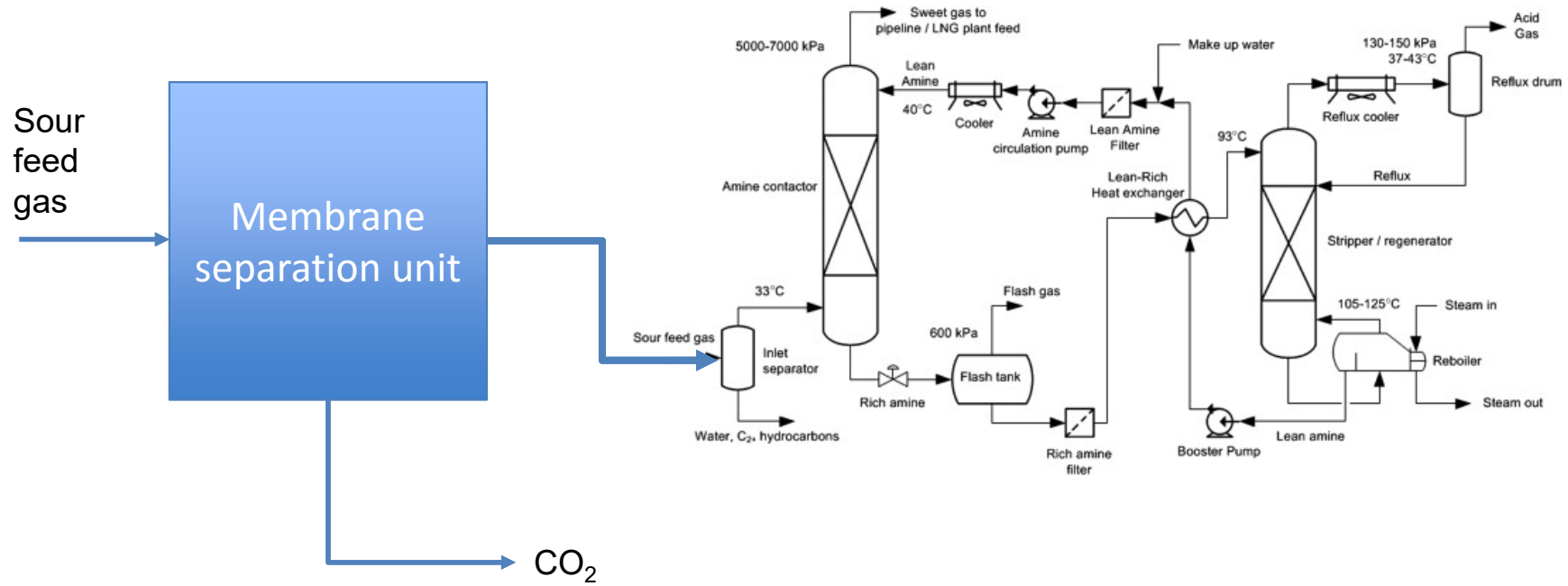
Run absorber hard to get 50 ppmv CO₂ for LNG prep. (vs 2% pipeline gas)

Optimized solvent rate = 1258m³/h

Summary amine unit requirements for Feed 1 and Feed 2

	Feed1	Feed2
CO ₂ , %mol	10	50
H ₂ S, %mol	0	1
Solvent rate m ³ /hr	1258	4105
Absorber stages	20	20
Absorber diameter, m	10	15
Stripper stages	7	7
Reboiler duty, Btu/gal lean solvent	880	880

Hybrid membrane-amine unit



Method to model membrane separation unit

Material: cellulose acetate (CA) membrane

Selectivity $P_{\text{CO}_2}/P_{\text{CH}_4} = 15$ (Niu and Rangaiah, 2014)

Selectivity $P_{\text{H}_2\text{S}}/P_{\text{CH}_4} = 19$

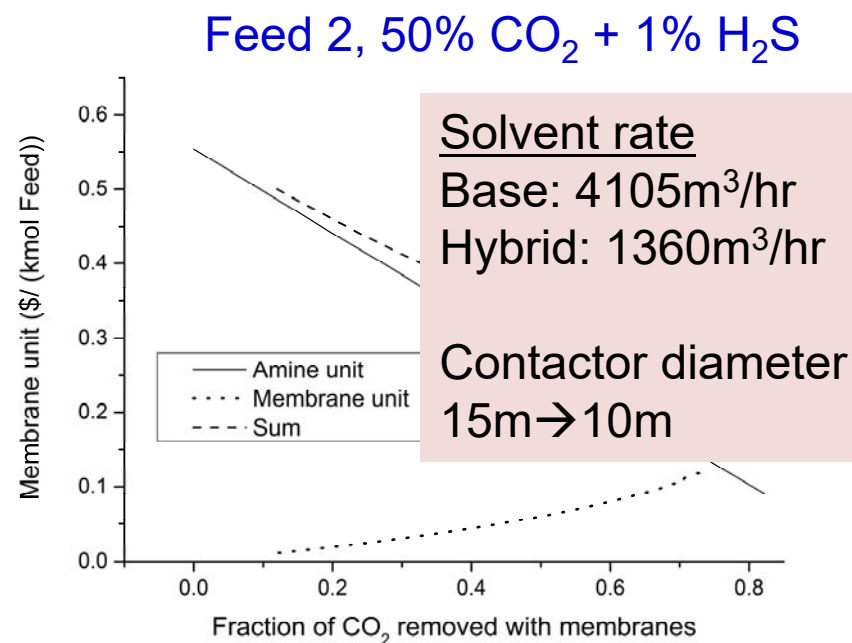
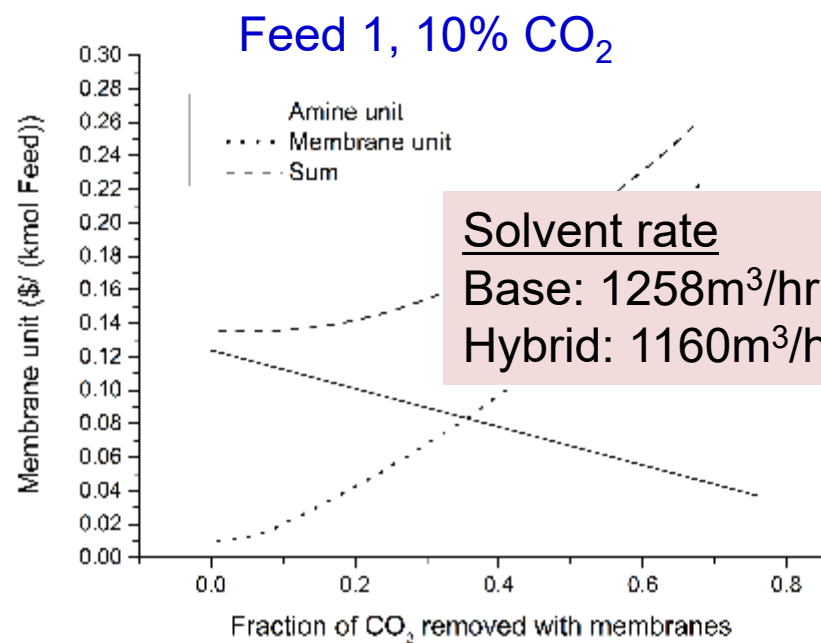
Permeate pressure = 1.38 bar

Membrane thickness = 100 nm (i.e. CA thickness on a substrate)

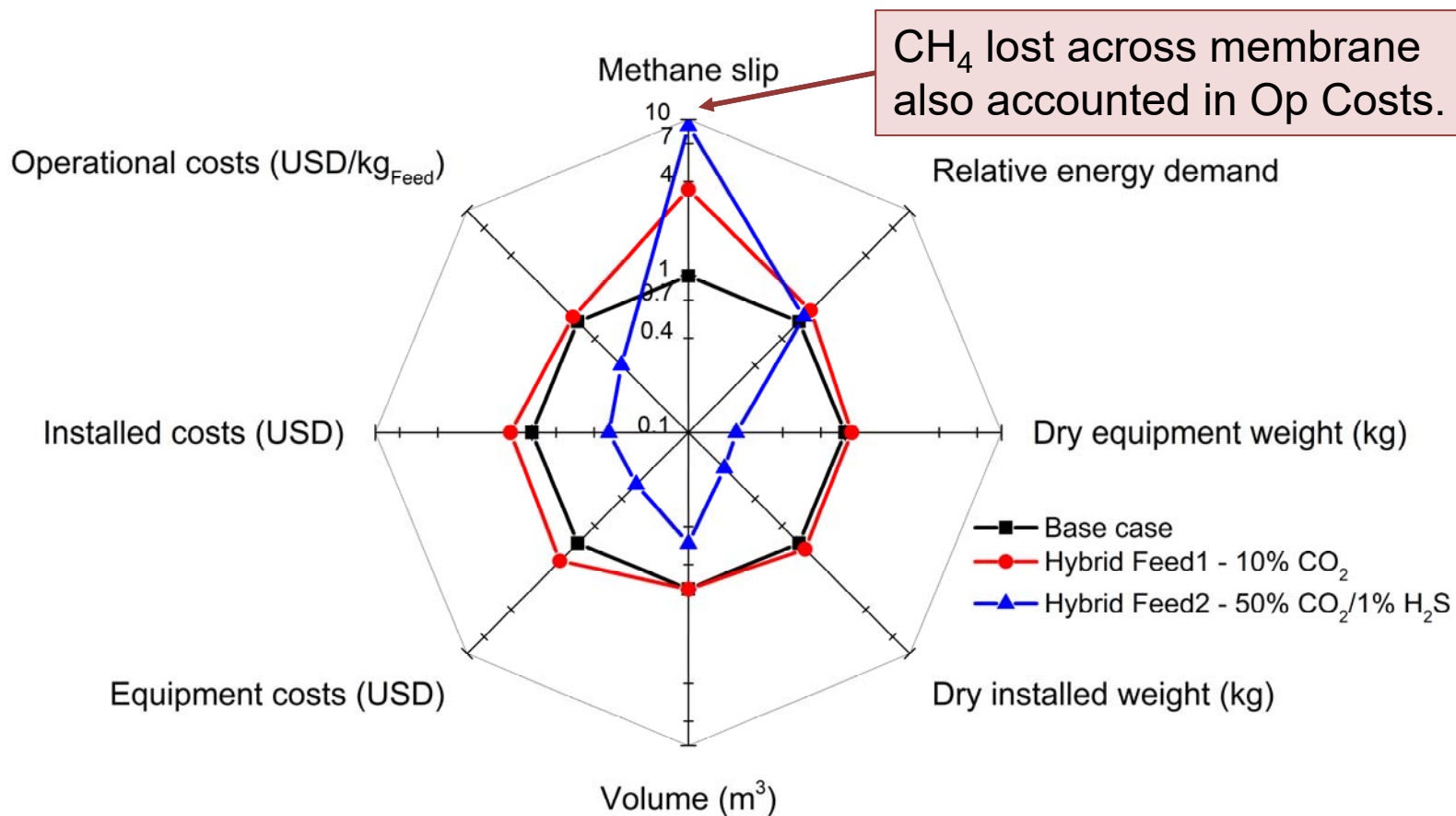
Modeled membrane unit in a user defined subflowsheet

How much CO₂ to remove in the membrane unit?

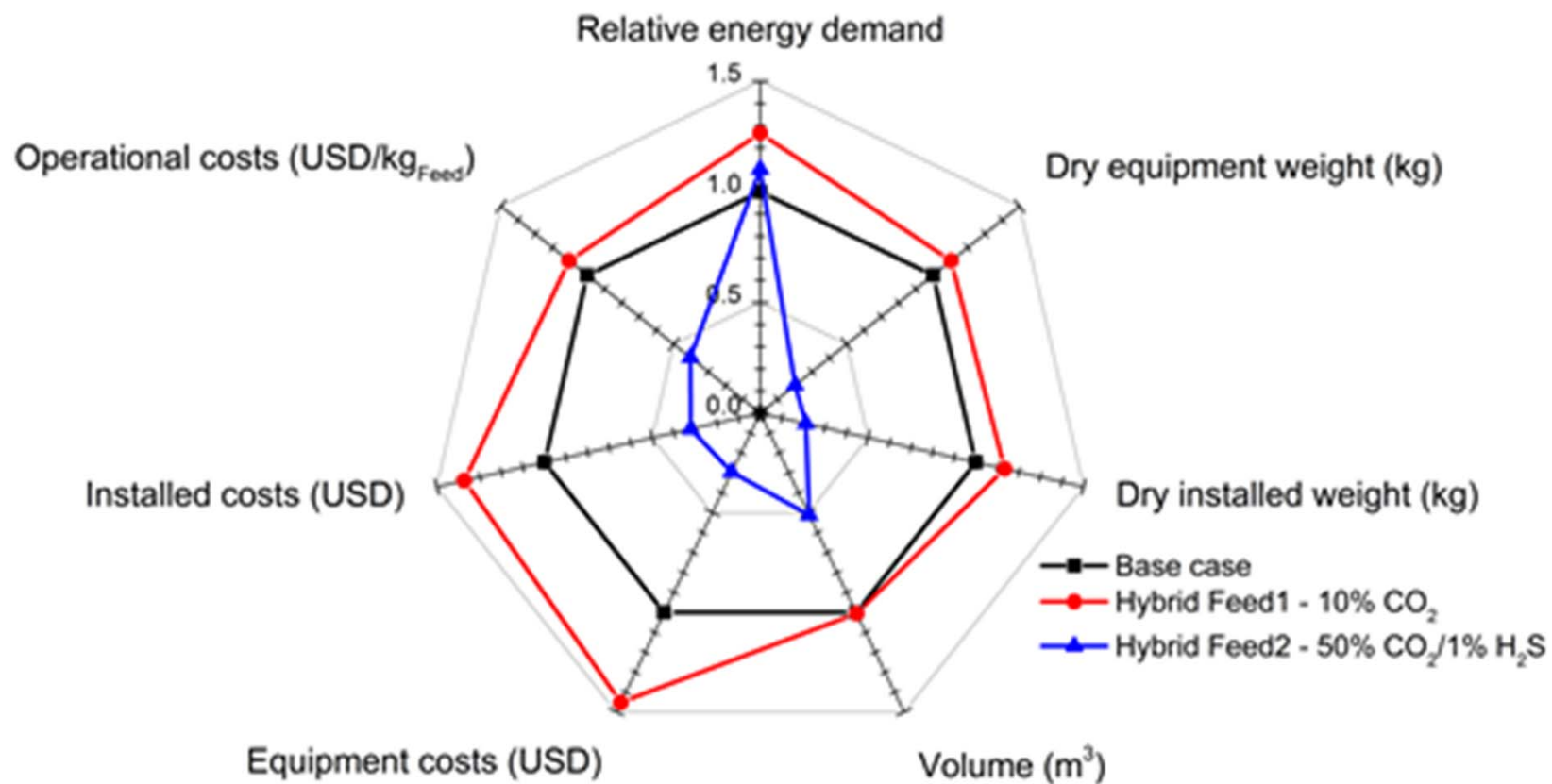
Each mole of CO₂ removed in membrane unit reduces solvent rate & reboiler duty required in the amine section. *Control = membrane area.*
 → Reducing hydraulic load also effects equipment sizing



Process metrics spider plot (log scale)



Comparison of process metrics



Feed 1 10% CO₂ summary

No clear advantage of hybrid system for this feed scenario and membrane properties.

- 7.8% lower solvent rate in hybrid process, doesn't impact column size
- 30% drop in energy for reboiler, pumps in amine unit

Those benefits offset by:

- methane slip in membrane unit
- Small increase in equipment weight (MSU + base case amine unit)
- Hybrid process costs all increase

Feed 2 50% CO₂ + 1% H₂S Summary

For very sour gas feed there is potential to use bulk separation properties of the membrane unit.

- 67% reduced solvent rate in hybrid process
- 80% reduced equipment weight, 50% reduced plant volume

Methane slips increases → ~10% CH₄ in feed lost across membrane

- Still energy savings allow 40% reduction in operating costs
- Methane slip has implications on any CO₂ processing/storage plans

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Niu, Mark Wendou, G. P. Rangaiah. 2014. Retrofitting amine absorption process for natural gas sweetening via hybridization with membrane separation (in *International Journal of Greenhouse Gas Control* **29**: 221-230)

Roussanaly, S., R. Anantharaman, K. Lindqvist. 2014. Multi-criteria analyses of two solvent and one low-temperature concepts for acid gas removal from natural gas (in *Journal of Natural Gas Science and Engineering* **20**: 38-49).

Rufford, Thomas E., Simon Smart, Guillaume C.Y. Watson et al. 2012. The removal of CO₂ and N₂ from natural gas: A review of conventional and emerging process technologies (in *Journal of Petroleum Science and Engineering* **94-95**: 123-154).



Methane slip (MS) The methane slip depicts the percentage of methane lost during the purification process. It is defined in Equation 5.1.

$$MS = 1 - \frac{CH_4 \text{ in the product gas}}{CH_4 \text{ in the raw natural gas}} \quad (5.1)$$

Relative energy demand (RED) This indicator is the ratio of lost and required energy based on the combustion heat of the product gas \dot{Q}_{Prod} , depicted in Equation 5.2. The absolute energy demand can be calculated by summing up the differences in the combustion heats of the raw gas \dot{Q}_{Raw} along with the product gas \dot{Q}_{Prod} , electrical energy P_{el} and the required heat of the reboiler \dot{Q}_{Reb} . The electrical energy P_{el} in the gas facilities is provided by gas turbines. For calculating P_{el} a turbine efficiency of $\eta = 0.4$ is assumed.

$$RED = \frac{\dot{Q}_{Raw} - \dot{Q}_{Prod} + P_{el}/\eta + \dot{Q}_{Reb}}{\dot{Q}_{Prod}} \quad (5.2)$$

Operational costs Table 5.2 provides the information needed for calculating the operational costs. Maintaining costs are neglected in this paper.

Table 5.2: Values for calculating the operational costs of base and hybrid case

	Value	Unit	Source
Cost for reboiler steam	0.0145	\$/kg	[20]
Cost for electricity	0.07	\$/kWh	[20]
Cost for cooling water	0.01	\$/t	[20]
Cost for process water	0.0005	\$/kg	[20]
CH ₄ Loss	4	\$/MMBtu	[20], [39]
Lower heating value methane	10	kWh/m ³	[40]
Cost for MDEA	5.5	\$/kg	[41]
Cost for PZ	7.7	\$/kg	[41]
Solvent Degradation	0.25	kg/t _{CO₂}	[20]
Temperature saturated steam	140	°C	
Efficiency of reboiler heating	0.9		