Enzymatically Driven, Cost-Efficient, Capture of Flue Gas CO_2

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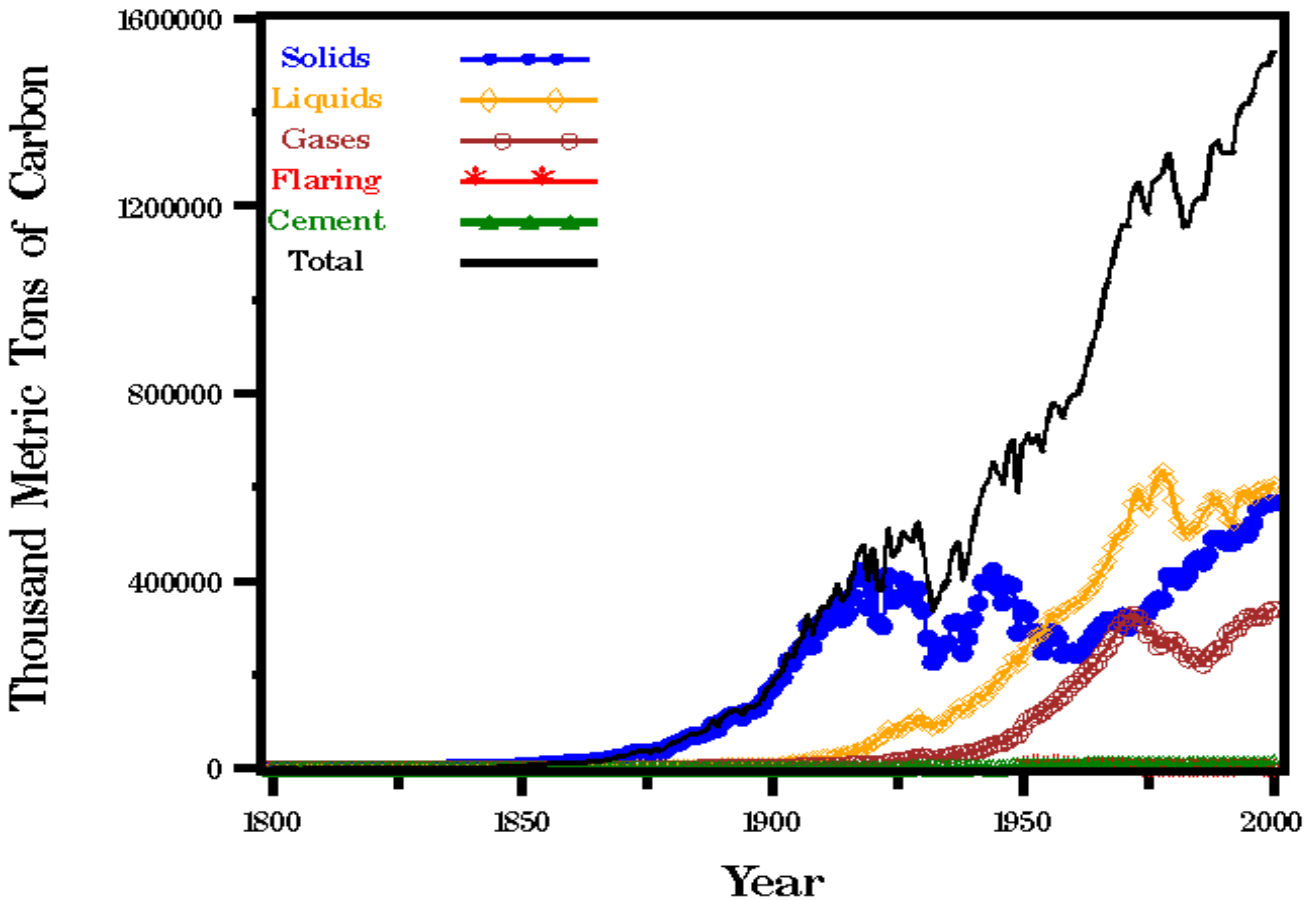
Outline

- Importance of CO_2 capture
- Current technology
- Membrane technology
- HFCLM
- Results
- Process Engineering
- Economics
- Conclusions

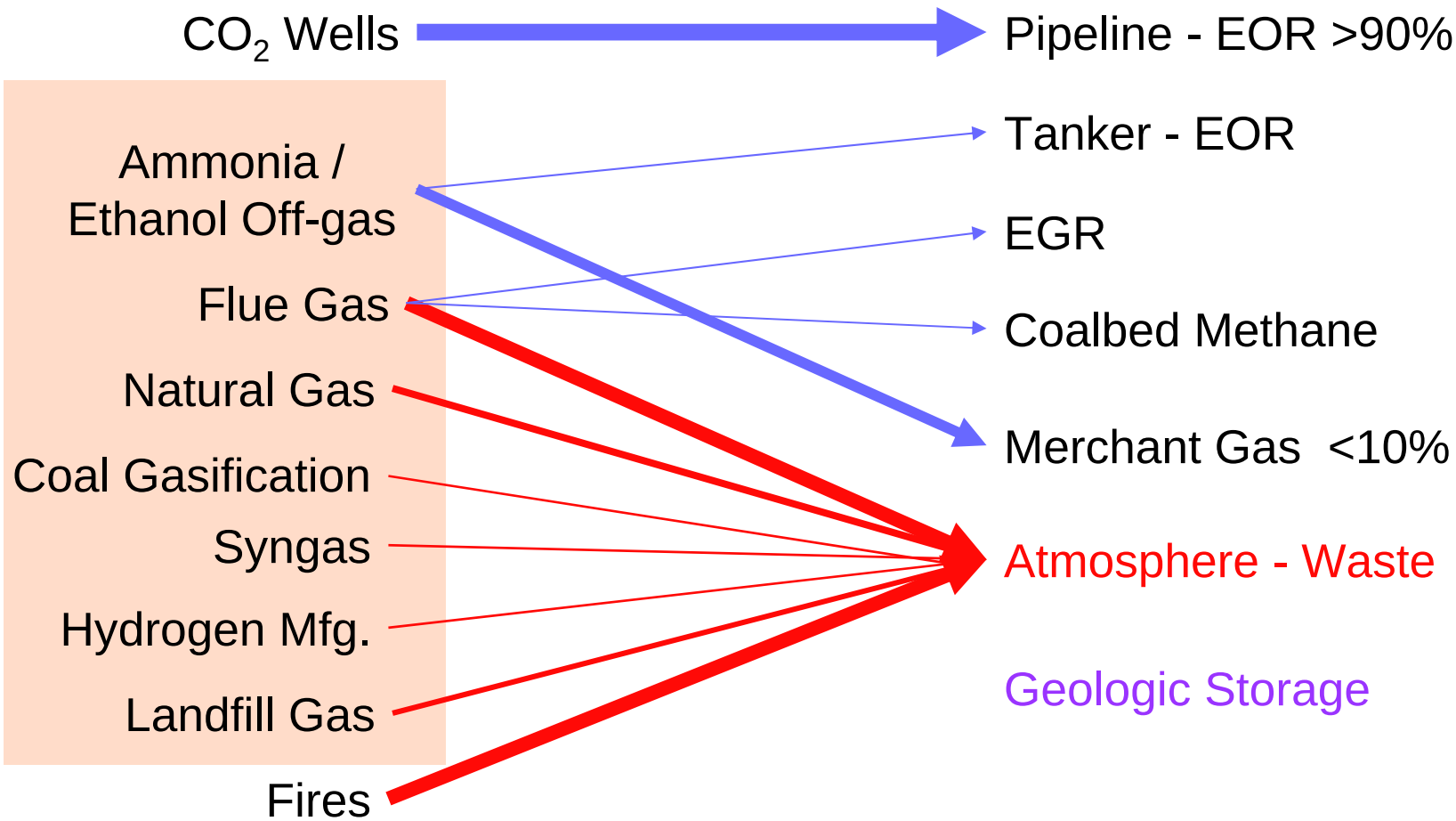
CO_2 Emissions in the USA



Source: http://cdiac.esd.ornl.gov/trends/emis/em.cont.htm

SOURCES and USES of CO_2

Today

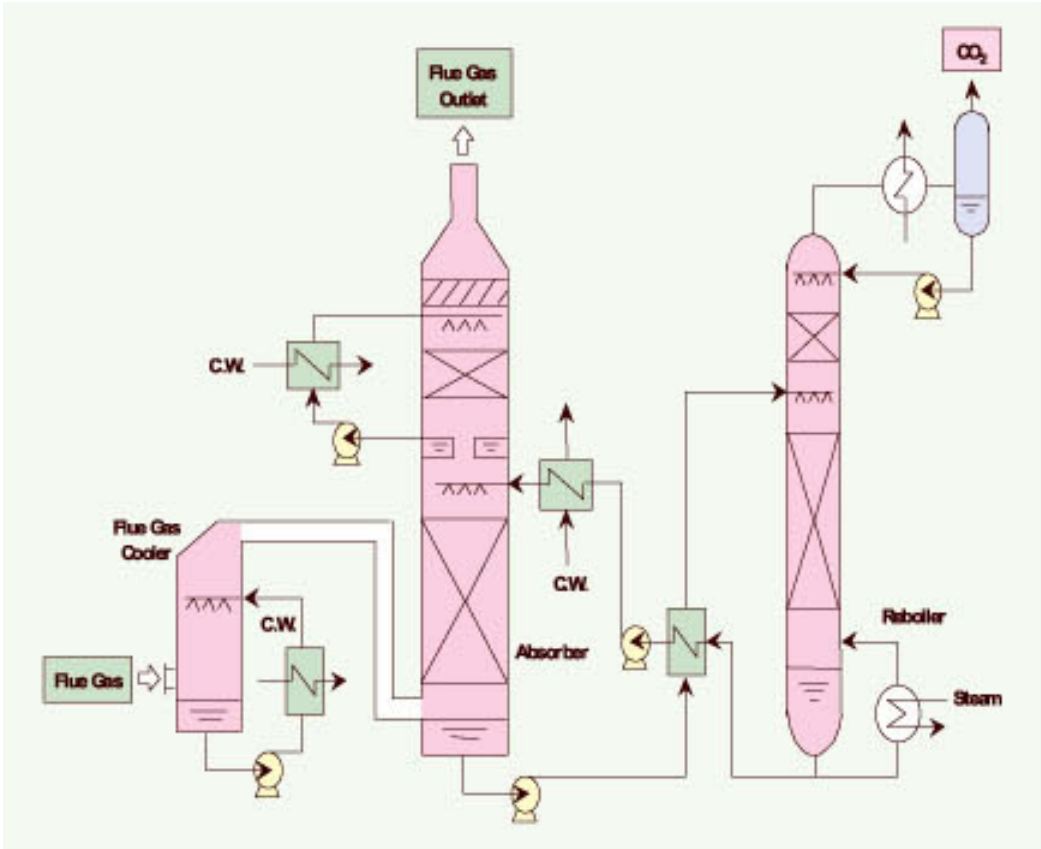


Current Technology

- Adsorption
- Physical Absorption
- Chemical Absorption
  - Potassium Carbonate
  - Primary Amine
  - Facilitated Carbonate
    - Piperazine
    - Secondary Amine
    - Carbonic Anhydrase

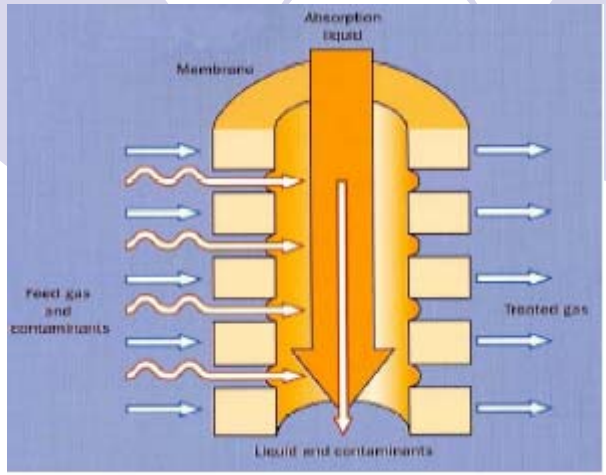
Current Technology - Amine

- Absorber/Stripper
- Energy intensive
  - Liquid Pumping
  - Thermal Amine Reactivation
- Corrosive
- Amine degradation and loss

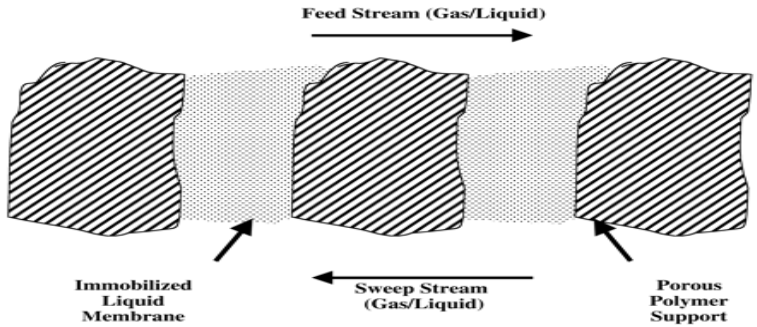


Liquid Membrane Technologies

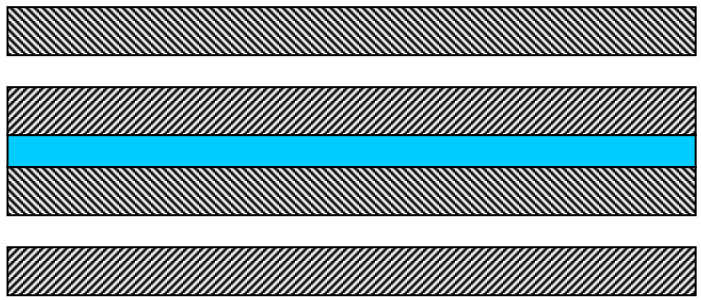
Membrane Contactor



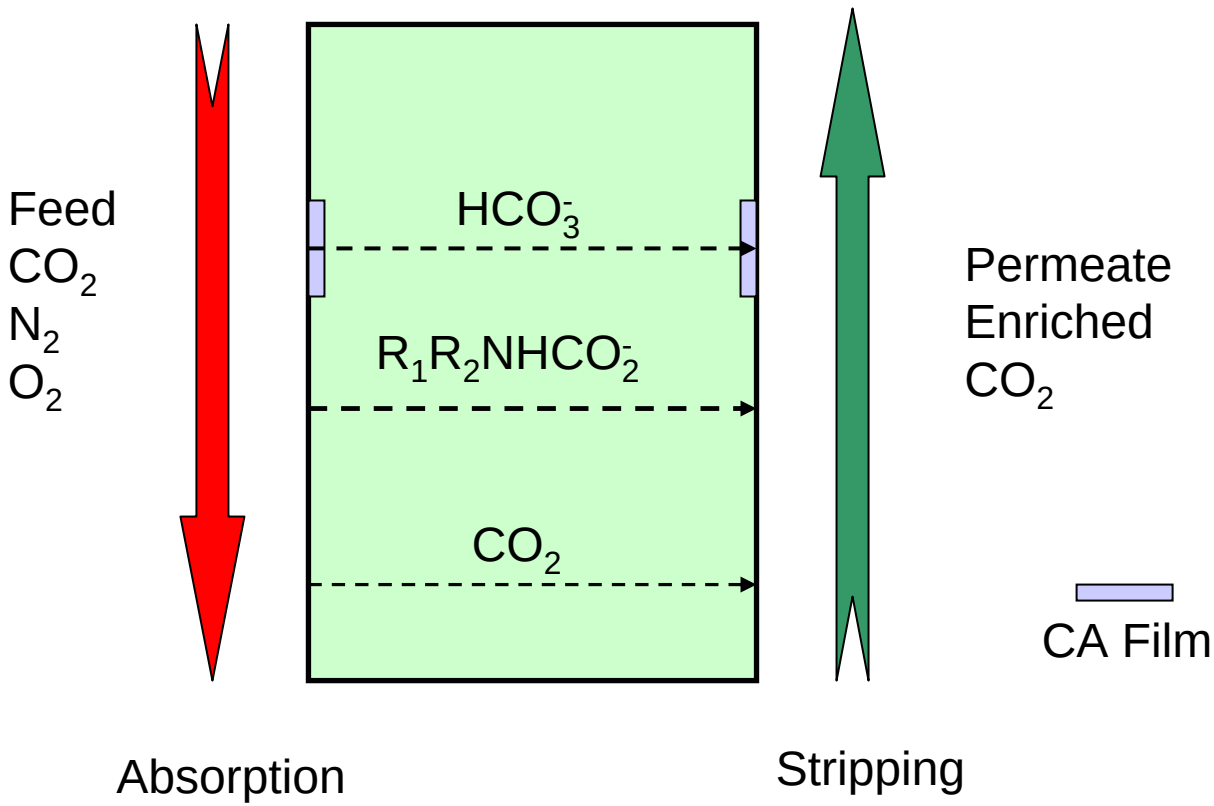
Supported/Immobilized Liquid Membrane (SLM/ILM)



Contained Liquid Membrane (CLM)

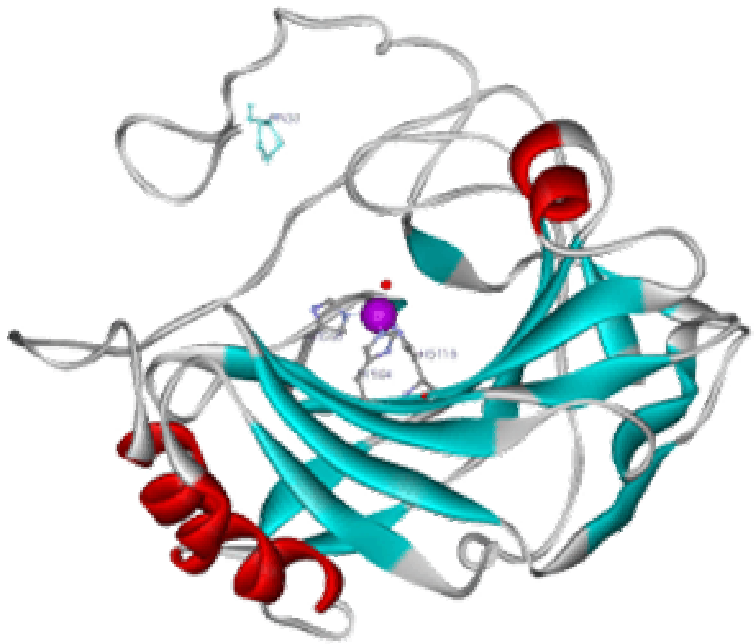


Facilitated Membrane Transport

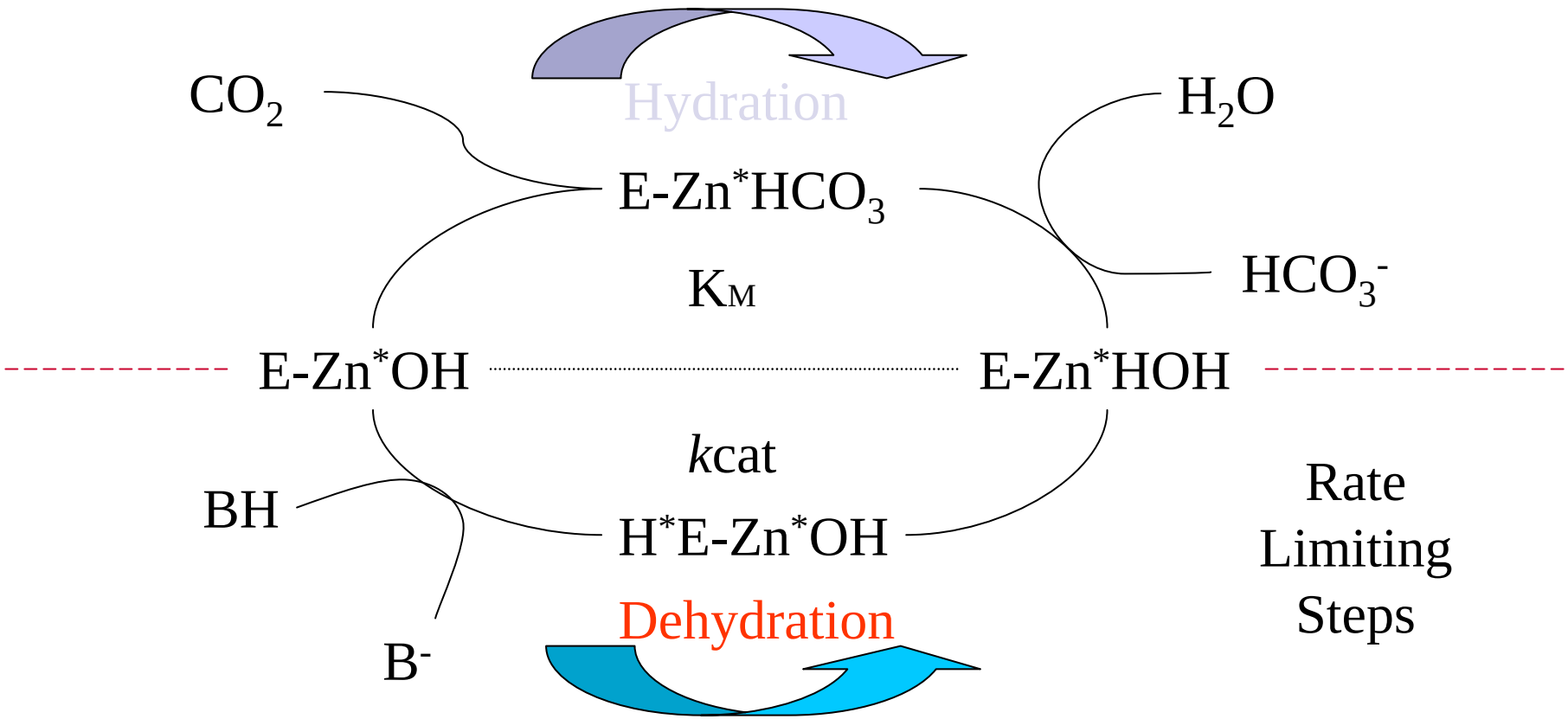


Carbonic Anhydrase (CA)

- The fastest catalyst for CO_2 hydration and dehydration reaction.
- 6 families
- Many isozymes



CA/H_2O/CO_2/HCO_3^- Reaction



Simulation – CA/HCO_3^-/CO_2

- Facilitated Mass Transport Model

$\alpha = [CO_2]/[CO_2]_0$; $\beta = [HCO_3^-]/C_M$;

$K_{11} = (L^2*kcat*E_0)/(DCO_2*[CO_2]_0)$; $K_{22} = Kmf*C_M/(Kmr*[CO_2]_0)$;

$K_{33} = Kmf/[CO_2]_0$; $K_{44} = (L^2*kco_2)/DCO_2$;

$K_{55} = (L^2*k_{OH}*KW)/(2*DCO_2*K_3)$;

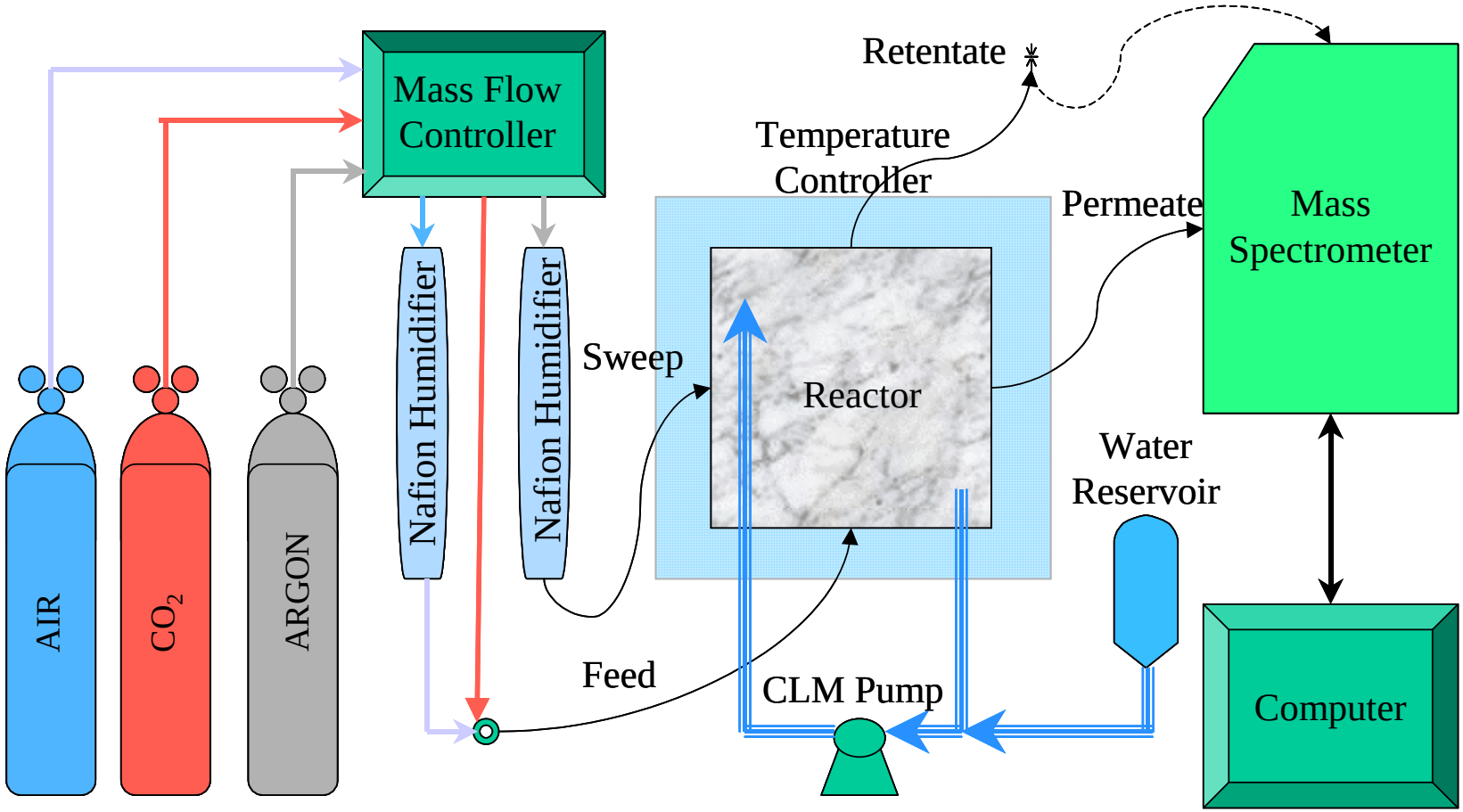
$K_{66} = -2*(L^2*C_M*kcat*E_0*K_3)/(DCO_2*[CO_2]_0^2*K_{Eq})$;

$K_{77} = -2*(L^2*C_M*k_{CO2}*K_3)/(D_{CO_2}*[CO_2]_0*K_{Eq})$;

$K_{88} = -(L^2*C_M*K_W*k_{OH})/(D_{CO_2}*[CO_2]_0*K_{Eq})$;

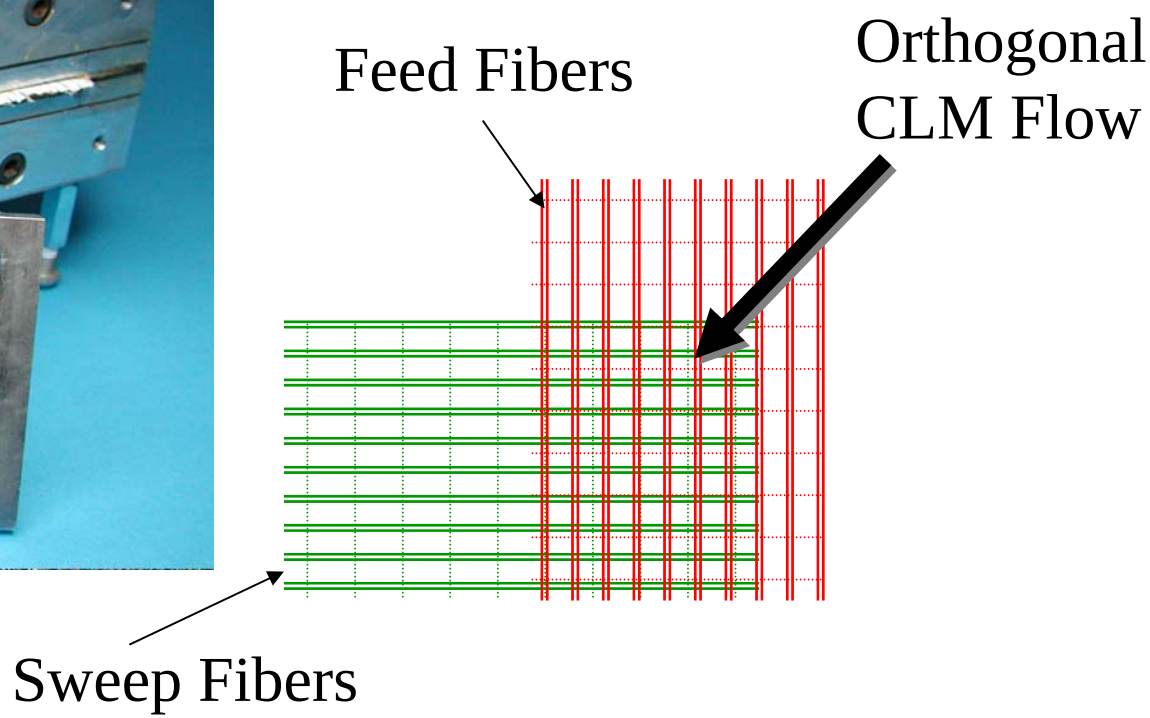
$$\frac{d^2\alpha}{dy^2} = \frac{K_{11}\alpha}{\alpha + K_{22}\beta + K_{33}} + K_{44}\alpha + K_{55} * (\frac{\alpha}{\beta} - \alpha) + \frac{K_{66}\beta^2}{(\alpha + K_{22}\beta + K_{33})(1-\beta)} + \frac{K_{77}\beta^2}{1-\beta} + K_{88}\beta$$

Experimental Setup



Hollow Fiber Contained Liquid Membrane (HFCLM) Permeator

Orthogonal Fibers

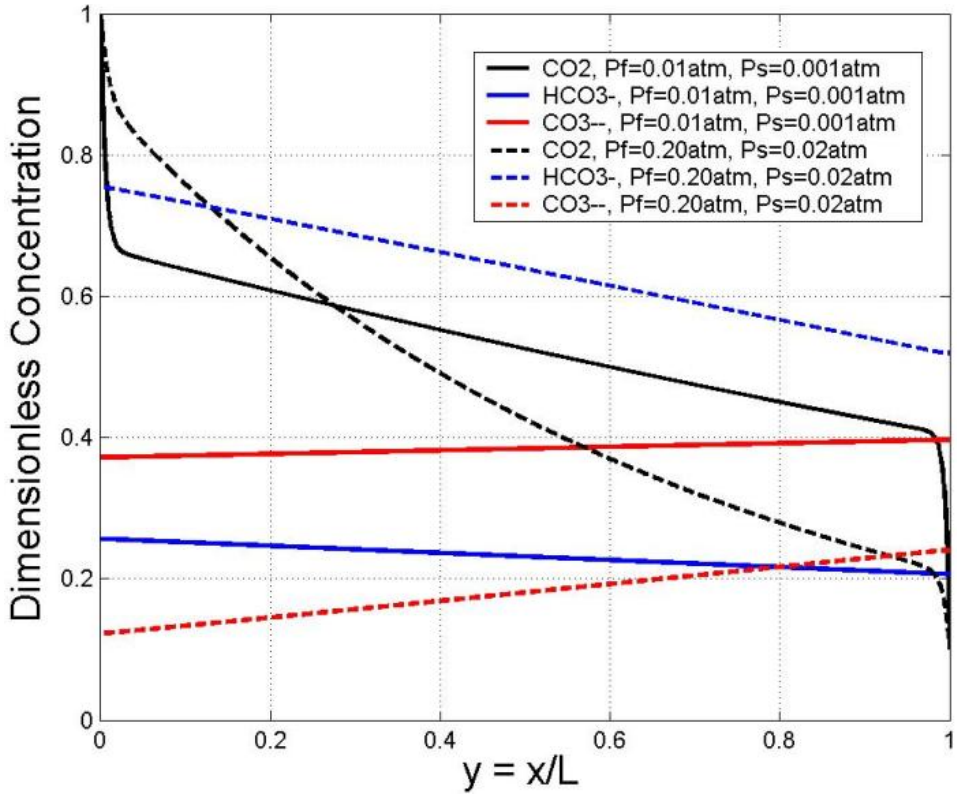


Simulation Results

- Transmembrane Concentration Profile - 3.0 mg/ml CA + 1.0M $NaHCO_3$

The experimental parameters used for this simulation are:

L(CLM thickness) = 200μm
CO_2%(f) = 1.0%, CO_2%(s) = 0.1%
or
CO_2%(f) = 20.0%, CO_2%(s) = 2.0%
Pf = Ps = 101.3 kPa
T (temperature) = 298.15 K

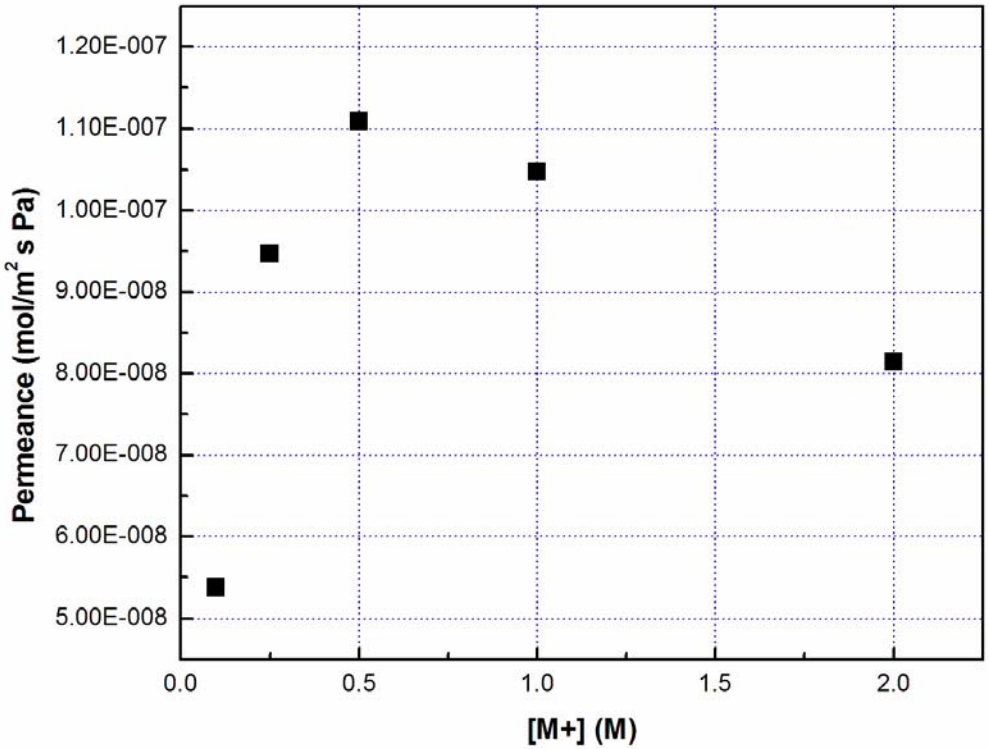


Effect of Metal Bicarbonate

- High [M+]:
  - high [HCO_3^-]gradient is possible, high facilitation, high selectivity
  - low diffusivity, longer time is required to reach steady-state

Parameters:

%CO_2 (feed) = 1%
%CO_2 (sweep) = 0.1%
L = 400 μm
[CA] = 3 mg/ml
*k*cat = 1E6 (s^{-1})
No external mass transport resistance

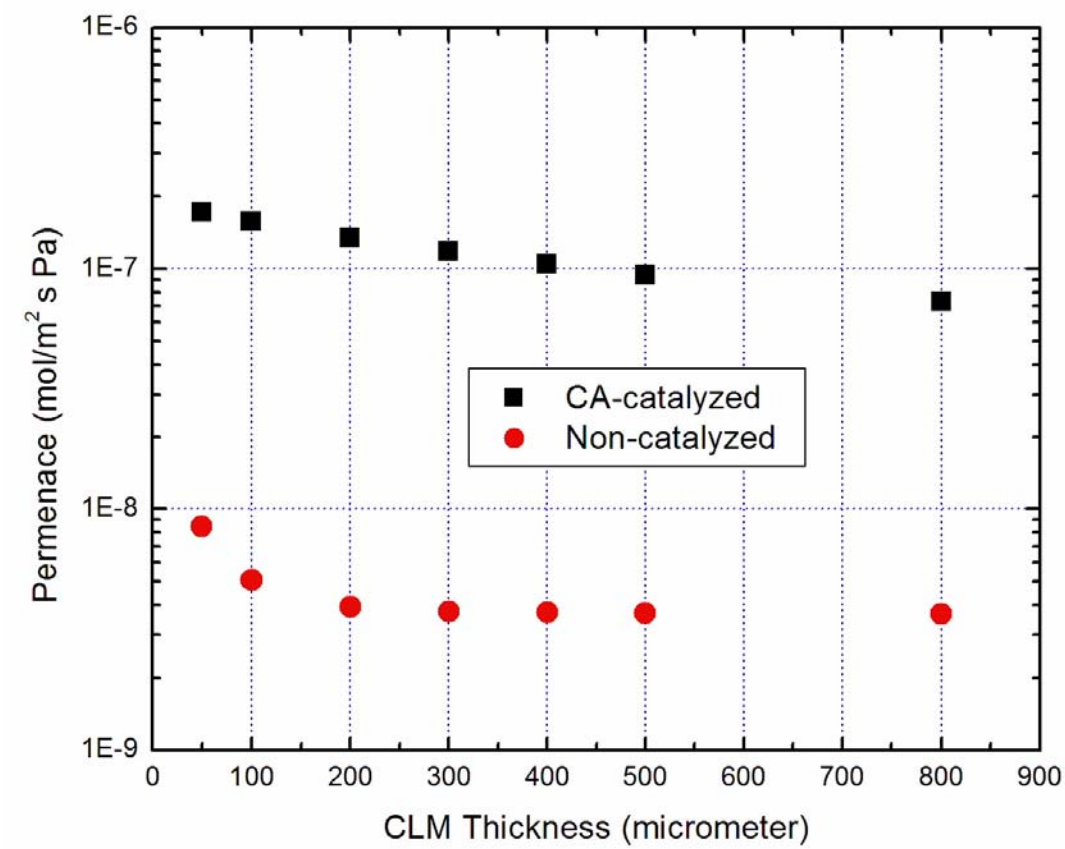


Effect of CLM thickness

- Thiele Modulus, $\Phi = (k\ L^2/D)^{1/2}$
- Fast reaction / Thick membrane → Chemical Equilibrium
- Permeance $\propto$ 1/L

Parameters:

%CO_2 (feed) = 1%
%CO_2 (sweep) = 0.1%
*k*cat = 1E6 (s^{-1})
[CA] = 3 mg/ml
[M+] = 1M
No external mass transport resistance



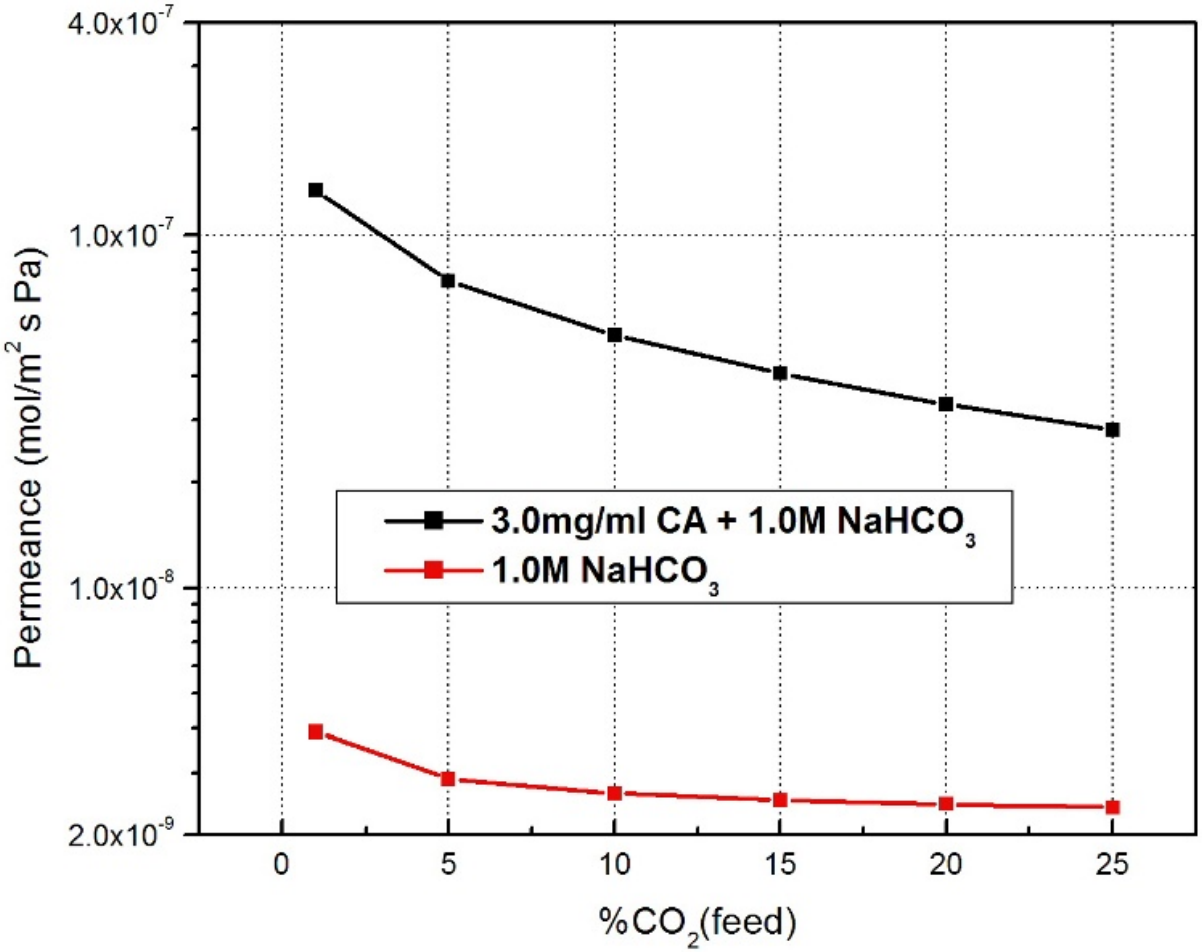
Calculated Permeance

CO_2%(Feed)	CO_2%(Sweep)	CO_2 permeance (mol/m^{2*} s Pa)
5.0	0.5	5.09E-8
10.0	1.0	3.34E-8
15.0	1.5	2.5E-8
20.0	2.0	1.99E-8

**** Based on effective membrane area***

Results - Simulation

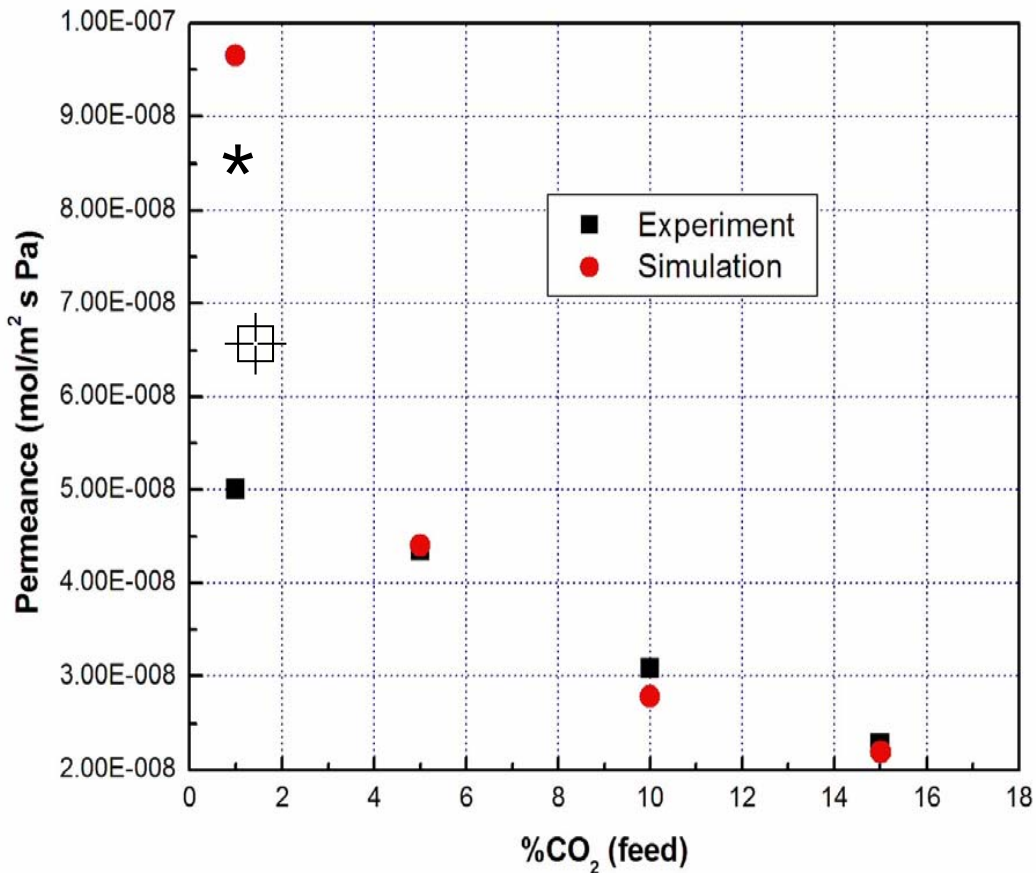
- Permeance vs. CO_2%
- CA increase CO_2 permeance 10 times or more



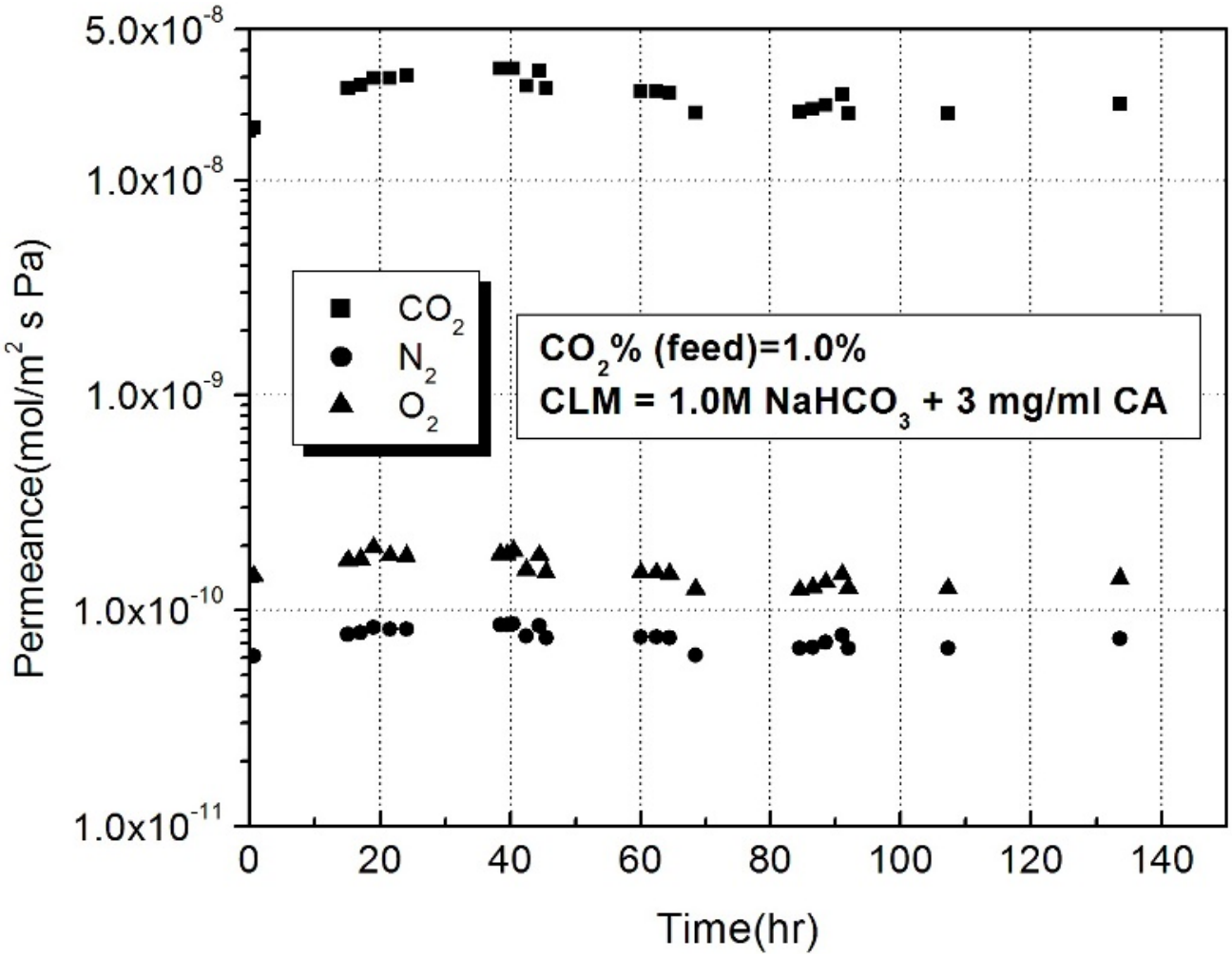
Model Validation – Actual vs. Predicted

- Celgard X30-240 micro-porous hollow fiber
- # of feed fibers = # of sweep fibers
- nominal porosity = 40%
- Total membrane surface area = 0.189 m^2
- Effective membrane area = 0.076 m^2
- No CLM pumping

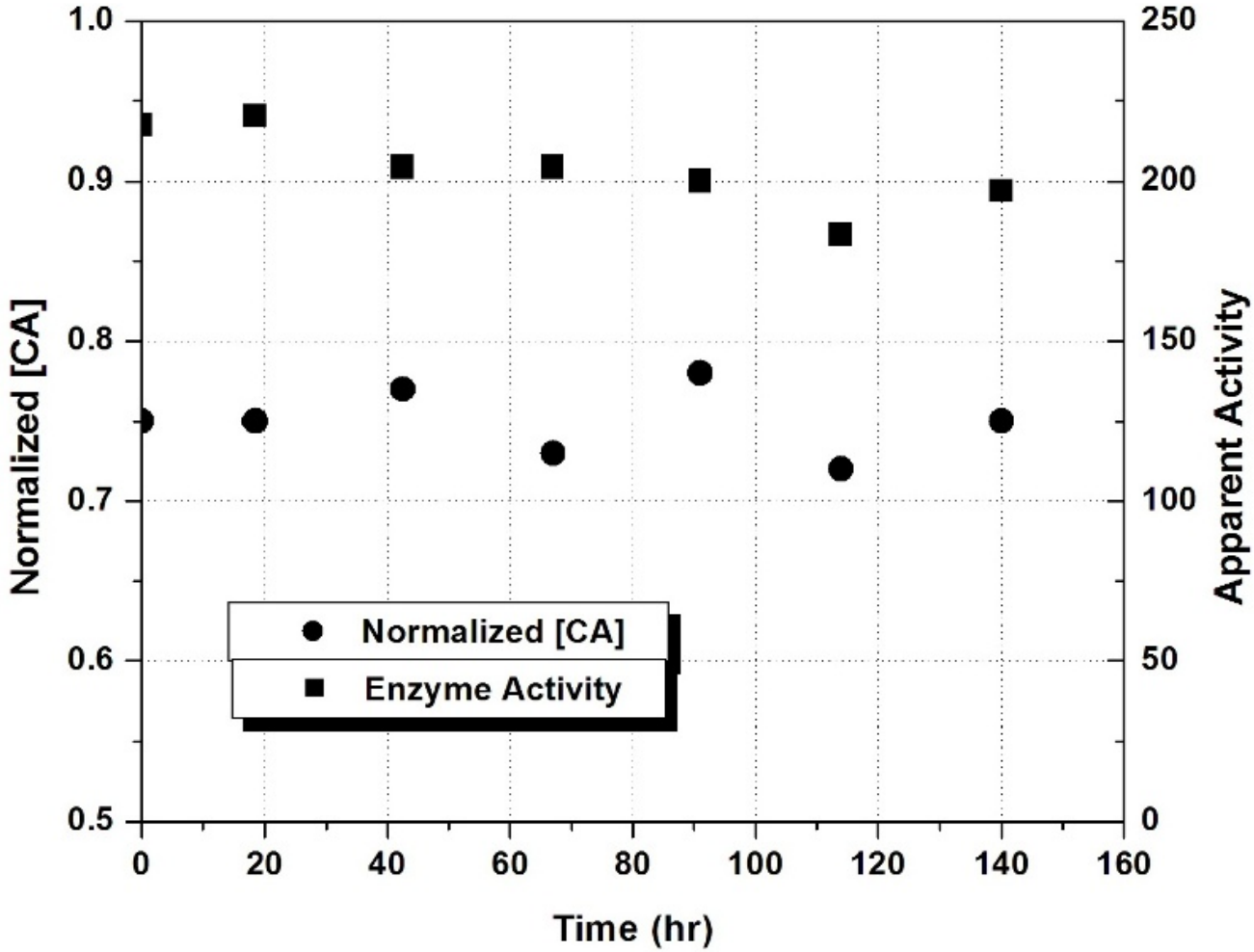
In the simulation *k*cat = 1E6 (s^{-1})



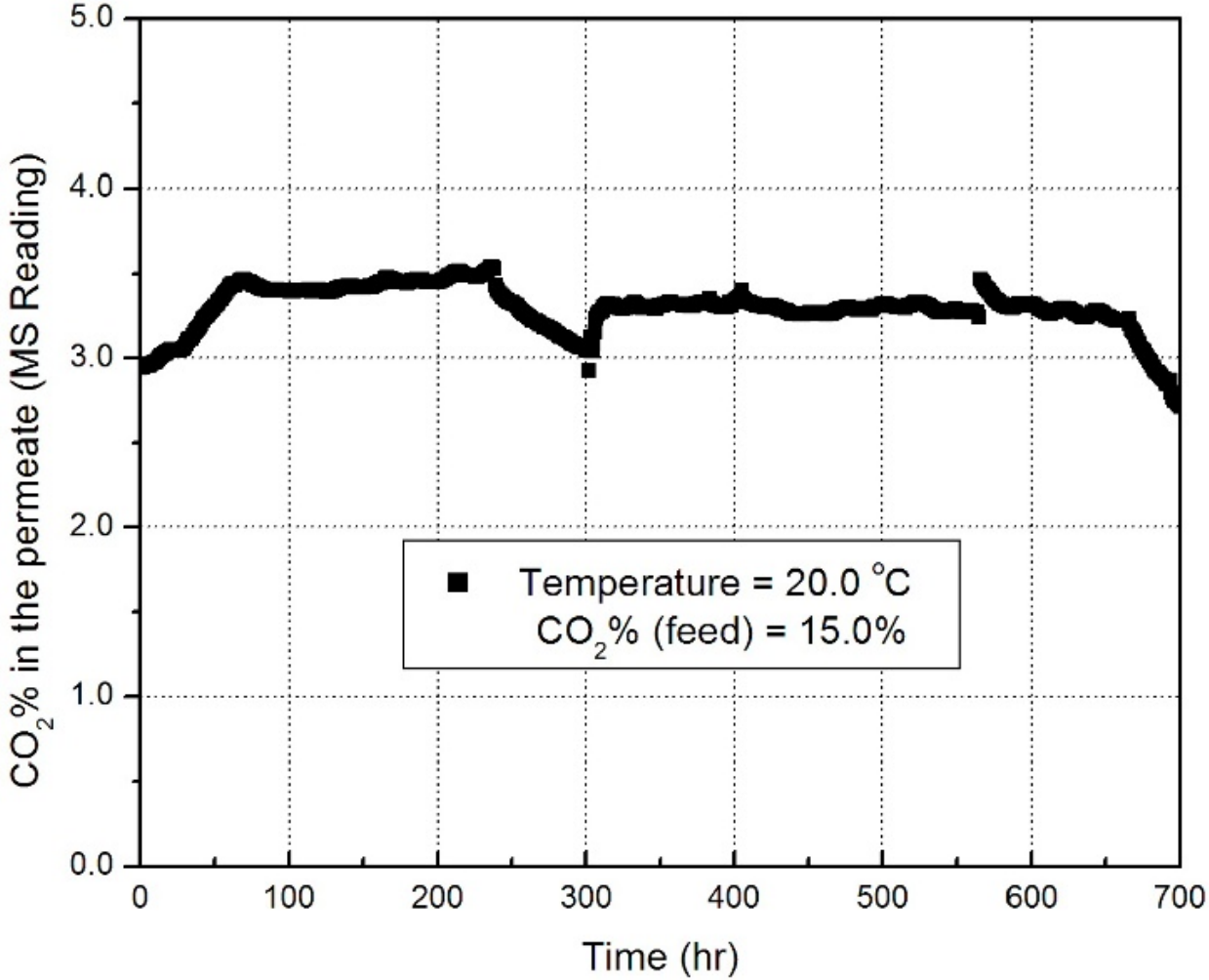
Stability and Longevity of HFCLM



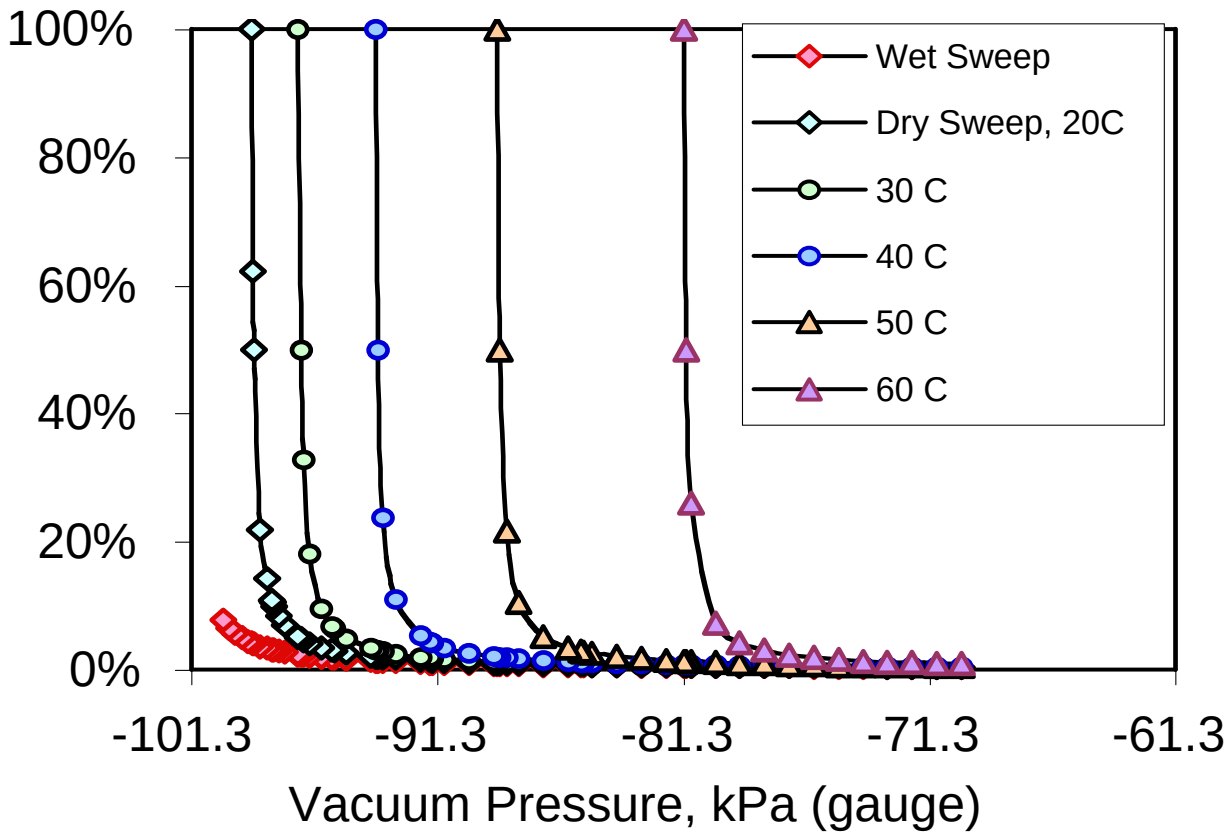
ENZYME STABILITY



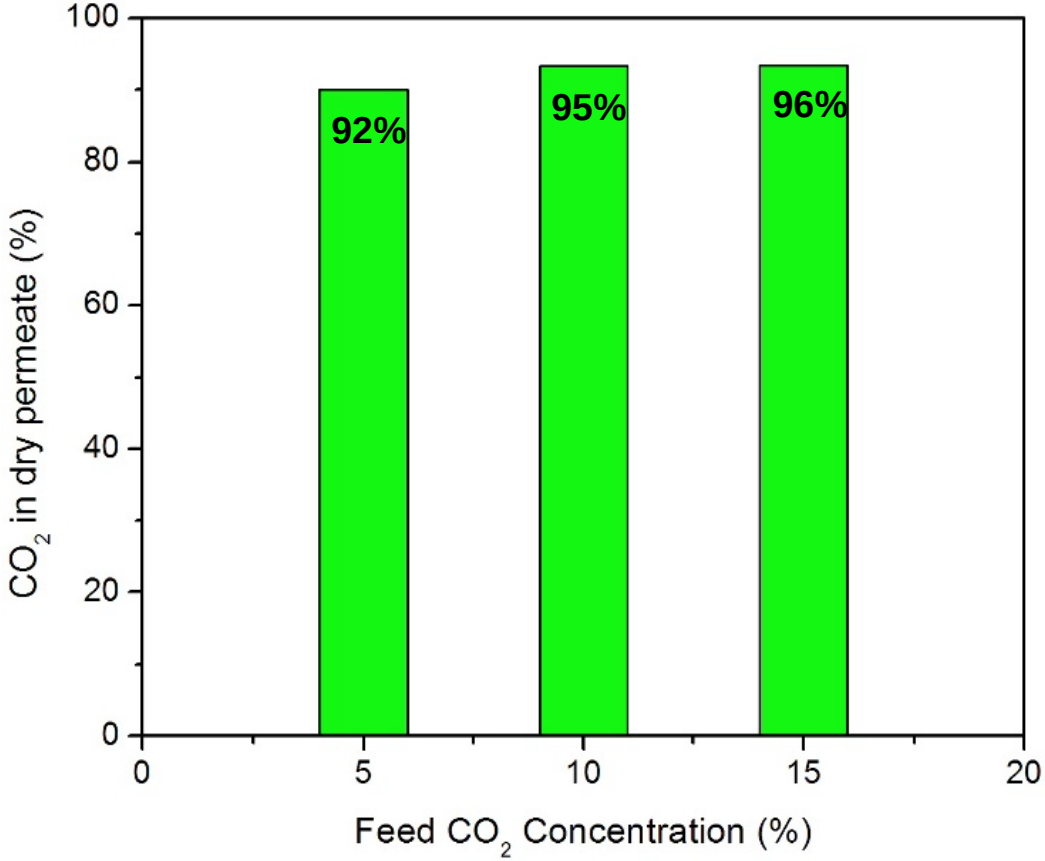
Stability and Longevity of CA/HFCLM



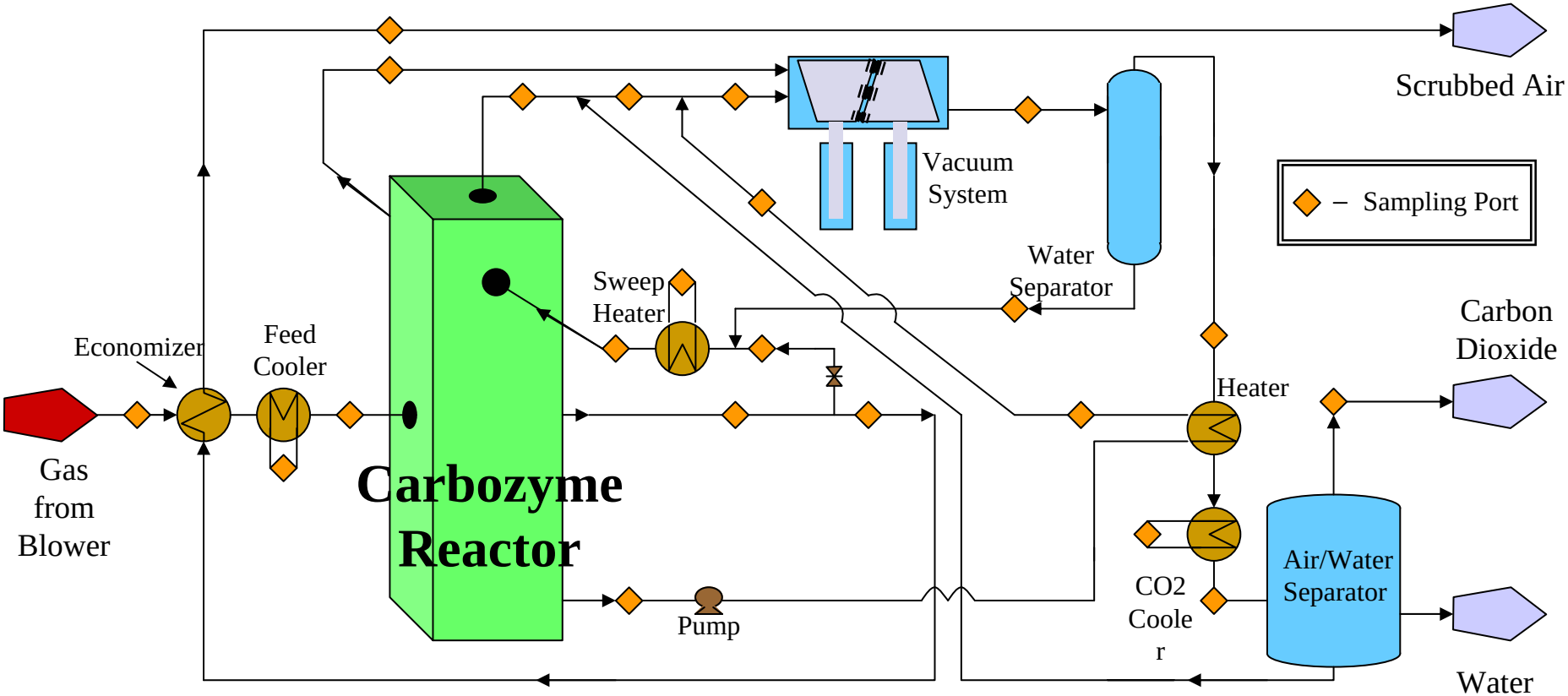
CALCULATED CO_2 CONCENTRATION DRY SWEEP GAS *vs.* VACUUM PRESSURE



FLUE GAS CO_2 CAPTURE

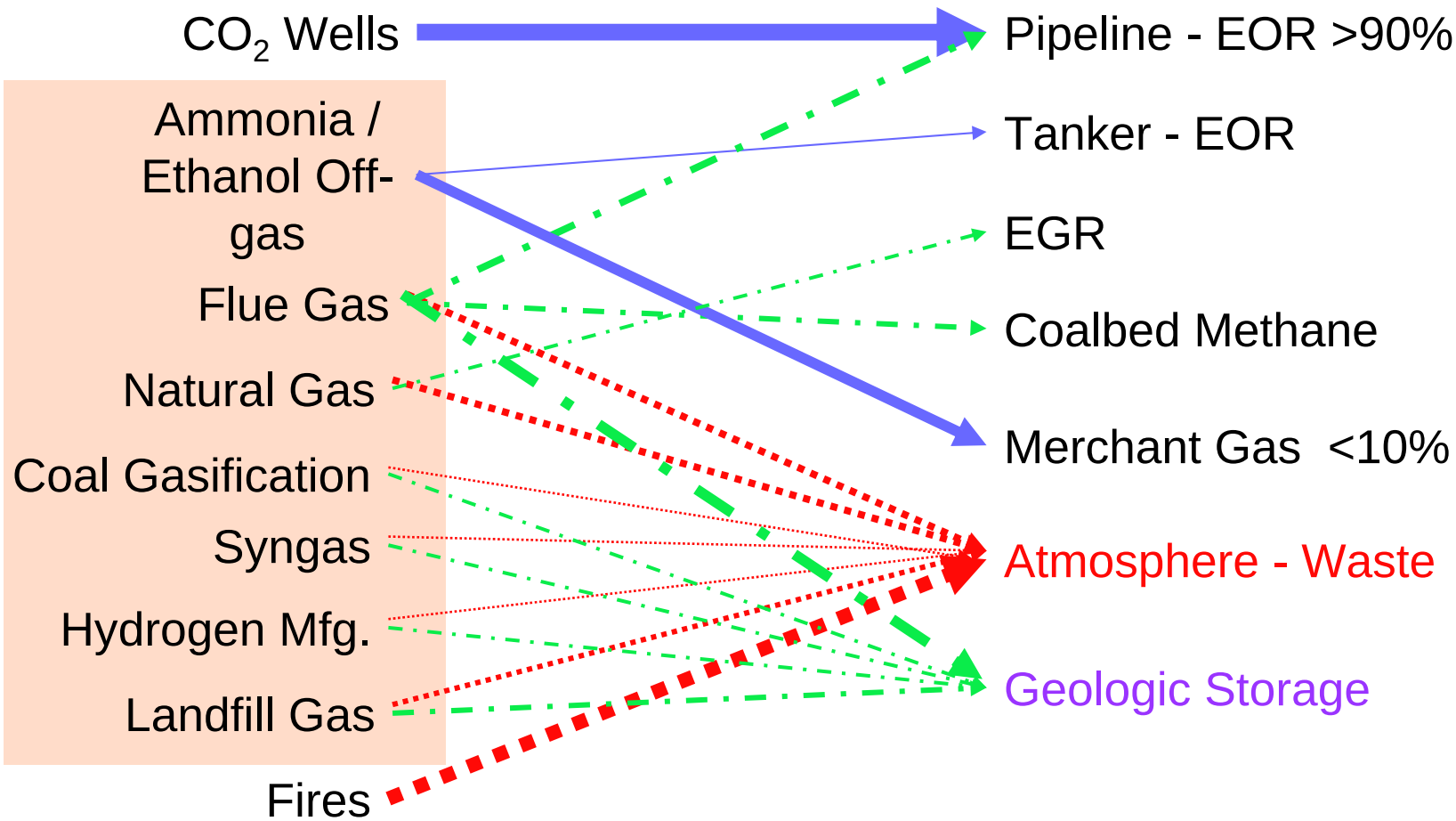


Carbozyme Process Engineering Design



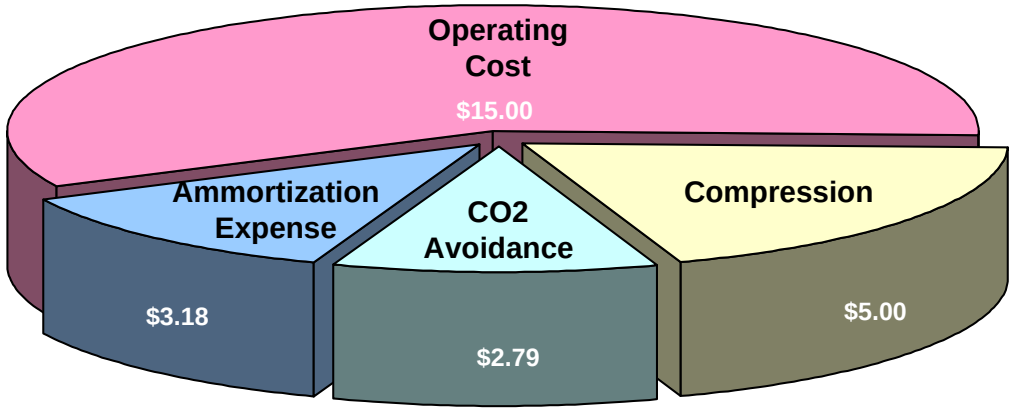
SOURCES and USES of CO_2

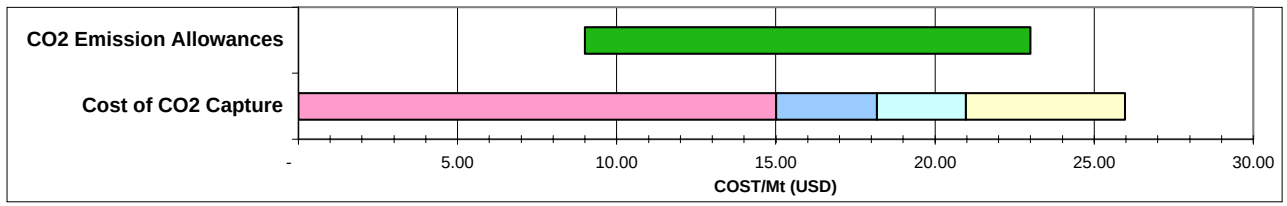
Tomorrow

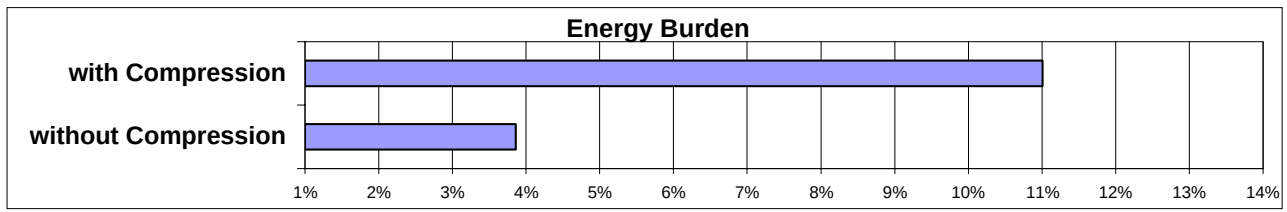


Economic Analysis

300MW Coal Burning Power Plant







Conclusions

- We examined facilitated CO_2 transport across a liquid membrane containing the biocatalyst CA. The performance of this facilitator was illustrated in terms of the CO_2 permeance and $CO_2/N_2/O_2$ selectivity.
- **Liquid membranes made of CA and alkaline carbonate gave very high permeance coupled with high selectivity and were stable for long periods of time.**
- Next generation CA-based transport is expected to increase permeance as much as 10X.

Acknowledgements

- NASA
- DOE