A Carbon Molecular Sieve Membrane-Based Reactive Separation Process for Pre-Combustion CO_2 Capture

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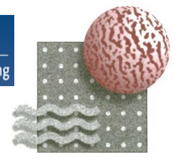
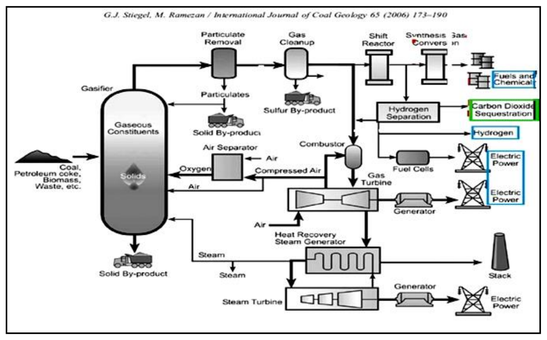
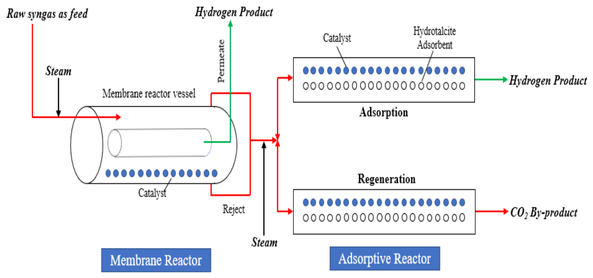


Technology Background

Adsorption-enhanced WGS membrane reactor (MR-AR) process for pre-combustion CO_2 capture



Conventional IGCC Power Plant

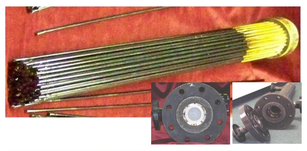


Advantages

- No syngas pretreatment required
- Improved WGS efficiency
- Significantly reduced catalyst weight usage requirements
- Efficient H_2 production, and superior CO_2 recovery and purity

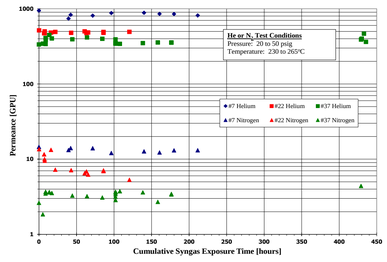
Key Technology Components

Carbon Molecular Sieve (CMS) Membranes

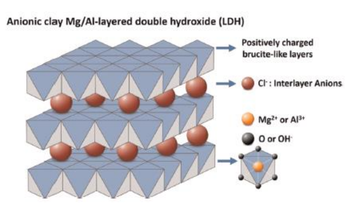


Field-tested at NCCC

Part ID	He [GPU]	N_2 [GPU]	H_2 [GPU]	CO_2 [GPU]	H_2/N_2 [-]	H_2/CO_2 [-]
HMR-61	578	2.5	550	1.0	219	558
HMR-67	450	1.6	581	2.8	354	211
HMR-68	591	3.0	675	2.7	227	248
MR-70	445	1.5	502	0.7	344	738
HMR-72	500	1.7	602	2.5	359	246
HMR-104	542	1.5	540	2.0	361	270



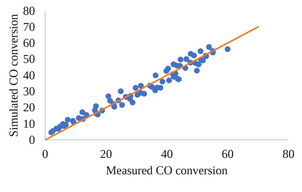
Hydrotalcite (HTC) Adsorbent



High-Pressure Adsorption Isotherm at 250°C

Co-Mo/Al_2O_3 Sour-Shift Catalyst

Reaction rate data generated and global kinetics model developed



$$-r_{CO} = A\, e^{\frac{-E}{RT}} p_{CO}^{a} p_{H_2O}^{b} p_{CO_2}^{c} p_{H_2}^{d} (1-\beta)$$

$$\beta = \frac{1}{K_{eq}} \frac{(P_{CO_2} \cdot P_{H_2})}{(P_{CO} \cdot P_{H_2O})} \qquad K_{eq} = \exp\left(\frac{4577.8}{T} - 4.33\right)$$

A[mol/(atm$^{(a+b+c+d)}$ · h · g)]	18957
E [J/mol]	58074
a	4
b	-1.46
c	0.13
d	-1.44

Root-Mean-Square Deviation (RMSD)	
Direct oxidation	3.38
Associative	5.12
Formate intermediate	8.04
Empirical model	3.32

Acknowledgement

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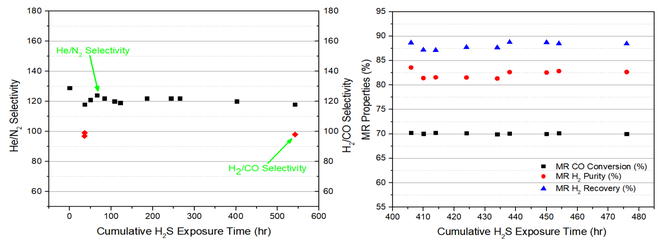
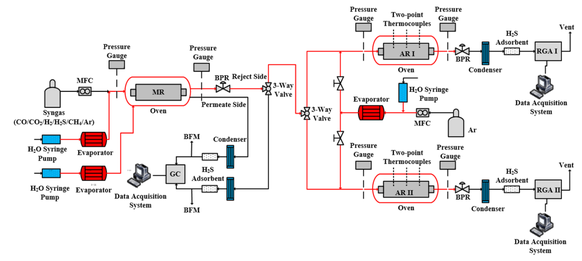
Lab-Scale Experiments

Membrane Reactor Studies

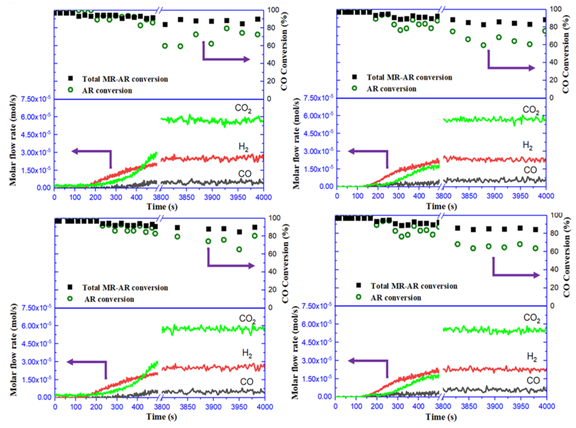


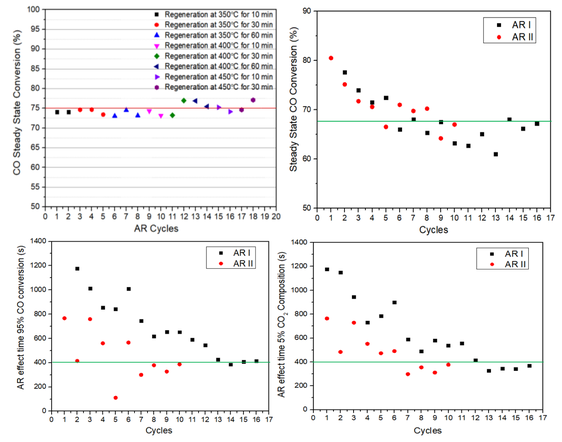
Lab-Scale Experimental Set-Up



The CMSM employed demonstrated robust and stable performance during the long-term run (>500 hr at T=250°C and P=25 bar)

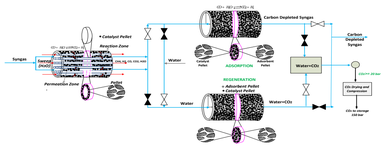
AR Studies

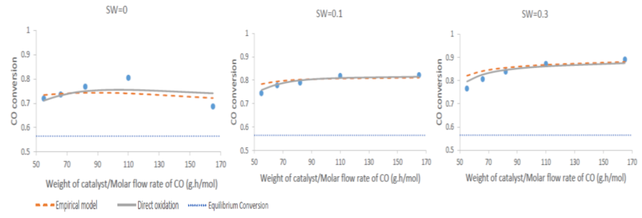




CO in the AR and total MR-AR conversion, and species molar flow rates. (Left Top) AR I, first cycle, (Right Top) AR II, first cycle, (Left Bottom) AR I, second cycle, (Right Bottom) AR II, second cycle. Temp.=250 °C, pressure=25 bar, H_2O/CO ratio=2.8, W/F_{CO}=55 g-h/mol.

Multi-Scale MR-AR Model for Process Scale-Up

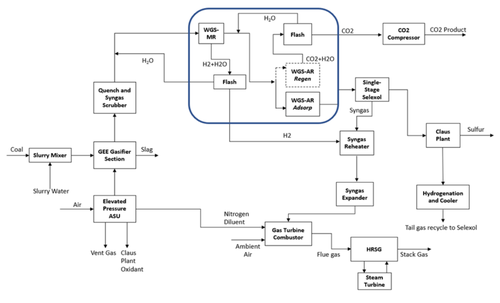




Experimental conversion for the MR with different sweep ratios and the corresponding MR model fits using both the empirical and microkinetic models. (300 °C, feed pressure of 15 bar, CMS#1)

The MR-AR model accounts for mass/energy balances in the catalyst, sorbent and the reactor fluid phases. The Dusty-Gas-Model is used to describe membrane transport. It describes well the laboratory data without resorting to adjustable parameters. It is used in the TEA calculations.

Preliminary TEA - MR-AR IGCC Process Scheme



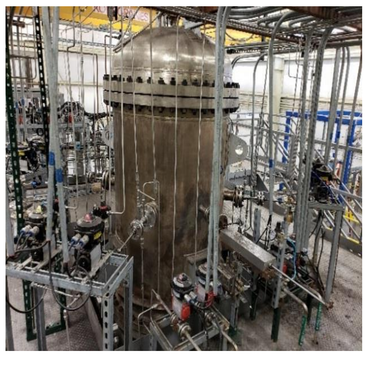
Designs	Net Power Production (MWe)	CO_2 Capture (%)
Shell IGCC w/o CCS – 1-Stage Selexol	622	0
Shell IGCC w/ CCS – 2-Stage Selexol	543	90
MR-AR IGCC Plant	593	92

	% CO Conversion	% H_2 Recovery	% CO_2 Purity	% CO_2 Recovery
Target	>95	>90	>95	>90
MR-AR Realization	99%	99	99	92

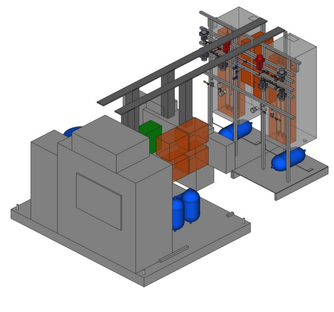
	Capital Cost ($/1000)	Variable Operating Cost ($)	Net Power (MWe)	N_2 Product (ton/h)	COE (No N_2 sale/ N_2 Sale) ($/MWh)	% COE reduction vs Baseline (No N_2 sale/ N_2 Sale)
IGCC CCS	$1,840,115	$46,580,032	543	0	135.4	0
MR-AR Realization	$1,539,820	$47,672,487	593	619	113.1 / 86.3	16.4% / 36%

	Net Power (MWe)	COE (No N_2 sale/ N_2 Sale) ($/MWh)	CO_2 Captured Cost (No N_2 sale/ N_2 Sale) ($/tonne)
IGCC CCS	543	135.4	63.2
MR-AR Realization	593	113.1 / 86.3	39.3 / 5.1

Field-Scale Study of the MR-AR Process



Uky-CAER Gasifier



MR-AR Skid