

REGENLOOP Project Team:



National Laboratory  
of the Rockies



COLORADO SCHOOL OF  
**MINES**

# Modular Cost-Effective Upgrading of Biogas to Liquid Hydrocarbons via Chemical Looping

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tcbi**mass**<sup>+</sup>2026

Photo from Adobe Stock 256750697

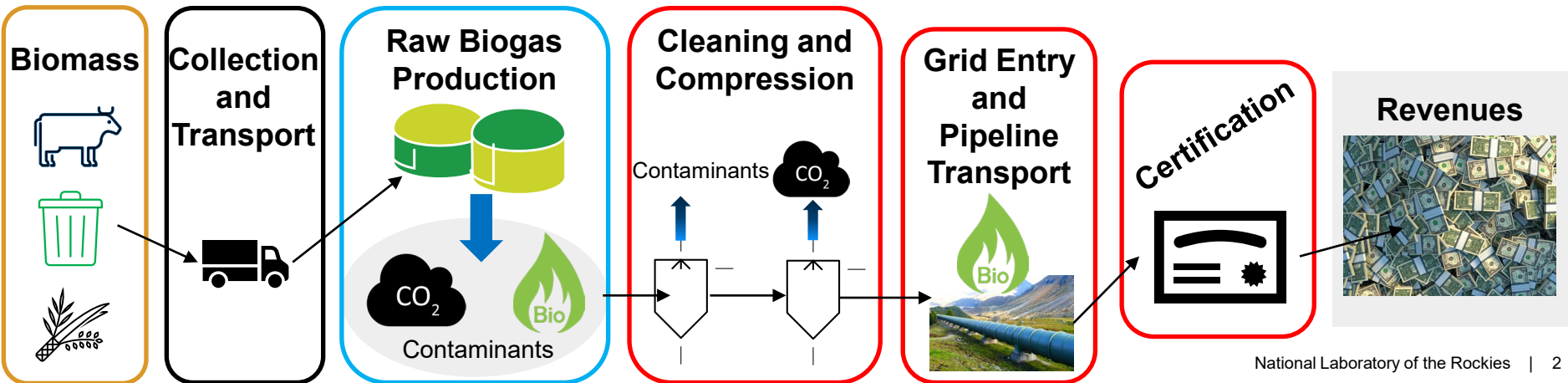
# Distributed Methane Resources: Challenges and Opportunities

- US biogas market potential: 140 BCM (\$15-250B)
- Pipeline quality biogas (biomethane) is hard to scale due to costly production chain -> 5-10x natural gas price
- Many biogas projects struggle economically (<6% utilization)

Data from: (1) IEA, Outlook for Biogas and Biomethane, 2025; (2) DOE, 2023 Billion-Ton Report

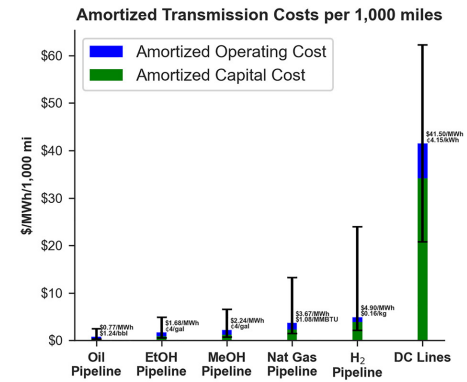


Source:  
<https://www.hydrogenfuelnews.com/de-sulfurization-biogas-plants/8562263/>



# Distributed Methane Resources: Challenges and Opportunities

- US biogas market potential: 140 BCM (\$15-250B)
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Source: [1]

Data from: (1) IEA, Outlook for Biogas and Biomethane, 2025; (2) DOE, 2023 Billion-Ton Report

## Liquid Fuels and Chemicals:

- High energy density: aviation/maritime requirement
- Easy to store and transport
- ~\$800B of existing infrastructure
- Improve U.S. energy resiliency

**BUT**

## Distributed Chemicals Production:

- Unfavorable scale for chemical plants: large capital intensity
- Complex process chains: high OPEX
- Dependency on incentives for commercial viability

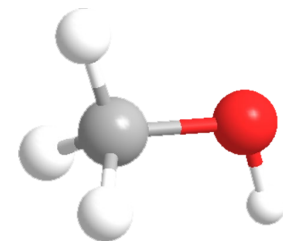
[1] D. DeSantis, B. D. James, C. Houchins, G. Saur, and M. Lyubovsky, "Cost of long-distance energy transmission by different carriers," *iScience*, vol. 24, no. 12, p. 103495, Dec. 2021, doi: [10.1016/j.isci.2021.103495](https://doi.org/10.1016/j.isci.2021.103495).

[2] <https://www.precedenceresearch.com/oil-and-gas-infrastructure-market>

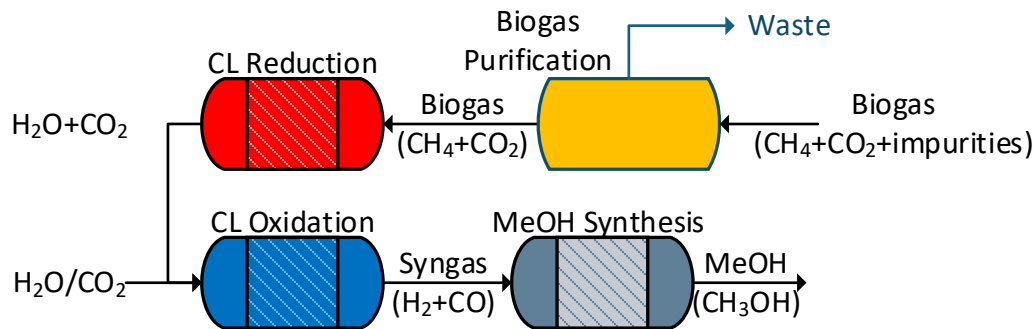
# Our Proposed Idea

## Chemical looping reforming of biogas to methanol (MeOH)

Methanol is a versatile chemical commodity and potential maritime fuel



Source: American Chemical Society  
<https://www.acs.org/molecule-of-the-week/archive/m/methanol.html>



- Reduced raw biogas purification (no need to remove CO<sub>2</sub>)
- Reforming without excess oxidizer (less water, CO<sub>2</sub> utilization in cycle)
- Reduced syngas conditioning (high oxidizers conversion)
- Feedstock flexibility: syngas composition can be maintained via make-up H<sub>2</sub>O/CO<sub>2</sub> ratio

# The REGENLOOP Process: Chemical Regenerator Effect

Patent pending

## Chemical looping reforming of methane (CLRM)

**Reduction (endothermic,  $\Delta_r H < 0$ ):**



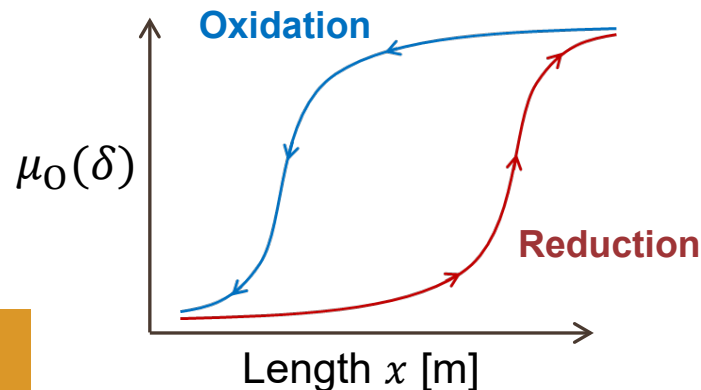
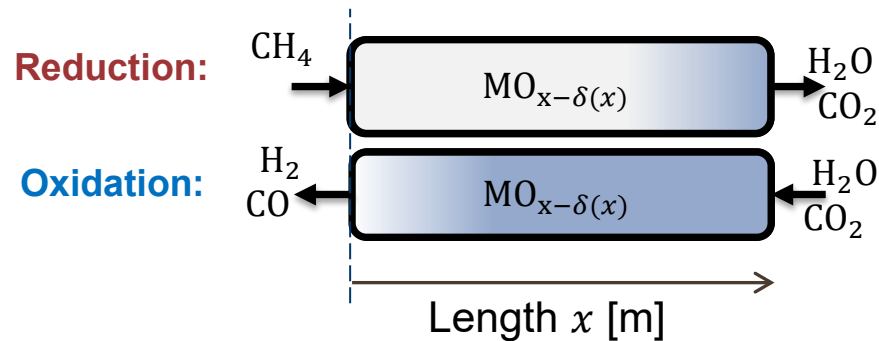
**Oxidation (exothermic,  $\Delta_r H < 0$ ):**



Chemical potential gradient (nonstoichiometric oxides)  $\rightarrow$  thermodynamic driving force

Experimental demonstrations for WGS, RWGS, and reforming exhibit higher  $X$  than catalytic processes

## Countercurrent Regenerative Reactor



Extent of reduction:  
 $\Delta\delta = \delta_{\text{red}} - \delta_{\text{ox}}$

B. Bulfin, et al., *Chem. Eng. J.* **2023**, 461.

I.S. Metcalfe, et al., *Nat. Chem.* **2019**, 11 638–643.

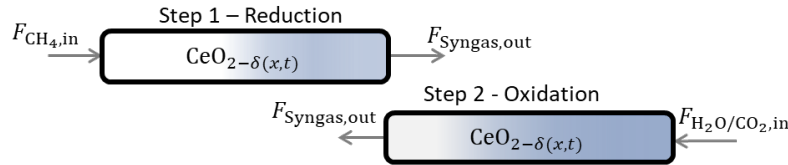
Y. Yan, et al., *Journal of CO<sub>2</sub> Utilization*, **2024**, 88 102935.

B. Bulfin, et al., *Chem. Eng. J.* **2024**, 488.

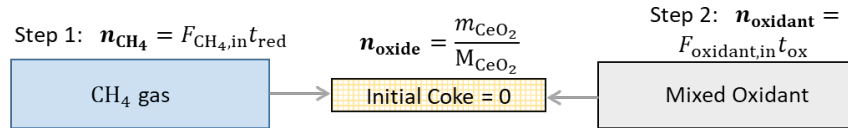
# Reactor Modeling Framework: Discretized Batch Reactor Model

- Infinitely fast kinetics (thermodynamic limit)
- Coking mechanism can be turned on/off
- Solid and gas elements are discretized
- Moving gas elements through reactor
- Iterative solution: fixing  $n_{\text{CH}_4}$  and  $n_{\text{oxidizer}}$  and sweeping over  $n_{\text{oxide}}$  with target  $X$

a) Temporal and spatial problem

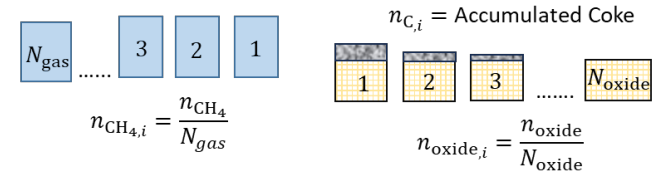


b) Parametrized to  $n_{\text{CH}_4}$ ,  $n_{\text{oxidant}}$ , and  $n_{\text{oxide}}$

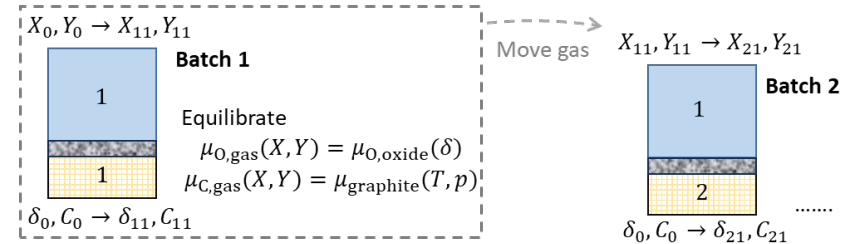


**Note:** results are with no coking mechanism due to coking kinetic limitations

c) Discretization of step 1



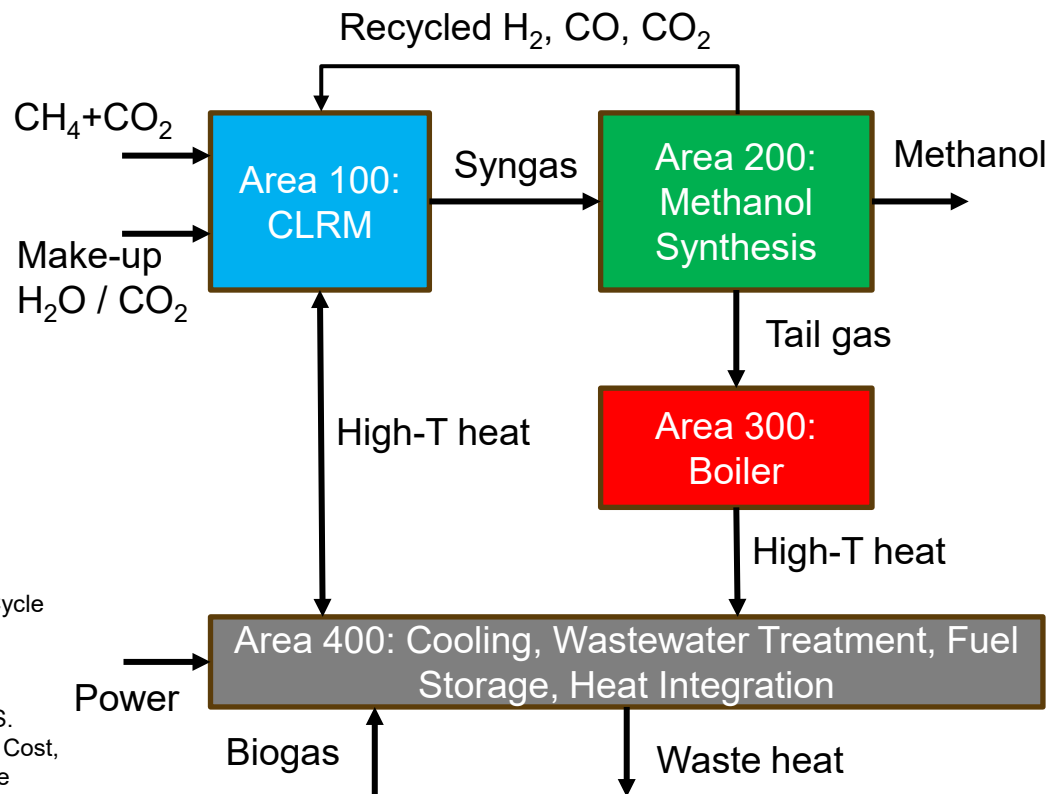
d) Sequential batch dual equilibrium calculations



# MeOH Plant General Design

Plant size: 200 tpd

- CLRM reactors: inverse flow reactor design with internal thermocline
- CLRM consist of reactor pairs
- GTL, boiler, and utilities based on [1,2]
- Replaced original syngas production (via RWGS) with CLRM
- Integrated recycle loops between CLRM and GTL sections
- Extra high-T process heat from biogas



[1] G. Zang, P. Sun, A. Elgowainy, M. Wang, Technoeconomic and Life Cycle Analysis of Synthetic Methanol Production from Hydrogen and Industrial Byproduct  $\text{CO}_2$ , Environ. Sci. Technol. 55 (2021) 5248–5257.

<https://doi.org/10.1021/acs.est.0c08237>.

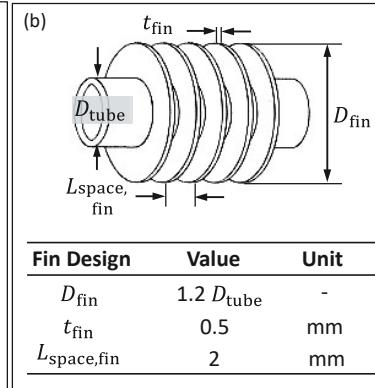
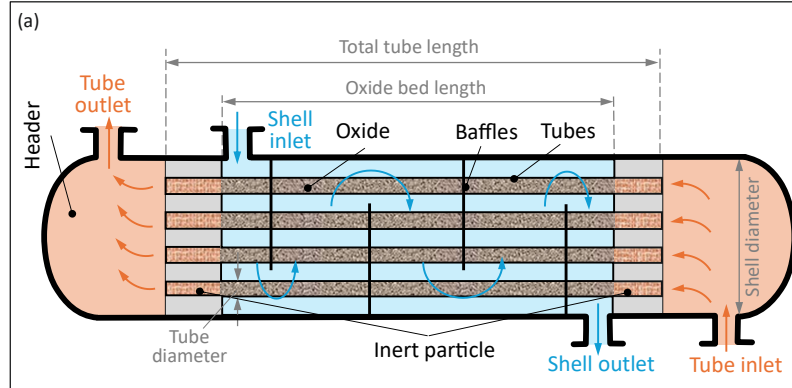
[2] G. Zang, P. Sun, E. Yoo, A. Elgowainy, A. Bafana, U. Lee, M. Wang, S. Supekar, Synthetic Methanol/Fischer–Tropsch Fuel Production Capacity, Cost, and Carbon Intensity Utilizing  $\text{CO}_2$  from Industrial and Power Plants in the United States, Environ. Sci. Technol. 55 (2021) 7595–7604.

<https://doi.org/10.1021/acs.est.0c08674>.

# Main Reactor Design Parameters

- Multi-tubular reactor driven by high-T HTF (flue gas)
- Solving single reactor tube -> number of required tubes to meet syngas demand
- Kern method to size the reactor (limited by  $D_{\text{shell}}$ , increase no. of reactors if needed)
- Heat of reduction and oxidation solved from thermal design
- Tuning flow rates and switch times to maximize yield and conversion

| Reactor Design                           | Value          | Unit   |
|------------------------------------------|----------------|--------|
| Redox material                           | CeZr15         | -      |
| Redox $T/p$                              | 850/15         | °C/bar |
| Tube size                                | NPS 3 x Sch 80 |        |
| Oxide bed length, $L_{\text{oxide}}$     | 15             | m      |
| Inner shell diameter, $D_{\text{shell}}$ | 2.687          | m      |
| Tubes per reactor, $N_{\text{tube}}$     | 378            | -      |



# Plant and Financial Parameters

- GTL capital cost from [1] (scaled)
- CLRM capital cost from [1-3]
- DCF analysis using ProFAST [4]
- Analysis \$ year: 2024

| Parameter                                 | Value     |
|-------------------------------------------|-----------|
| CH <sub>4</sub> /CO <sub>2</sub> fraction | 80% / 20% |
| Oxidizer to reductant ratio               | 4:1       |
| Oxide to reductant ratio                  | 57        |
| Cycle time (symmetric)                    | 4 minutes |

| Technoeconomic Analysis Parameter | Value          | Unit   |
|-----------------------------------|----------------|--------|
| Annual capacity factor            | 90%            |        |
| <b>Nameplate capacity</b>         | <b>200 t/d</b> |        |
| Construction period               | 3              | years  |
| Debt-to-equity ratio              | 62.6           | %      |
| Inflation rate                    | 2.5            | %/year |
| Plant lifetime                    | 30             | years  |
| Nominal ROI                       | 13             | %      |
| Nominal debt interest rate        | 7              | %      |
| Type of debt                      | Revolving debt |        |
| Cash on hand                      | 3              | months |
| Effective tax rate                | 25.74          | %      |
| Capital gain tax                  | 15             | %      |
| Depreciation schedule             | 7-years MARCS  |        |

[1] G. Zang, P. Sun, A. Elgowainy, M. Wang, Technoeconomic and Life Cycle Analysis of Synthetic Methanol Production from Hydrogen and Industrial Byproduct CO<sub>2</sub>, Environ. Sci. Technol. 55 (2021) 5248–5257. <https://doi.org/10.1021/acs.est.0c08237>.

[2] G. Towler, R. Sinnott, Chemical Engineering Design, 3rd ed., Elsevier, 2022. <https://doi.org/10.1016/C2019-0-02025-0>.

[3] R. Turton, J.A. Shaeiwitz, D. Bhattacharyya, W.B. Whiting, Analysis, synthesis, and design of chemical processes, Fifth edition, Prentice Hall, Boston Columbus Indianapolis New York San Francisco Amsterdam, 2018.

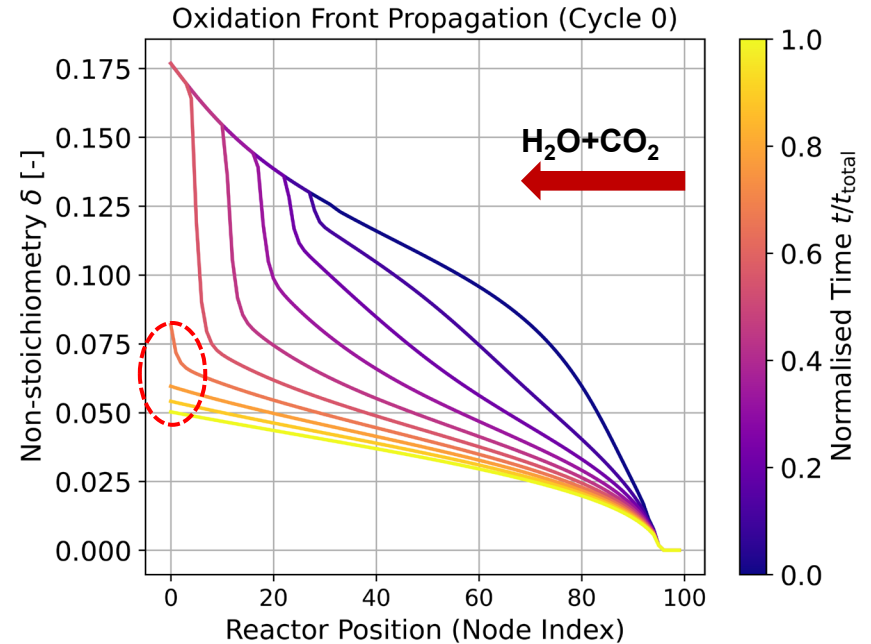
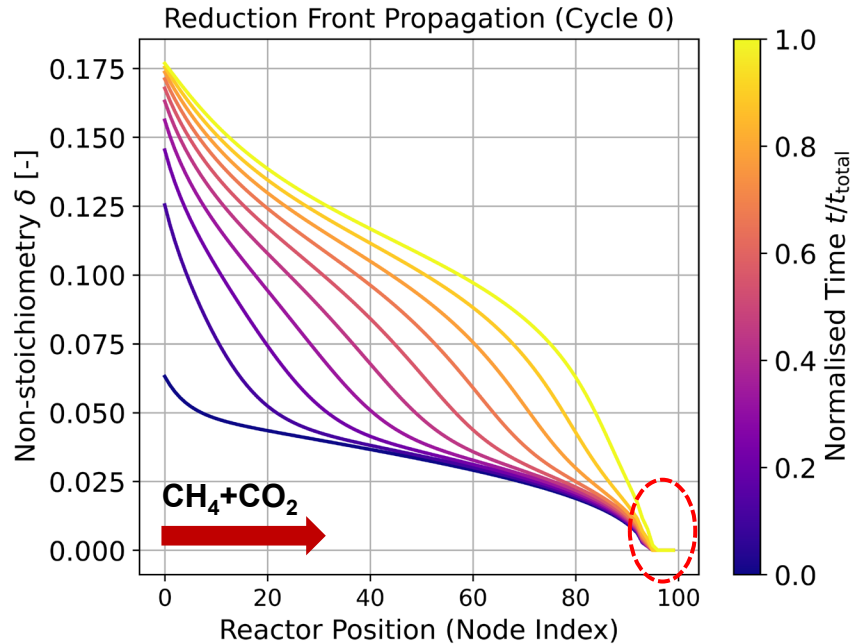
[4] J. Kee, M. (Misho) Penev, ProFAST (Production Financial Analysis Scenario Tool) [SWR-23-88], (2023). <https://doi.org/10.11578/DC.20231211.1>.

# Results

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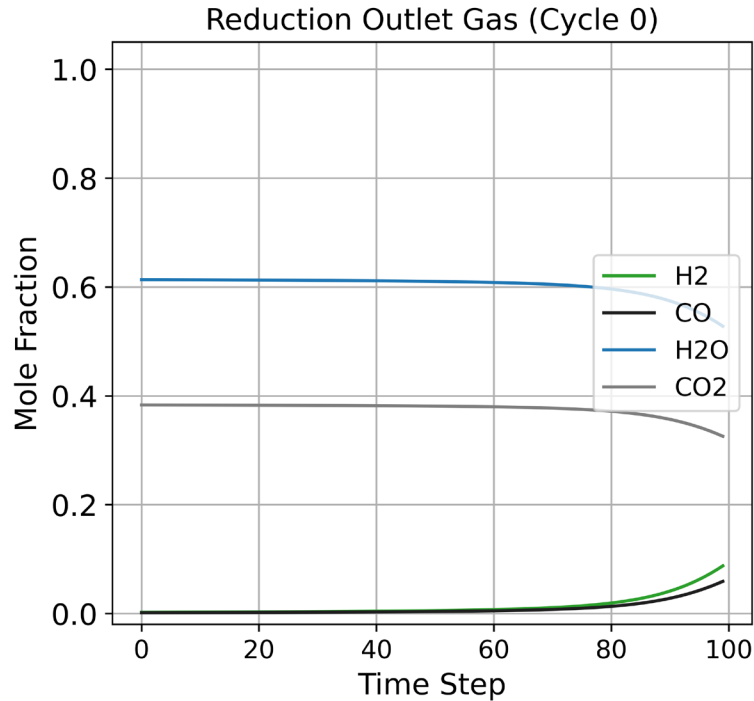
# Reactor Sizing and Performance

$\delta$ =non-stoichiometry  
extent (O in solid)



- Model solved non-dimensionally for 1 mol of reductant ( $\text{CH}_4 + \text{CO}_2$ )
- Iterative solution until quasi steady-state

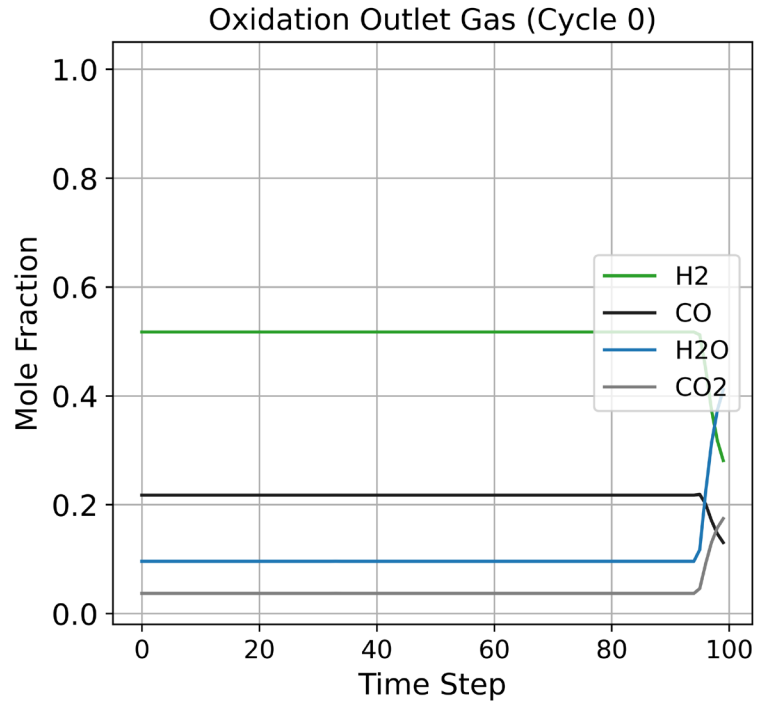
# Reactor Sizing and Performance



Reduction:

$$X_{\text{CH}_4} = 1$$

$$S_{\text{H}_2\text{O}} = 0.978$$



Oxidation:

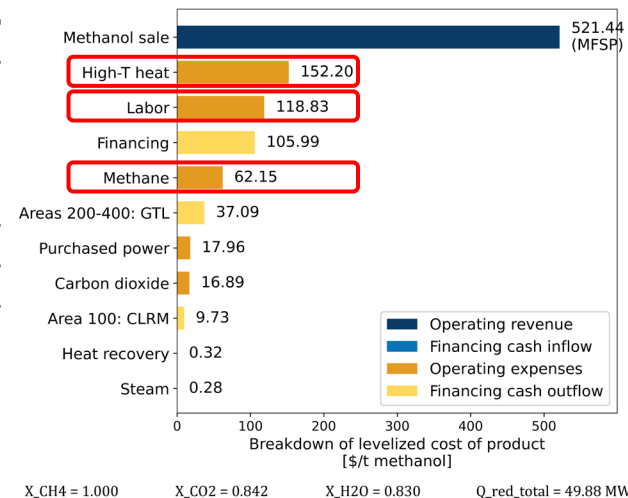
$$X_{\text{CO}_2, \text{ox}} = 0.842$$

$$X_{\text{H}_2\text{O}, \text{ox}} = 0.83$$

# Baseline Case Financial Results

**LCOP of MeOH = \$521.44/t**

| Process Area                                     | Purchased Cost      | Installed Cost      |
|--------------------------------------------------|---------------------|---------------------|
| Area 100: REGENLOOP                              | \$5,925,439         | \$12,775,003        |
| Area 200: MeOH Synthesis                         | \$8,786,462         | \$13,282,136        |
| Area 300: Boiler and Thermal Management          | \$4,495,673         | \$8,092,212         |
| Area 400: Cooling, Wastewater Treatment, Storage | \$9,326,591         | \$19,375,265        |
| <b>Total Direct Cost</b>                         | <b>\$28,534,165</b> | <b>\$53,524,614</b> |
| Indirect Costs                                   | % of TDC            | Cost                |
| Site preparation                                 | 1.84%               | \$984,250           |
| Design and engineering                           | 12.92%              | \$6,914,153         |
| Project contingency                              | 15.80%              | \$8,454,979         |
| Permitting, start-up, other costs                | 13.79%              | \$7,381,878         |
| Offsites                                         | 4.77%               | \$2,555,001         |
| <b>Total Indirect Cost</b>                       | <b>49.12%</b>       | <b>\$26,290,260</b> |
| <b>Fixed Capital Investment Excluding Land</b>   |                     | <b>\$79,814,875</b> |
| Land                                             |                     | \$250,126           |
| <b>Total Capital Investment (TCI)</b>            |                     | <b>\$80,065,001</b> |
| <b>Capital Intensity</b>                         | <b>\$(t/y)</b>      | <b>1219</b>         |



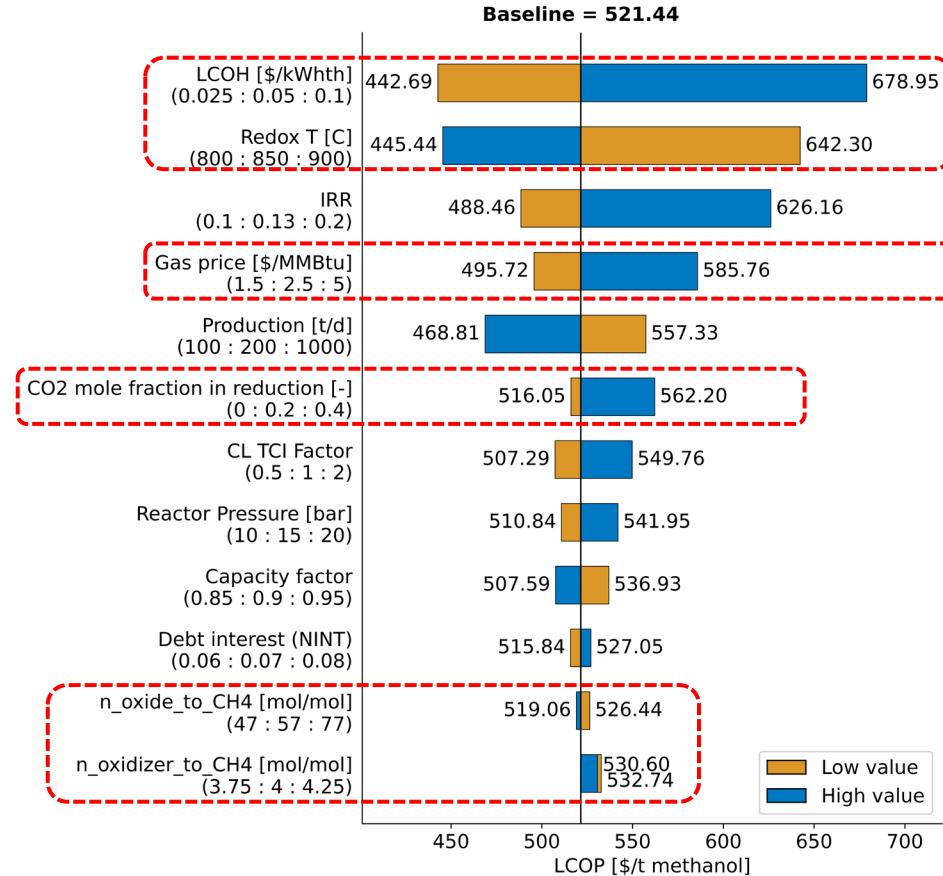
**OpEx dominated cost stack**

# Sensitivity Analysis

## Main cost drivers and insights:

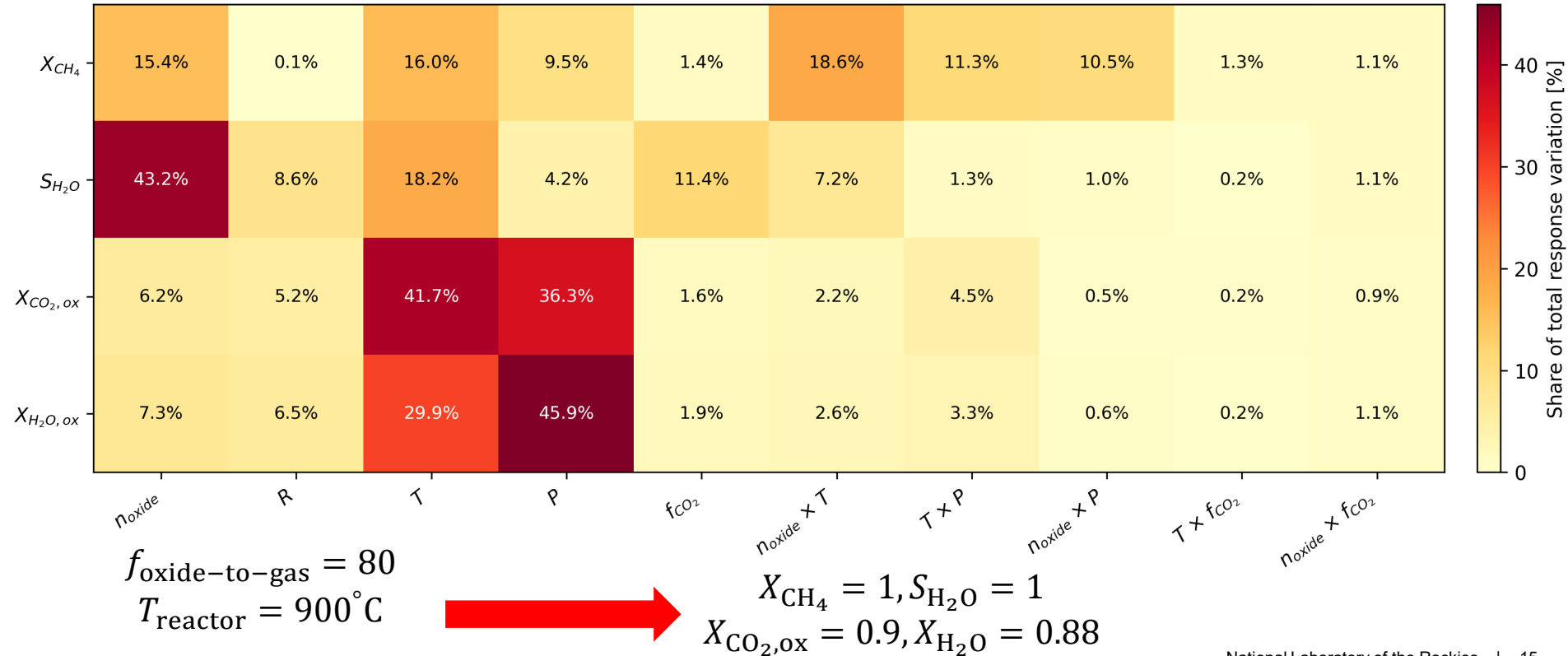
- High-quality heat and redox temperature
- Feedstock price and quality
- Ratios of oxide to  $\text{CH}_4$  and oxidizer to  $\text{CH}_4$  has an optimal value, dependent on other parameters

*Room for process optimization*



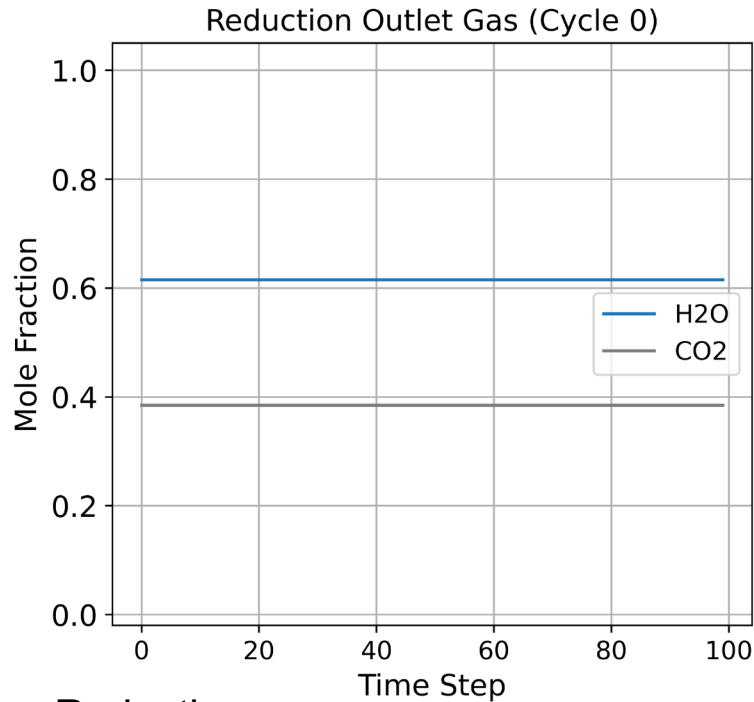
# CLRM Response Surface

CL tuning sensitivity: main effects and strongest interactions



# Optimized CLRM: Reactor Sizing and Performance

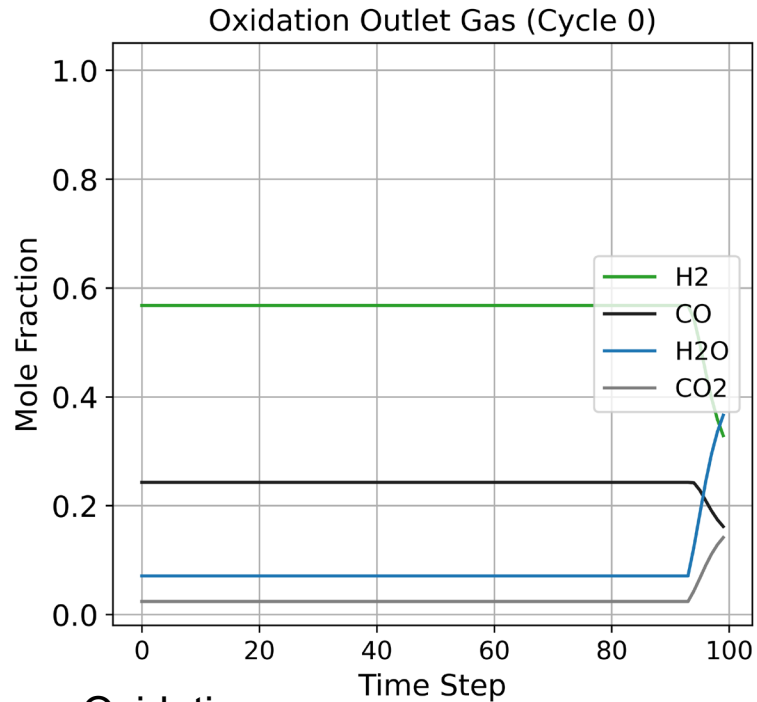
$$f_{\text{oxide-to-gas}} = 80 \text{ mol}$$
$$T_{\text{reactor}} = 900^{\circ}\text{C}$$



Reduction:

$$X_{\text{CH}_4} = 1$$

$$S_{\text{H}_2\text{O}} = 1$$



Oxidation:

$$X_{\text{CO}_2, \text{ox}} = 0.9$$

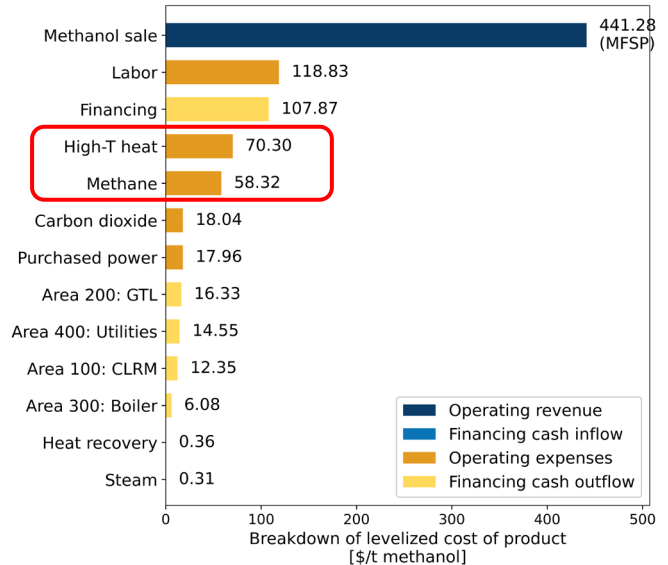
$$X_{\text{H}_2\text{O}, \text{ox}} = 0.88$$

# Optimized CLRM: TEA Results

$$f_{\text{oxide-to-gas}} = 80 \text{ mol}$$

$$T_{\text{reactor}} = 900^{\circ}\text{C}$$

## Baseline: Heat recovery only



$X_{\text{CH}_4} = 1.000$     $X_{\text{CO}_2} = 0.851$     $X_{\text{H}_2\text{O}} = 0.825$     $Q_{\text{red\_total}} = 40.69 \text{ MW}$

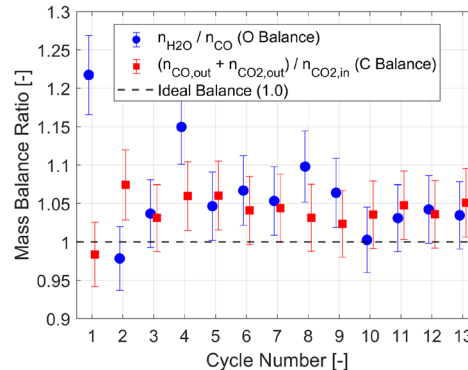
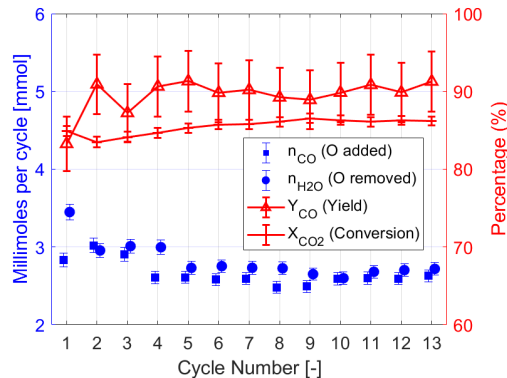
$$X_{\text{CH}_4} = 1, S_{\text{H}_2\text{O}} = 1$$

$$X_{\text{CO}_2, \text{ox}} = 0.9, X_{\text{H}_2\text{O}, \text{ox}} = 0.88$$

- **LCOP: \$441/t vs \$521/t**
- High  $X$  lowers heat and feedstock
- Increase in CLRM capital due to large reactor (more oxide)
- Decrease in GTL capital due to smaller syngas conditioning

# Experimental Work

- Benchtop reverse flow reactor (~100g OC)
- Running with pure gases ( $\text{CH}_4$ ,  $\text{H}_2$ ,  $\text{CO}_2$ )
- Atmospheric pressure, up to  $950^\circ\text{C}$
- Unattended operation (long cycling tests)
- Demonstrated on CL-RWGS
- 100x pilot reactor development in progress



# Summary and Conclusions

- Chemical regenerator-based CLRM can provide process intensification
- Reactor thermal design and management (endothermic/exothermic)
- Competitive MeOH price (~\$441/t) for small plant (200 tpd)
- LCOP most sensitive to feedstock and high-T heat cost, followed by feedstock price and financing parameters

## Future Work

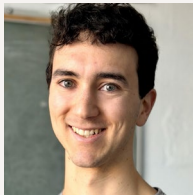
- Experimental demonstration of our CLRM process with varying  $\text{CH}_4:\text{CO}_2$
- Robust optimization of the CLRM to identify best operating regime
- Detailed Aspen Plus modeling of a biogas-based case study
- Integration of the raw biogas production into the plant



**Akbar Laksana**



**Alon Lidor**



**Luke Davenport**



**Rhushikesh  
Ghotkar**



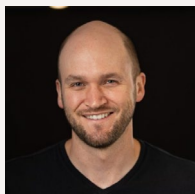
**Janna  
Martinek**



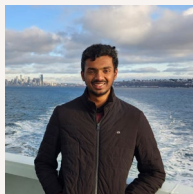
**Geetanjali  
Yadav**



**Brendan Bulfin**



**Kurt Van Allsburg**



**Phalgun  
Malupillai**



**Eva Ciuffetelli**

### **Other project partners:**

- Solar Fuels and Technology Lab (Ga Tech)
- High-Temperature Energy Conversion and Storage (Hi-TECS) Research Group (CSM)

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DOE TM: Gong Zhou (IESO, TES)

# Thank you! Questions?

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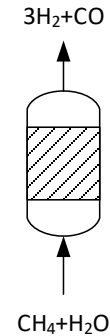
# Extra Slides

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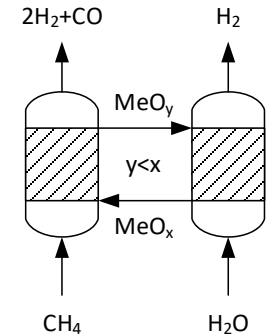
# Chemical Looping in 60 Seconds

- Chemical looping (CL): breaking a single chemical reaction into multiple sub-reactions using chemical intermediates to:
  - Minimize exergy losses
  - Allow enhanced separation of products/pollutants
  - Avoid unwanted side reactions
- Chemical intermediates: mostly solids undergoing reduction-oxidation (redox), calcination-carbonation, or other reactions
- Separation of steps:
  - Spatially (moving solids between reactors, steady state)
  - Temporally (redirecting flow, cycling batch process)
- Example on right: steam methane reforming (SMR)

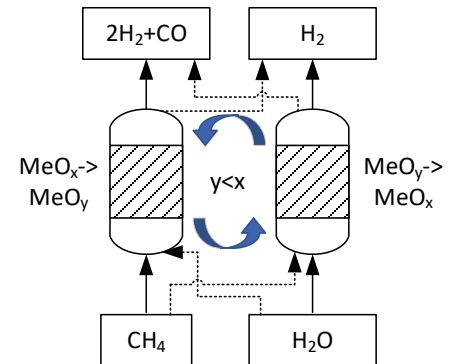
**Catalytic**



**CL with moving oxide**



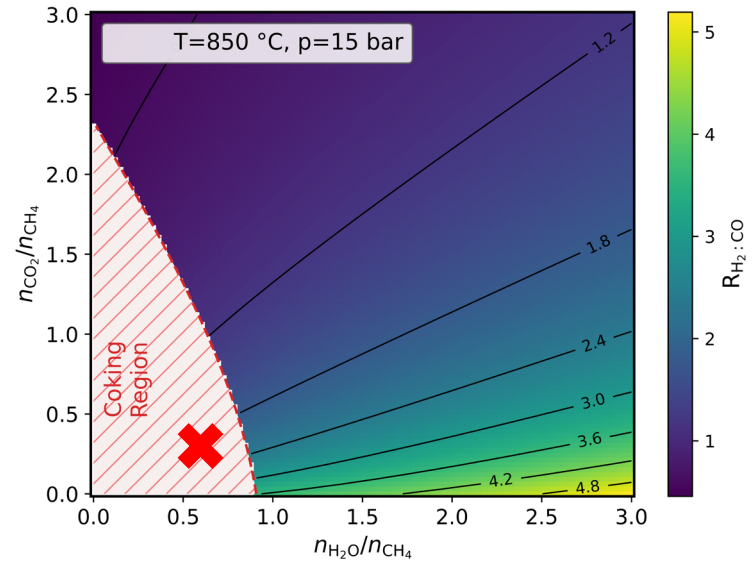
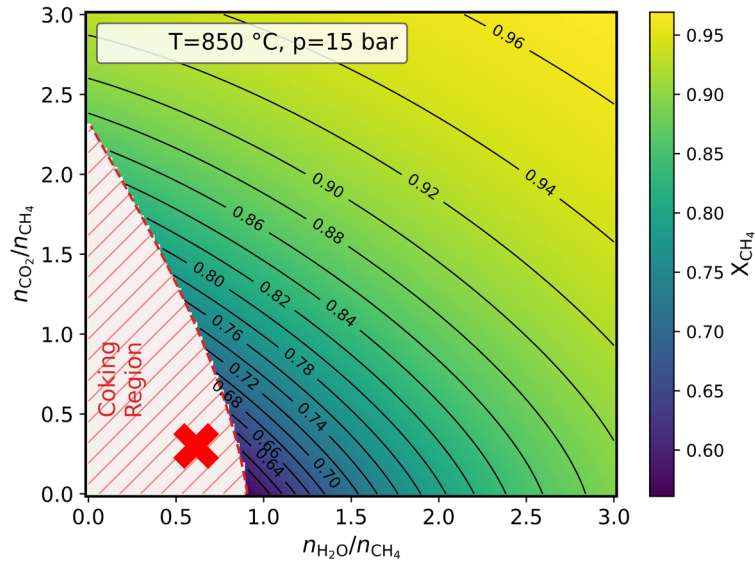
**CL with redirecting flow**



[1] N.M. Nguyen, F. Alobaid, P. Dieringer, B. Epple, Biomass-Based Chemical Looping Gasification: Overview and Recent Developments, Applied Sciences 11 (2021). <https://doi.org/10.3390/app11157069>.

[2] X. Zhao, H. Zhou, V.S. Sikarwar, M. Zhao, A.-H.A. Park, P.S. Fennell, L. Shen, L.-S. Fan, Biomass-based chemical looping technologies: the good, the bad and the future, Energy Environ. Sci. 10 (2017) 1885–1910. <https://doi.org/10.1039/C6EE03718F>.

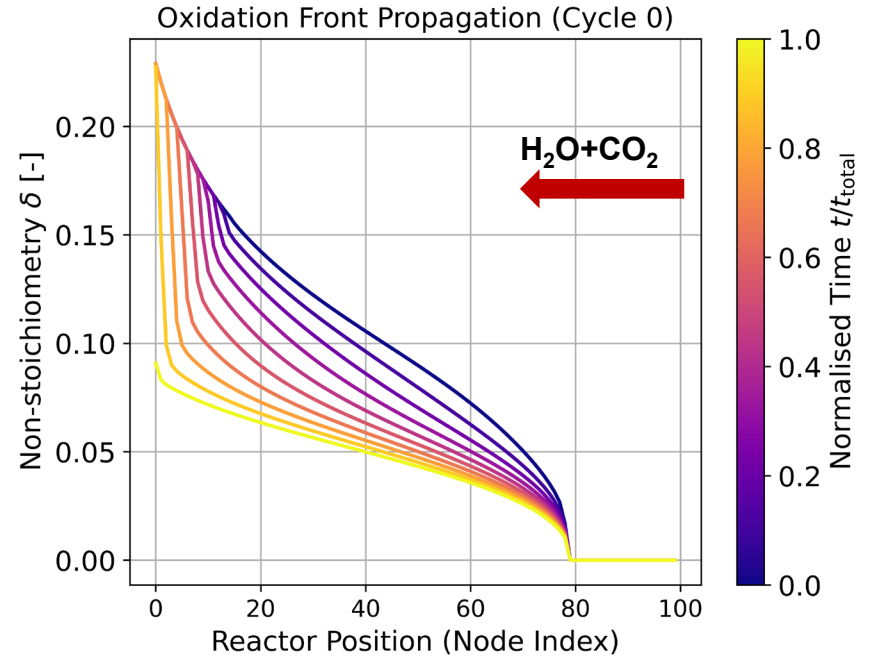
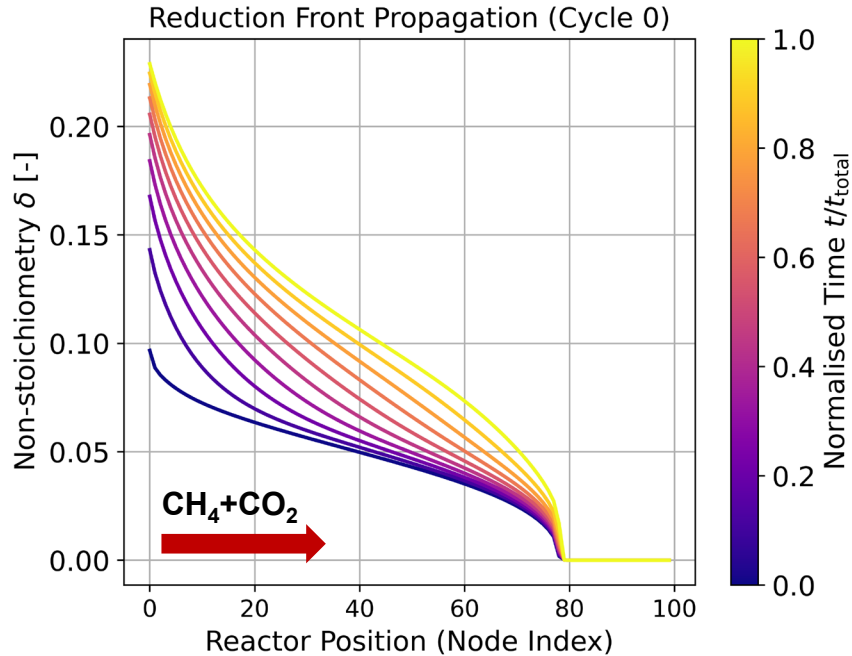
# Comparison with Equilibrium Limit of Catalytic Reforming of Methane



- Reforming without excess oxidizer in regimes prone to coking
- How can we improve  $X_{\text{CO}_2}$  and  $X_{\text{H}_2\text{O}}$  without sacrificing  $X_{\text{CH}_4}$  and  $S_{\text{H}_2\text{O}}$ ?

# Reactor Model for Optimized Case

$\delta$ =non-stoichiometry  
extent (O in solid)



$$\begin{aligned} f_{\text{oxide-to-gas}} &= 80 \\ R_{\text{oxidizer-to-CH}_4} &= 4 \\ T_{\text{reactor}} &= 900^\circ\text{C} \end{aligned}$$



$$\begin{aligned} X_{\text{CH}_4} &= 1, S_{\text{CH}_4} \cong 1 \\ X_{\text{CO}_2, \text{ox}} &= 0.9, X_{\text{H}_2\text{O}} = 0.88 \end{aligned}$$

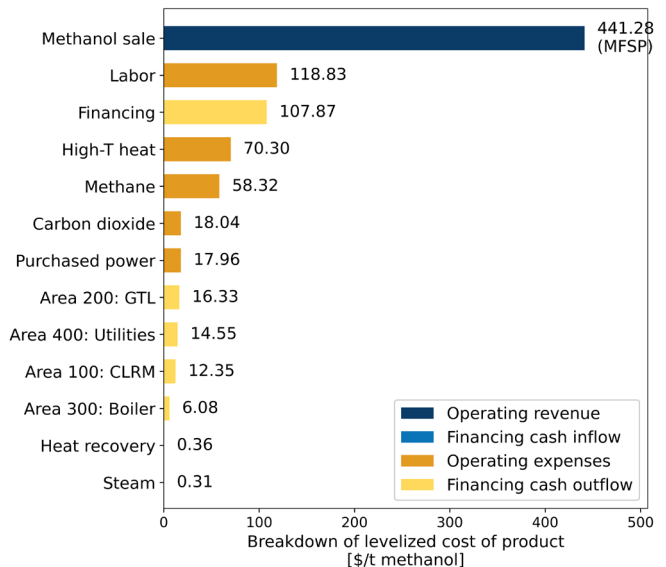
# Process Integration Options: Ideal Case

$$f_{\text{oxide-to-gas}} = 80 \text{ mol}$$

$$R_{\text{oxidizer-to-CH}_4} = 4$$

$$T_{\text{reactor}} = 900^\circ\text{C}$$

## Baseline: Heat recovery only

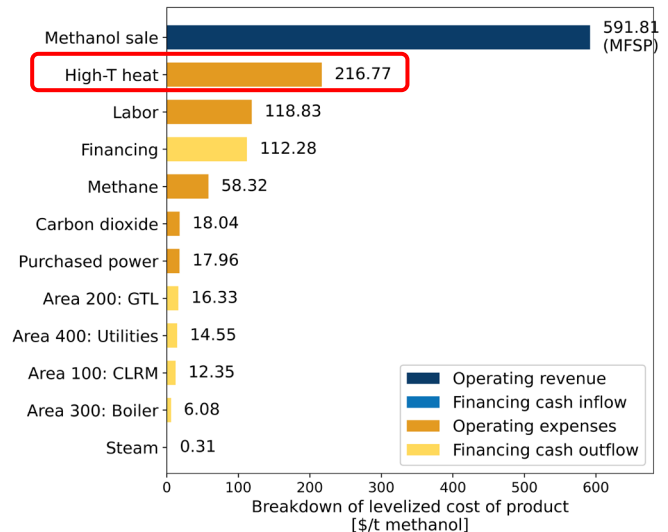


$X_{\text{CH}_4} = 1.000$     $X_{\text{CO}_2} = 0.851$     $X_{\text{H}_2\text{O}} = 0.825$     $Q_{\text{red\_total}} = 40.69 \text{ MW}$

$$X_{\text{CH}_4} = 1, S_{\text{H}_2\text{O}} \cong 1$$

$$X_{\text{CO}_2, \text{ox}} = 0.9, X_{\text{H}_2\text{O}, \text{ox}} = 0.88$$

## No waste heat recovery



$X_{\text{CH}_4} = 1.000$     $X_{\text{CO}_2} = 0.851$     $X_{\text{H}_2\text{O}} = 0.825$     $Q_{\text{red\_total}} = 40.69 \text{ MW}$

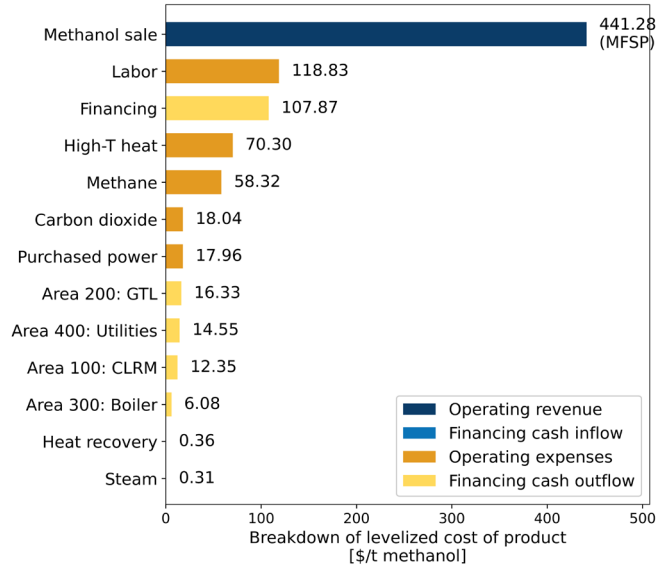
# Process Integration Options: Ideal Case

$$f_{\text{oxide-to-gas}} = 80 \text{ mol}$$

$$R_{\text{oxidizer-to-CH}_4} = 4$$

$$T_{\text{reactor}} = 900^\circ\text{C}$$

## Baseline: Heat recovery only

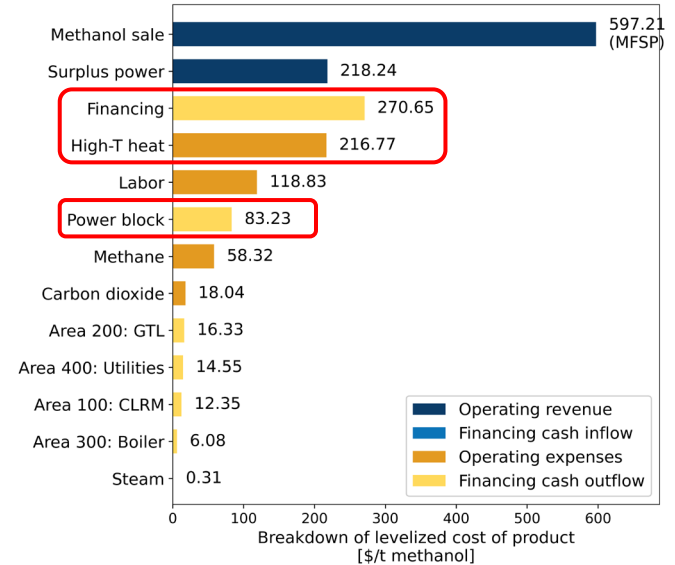


$X_{\text{CH}_4} = 1.000$     $X_{\text{CO}_2} = 0.851$     $X_{\text{H}_2\text{O}} = 0.825$     $Q_{\text{red\_total}} = 40.69 \text{ MW}$

$$X_{\text{CH}_4} = 1, S_{\text{H}_2\text{O}} \cong 1$$

$$X_{\text{CO}_2, \text{ox}} = 0.9, X_{\text{H}_2\text{O}, \text{ox}} = 0.88$$

## Waste heat power block



$X_{\text{CH}_4} = 1.000$     $X_{\text{CO}_2} = 0.851$     $X_{\text{H}_2\text{O}} = 0.825$     $Q_{\text{red\_total}} = 40.69 \text{ MW}$

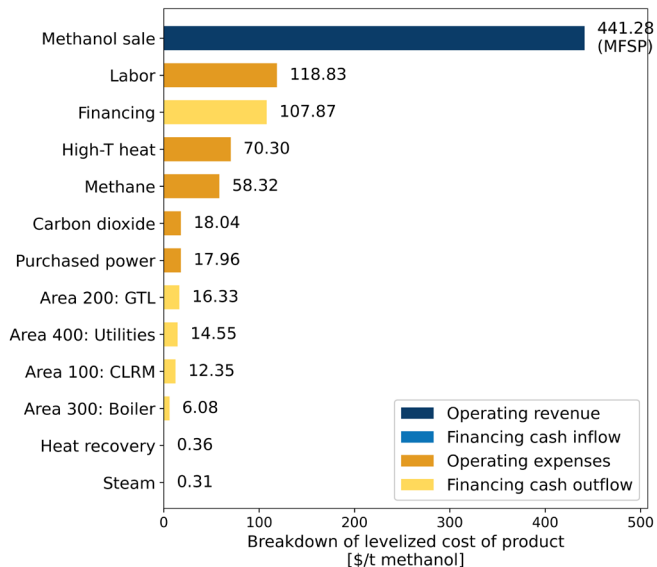
# Process Integration Options: Ideal Case

$$f_{\text{oxide-to-gas}} = 80 \text{ mol}$$

$$R_{\text{oxidizer-to-CH}_4} = 4$$

$$T_{\text{reactor}} = 900^\circ\text{C}$$

## Baseline: Heat recovery only

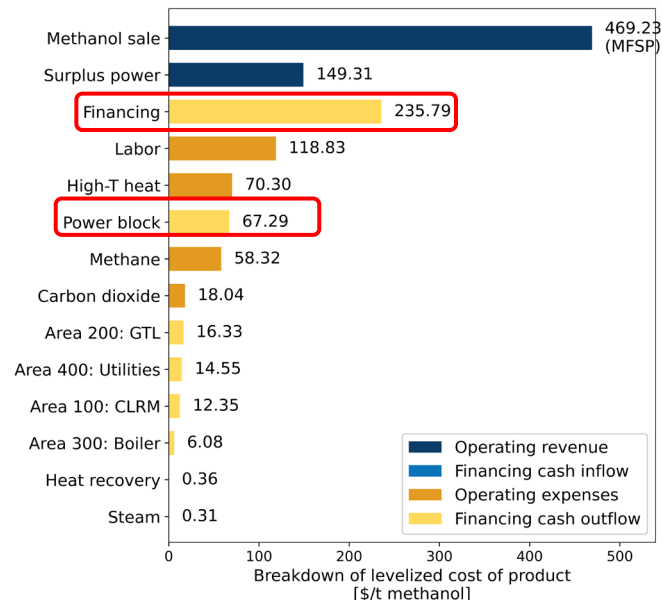


$X_{\text{CH}_4} = 1.000$   $X_{\text{CO}_2} = 0.851$   $X_{\text{H}_2\text{O}} = 0.825$   $Q_{\text{red\_total}} = 40.69 \text{ MW}$

$$X_{\text{CH}_4} = 1, S_{\text{H}_2\text{O}} \cong 1$$

$$X_{\text{CO}_2, \text{ox}} = 0.9, X_{\text{H}_2\text{O}, \text{ox}} = 0.88$$

## Combined HR and PB



$X_{\text{CH}_4} = 1.000$   $X_{\text{CO}_2} = 0.851$   $X_{\text{H}_2\text{O}} = 0.825$   $Q_{\text{red\_total}} = 40.69 \text{ MW}$