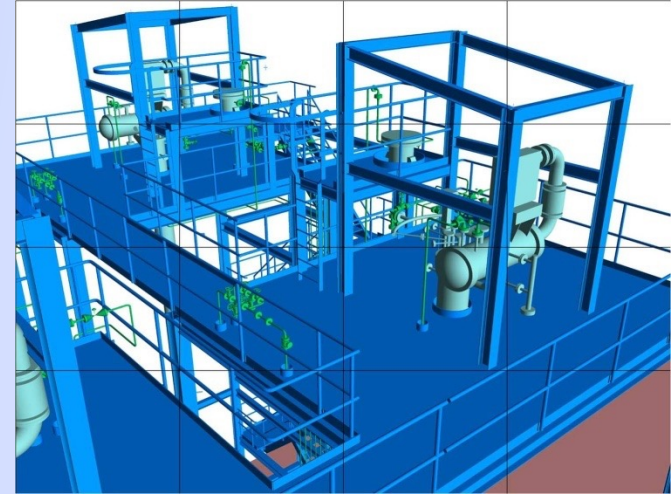# Decarbonization of Steam-Methane Reforming through Induction Heating

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Energy Systems Initiative Seminar; October 9, 2023

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Research Background

- Completed my PhD in Chemical Engineering at the University of the Witwatersrand, SA, 2012.

- Thesis Entitled: Interaction between reaction and phase equilibria in the Fischer–Tropsch reaction.

- Advised by Prof Diane Hildebrandt and Prof David Glasser.

Research Group at Purdue University

- Aim to Revolutionize the Chemical Industry.

- Decarbonization of the Chemical Industry through Renewable/Carbon-Free Electrification.

- Start with the Decarbonization of the most Energy Intensive Processes.

- Development and Demonstration of Modular, Integrated, and Intensified Processes.

- Algorithmic developments in Modeling and Optimization of Sustainable Processes.

Energy Transition

U.S. net electricity generation by fuel
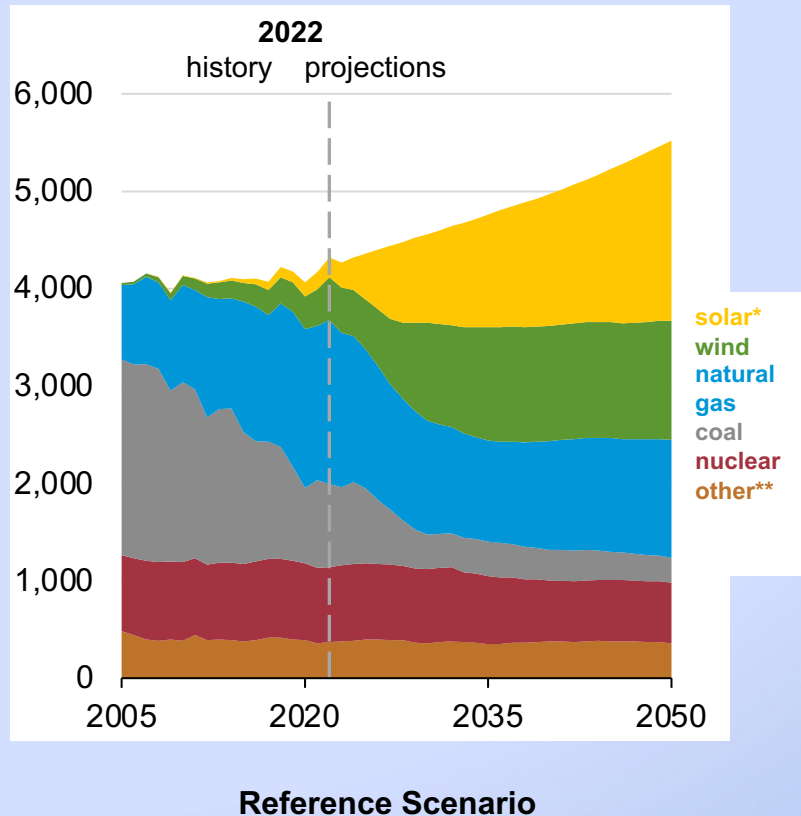billion kilowatthours
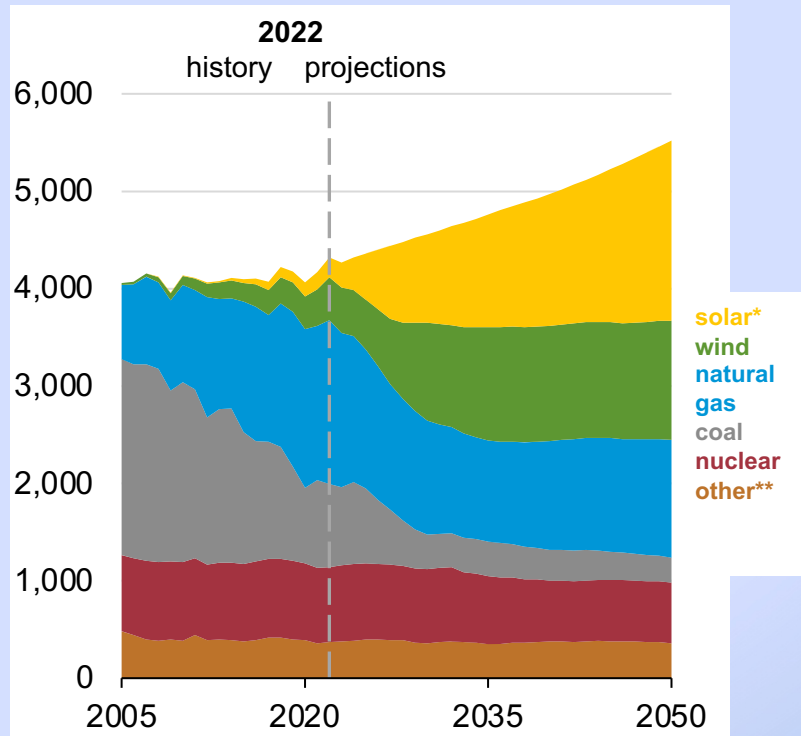
Reference Scenario

Source: US EIA, Annual Energy Outlook 2023

- The Energy Transition is already underway.

- The main challenge for Renewable Energy Sources is Energy Storage.

- Grid Stability is already a bottleneck for new power projects coming on stream.

- Battery Storage potential and capacity is extremely limited.

- Chemicals have a huge potential as energy storage vectors.

- Hydrogen can play a crucial role in facilitating the energy transition.

4

Energy Transition

U.S. net electricity generation by fuel
billion kilowatthours

2022
history   projections

solar*
wind
natural
gas
coal
nuclear
other**

6,000 — 5,000 — 4,000 — 3,000 — 2,000 — 1,000 — 0

2005   2020   2035   2050

Reference Scenario

Source: US EIA, Annual Energy Outlook 2023

- Renewable Energy Sources can help transform the Chemical Industry;

- On the other hand,

- The Chemical Industry can help facilitate the implementation of more Renewable Energy Power Plants.

Process Design in Energy and Sustainability: Towards a Net-Zero Carbon Economy

- Process Industries are notably Energy Intensive, currently Fossil Fuel Fired, and considered Difficult to Decarbonize.

- As more Electricity is either Renewable or otherwise Carbon-Free, Powering more Process Industry Operations with Electricity becomes a Decarbonization Strategy.

- Thus, designing electrically driven process units to take advantage of the carbon-free electricity is imperative.

Towards a Net-Zero CO_2 Chemical Industry

- The Chemical Industry grew out of the First Industrial Revolution where Steam produced by Fossil Fuels Combustion drove all the energy transfers in the Process.

- Curbing carbon dioxide emissions while maintaining quality of life is a global challenge for manufacturing processes that will require process innovation.

- One approach is replacing energy from the burning of carbon-based fuels with energy supplied by 'green' electrons.

- This goal can be achieved in some cases by simply replacing heat supplied by combustion with electrical heating.

Production Cost of Energy
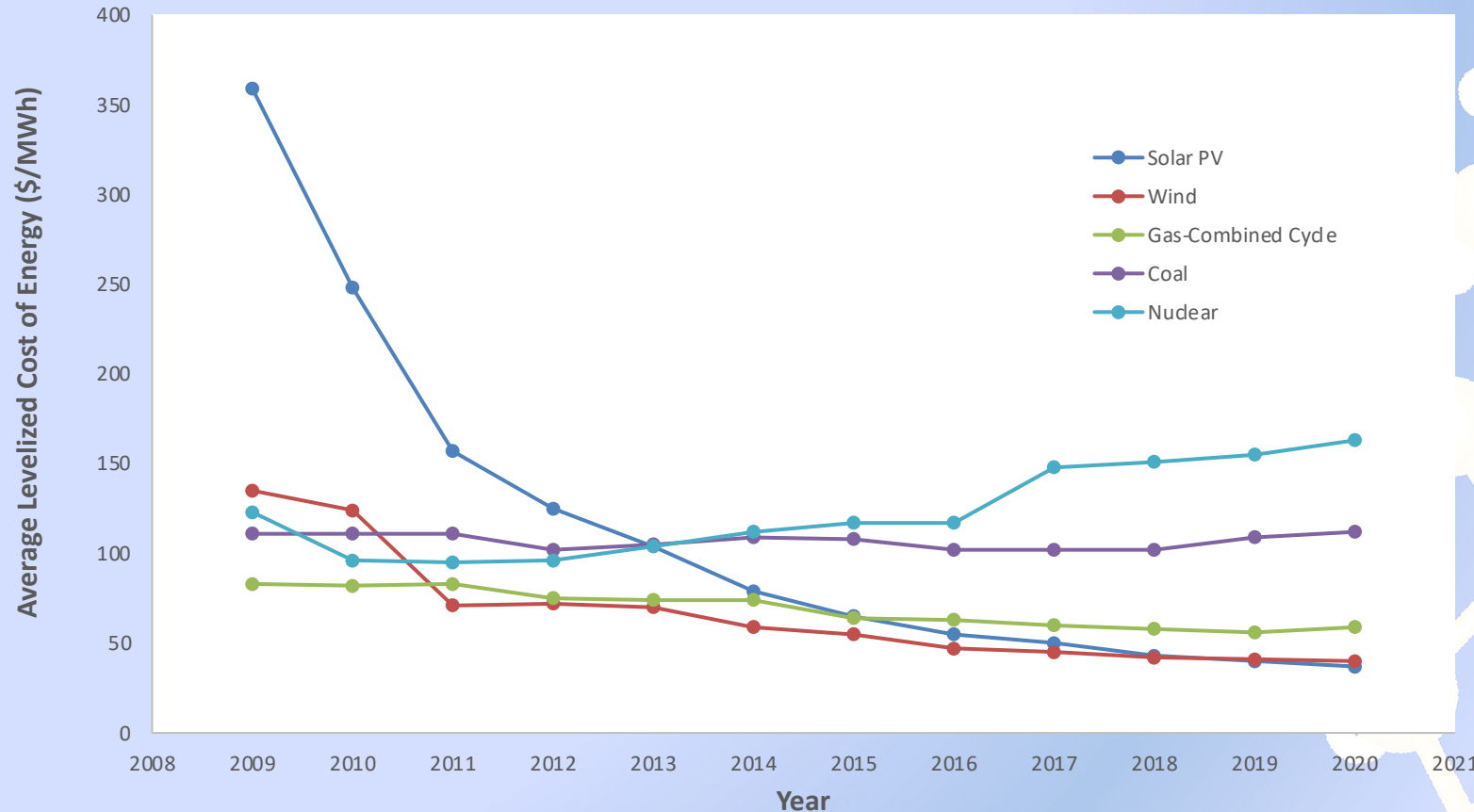
- Lazard's Levelized Cost of Energy Analysis.
- The cost of Solar PV and Wind Energy is now lower than fossil fuel installation.

Process Decarbonization through Electrification

- Thermal Energy is required for Increasing Temperature, Phase Change, and Endothermic Reactions.

- Electricity can Generate Heat via Resistance, Induction, Dielectric, Arc, and Gas Compression Mechanisms, among others.

- Electrical Heat can be delivered with High Precision and Tight Control.

Process Decarbonization through Electrification

- Electricity can power the movement of heat from sources at a lower temperature to needs at higher temperature.

- Electricity can also facilitate chemical reactions that are Thermodynamically difficult, especially the electrolysis of water to produce hydrogen.

- Almost all process industries can benefit from carbon-free electrification.

- The key will be to match the right electrical heating technology at the right temperature with the right application at the right scale.
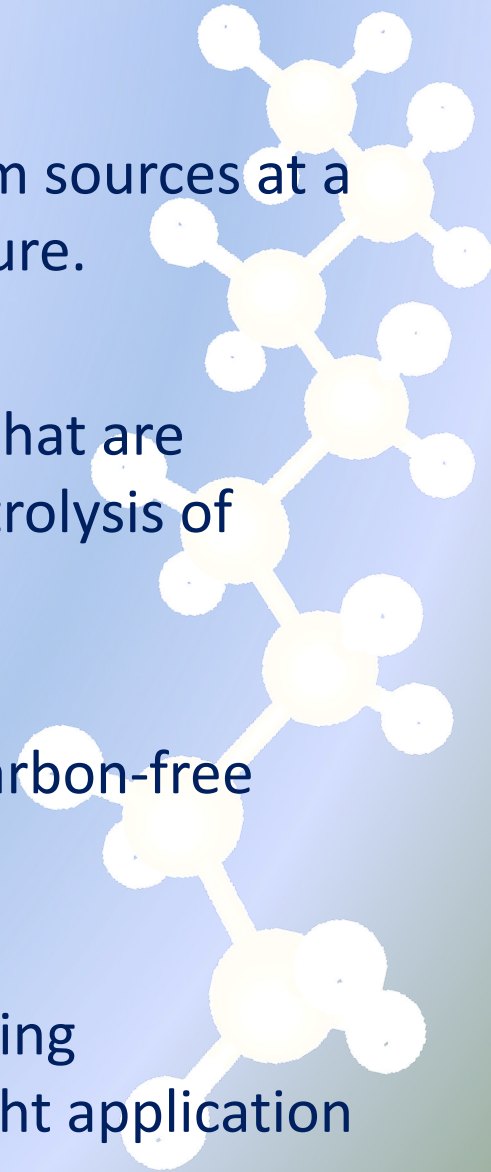
Energy Consumption and Carbon Footprint of High-Volume Commodity Chemicals
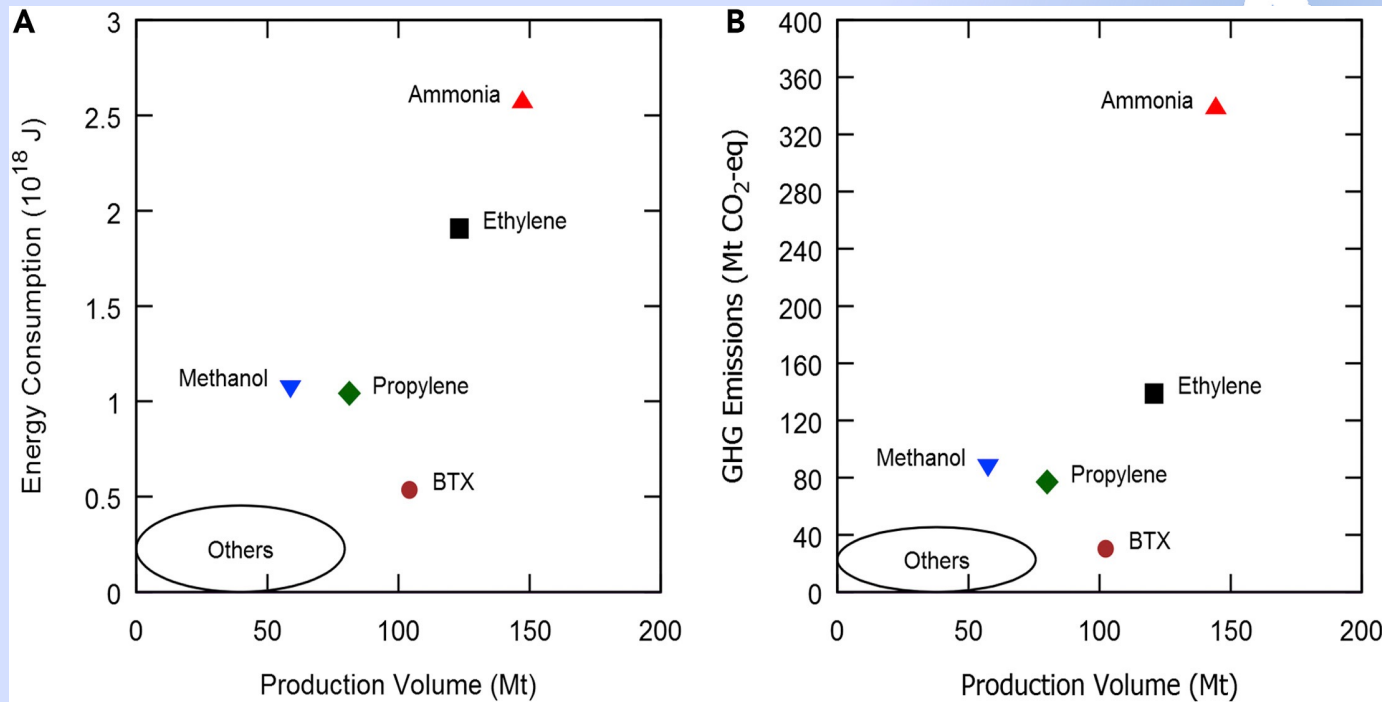
- Performing chemical synthesis with renewable electricity can reduce carbon emissions.

Figure Source: DECHEMA. Low carbon energy and feedstock for the European chemical industry. 2017

Process Decarbonization Starting with The Most Widely Applied Endothermic Reactions

- From a Shale/Natural Gas feed, we are looking at:

- Steam-Methane Reforming;

- Ethane Cracking to Ethylene; and
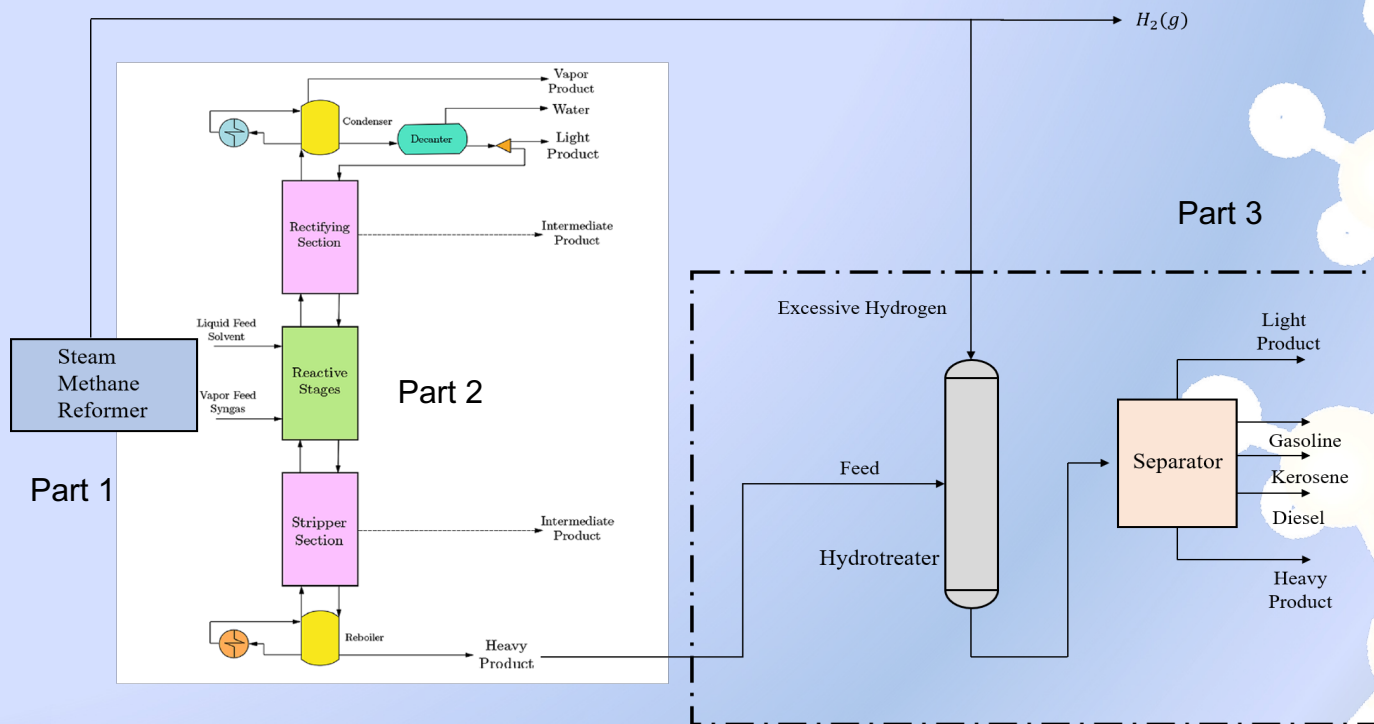
- Propane Cracking to Propylene.

Steam-Methane Reforming

- One of the most important reaction in the Chemical Industry.

- Most of the Hydrogen produced in the Chemical Industry is currently through Steam-Methane Reforming.

- Gas-to-Liquids processes have three main sections:
    1. Syngas Generation (Steam-Methane Reforming);
    2. Syngas Conversion (Fischer–Tropsch Reaction);
    3. Product Refining (Producing Final Product).

- Any carbon efficiency improvements on this Reforming step can have a huge impact on the entire Industry.
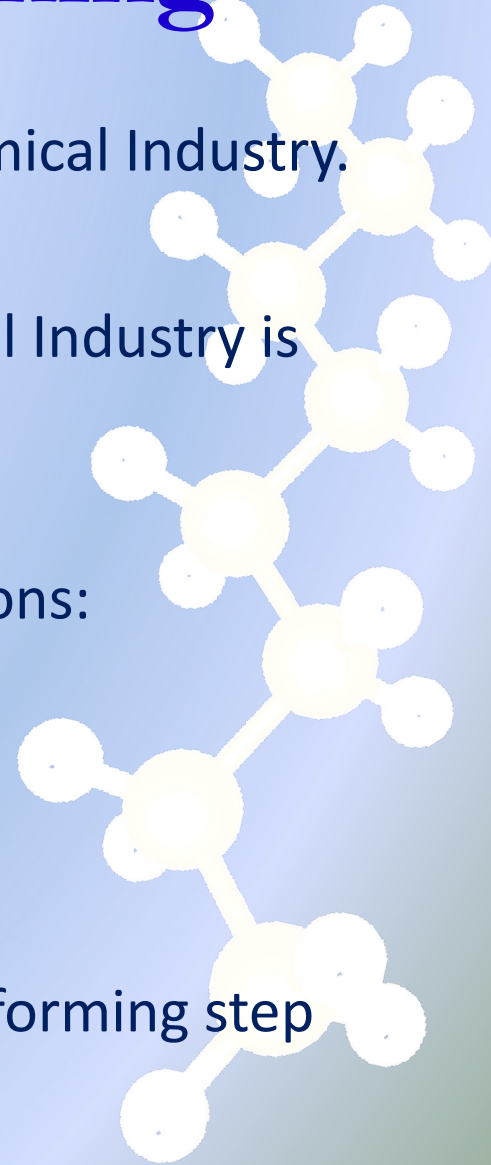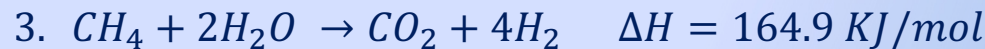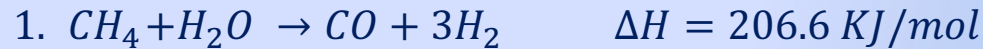
Modular Integrated Processes

- Gas-to-Liquids processes have three main sections:
    1. Syngas generation (Reforming)
    2. Syngas conversion (Fischer–Tropsch reaction)
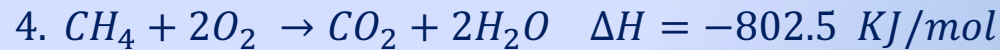   3. Product refining (to produce final products)

N. He, Y. Hu, C.M. Masuku, L.T. Biegler, 110th Anniversary: Fischer–Tropsch synthesis for multiphase product recovery through reactive distillation, *Ind. Eng. Chem. Res. 58* (2019) 13249–13259.

Steam and Partial Oxidation Reformer

Reactions: Oxidation reaction provides extra heat

- Steam reforming reactions:

  1. $CH_4 + H_2O \rightarrow CO + 3H_2$ $\quad\quad \Delta H = 206.6 \, KJ/mol$
  2. $CO + H_2O \rightarrow CO_2 + H_2$ $\quad\quad \Delta H = -41.1 \, KJ/mol$
  3. $CH_4 + 2H_2O \rightarrow CO_2 + 4H_2$ $\quad \Delta H = 164.9 \, KJ/mol$

- Oxidation reaction:

  4. $CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$ $\quad \Delta H = -802.5 \, KJ/mol$

Kinetics: Obtained from literature (Trimm & Lam, 1980; Xu & Froment, 1989)

$$DEN = 1 + K_{CO}P_{CO} + K_{H_2}P_{H_2} + K_{CH_4}P_{CH_4} + \frac{K_{H_2O}P_{H_2O}}{P_{H_2}}$$

$$R_1 = \eta_1 \frac{k_1}{P_{H_2}^{2.5}} \left(P_{CH_4}P_{H_2O} - \frac{P_{H_2}^3 P_{CO}}{Ke_1}\right)/DEN^2$$

$$R_2 = \eta_2 \frac{k_2}{P_{H_2}} \left(P_{CO}P_{H_2O} - \frac{P_{H_2}P_{CO_2}}{Ke_2}\right)/DEN^2$$

$$R_3 = \eta_3 \frac{k_2}{P_{H_2}^{3.5}} \left(P_{CH_4}P_{H_2O}^2 - \frac{P_{H_2}^4 P_{CO_2}}{Ke_3}\right)/DEN^2$$

$$R_4 = \eta_4 \frac{k_{4a}P_{CH_4}P_{O_2}}{(1 + K_{CH_4}^{ox}P_{CH_4} + K_{O2}^{ox}P_{O_2})^2} + \frac{k_{4b}P_{CH_4}P_{O_2}}{(1 + K_{CH_4}^{ox}P_{CH_4} + K_{O_2}^{ox}P_{O_2})}$$

D. Trimm, C.-W. Lam, The combustion of methane on platinum—alumina fibre catalysts—II design and testing of a convective-diffusive type catalytic combustor, *Chem. Eng. Sci. 35* (1980) 1731–1739.
J. Xu, G.F. Froment, (1989). Methane steam reforming, methanation and water-gas shift: I. Intrinsic kinetics, *AIChE J. 35* (1989) 88–96.
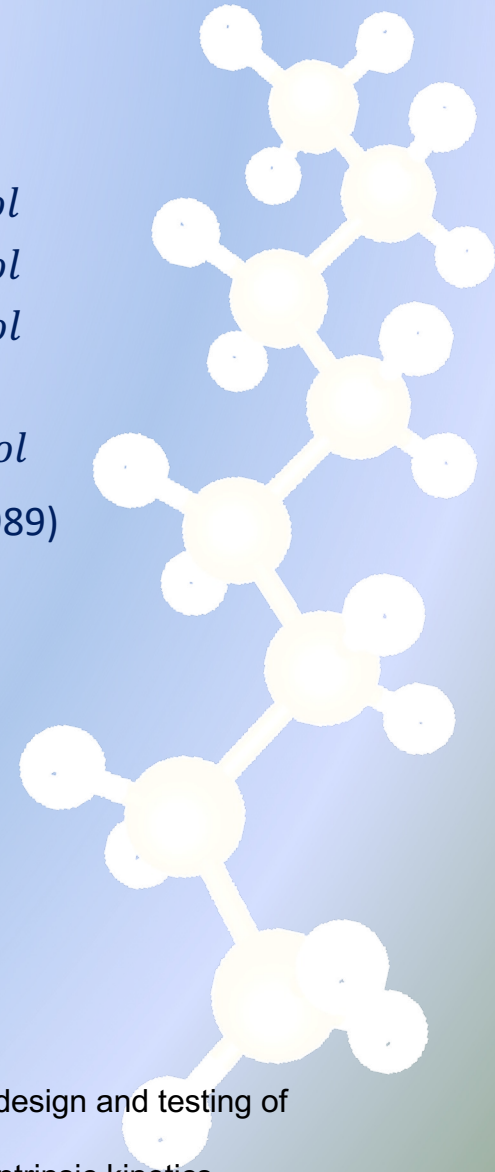
Reforming Modeling Assumptions

Heat balance: Oxidation reaction provides extra heat

$$\frac{dT}{dZ} = \frac{\sum_i A_c(1-\varepsilon)\rho_{cata}\Delta H_i R_i}{u\rho Cp} \qquad i = R_1, R_2, R_3, R_4$$

$$Cp_j = A + Bt + Ct^2 + Dt^3 + E/t^2 \qquad j = CH_4, H_2O, CO, H_2, CO_2, O_2$$

$$Cp = \frac{\sum_j Cp_j F_j}{F_{total}}$$

Pressure Drop:

$$\frac{dP}{dZ} = \frac{G\mu}{\rho d_p^2} \frac{(1-\varepsilon)^2}{\varepsilon^3} \left(150 + 1.75 \frac{Re_p}{1-\varepsilon}\right)$$

(Some parameters from Smet et al., 2001)

C.D. Smet, M.D. Croon, R. Berger, G. Marin, J. Schouten, Design of adiabatic fixed-bed reactors for partial the partial oxidation of methane to synthesis gas. Application to production of methanol and hydrogen-for-fuel-cells, *Chem. Eng. Sci. 56* (2001) 4849–4861.

Reforming simulation

Inlet & Operating Conditions

Term	value
Reaction inlet pressure (bar)	10
Reaction inlet temperature (K)	720
reactor diameter (m)	0.03
reactor length (m)	3
Catalyst porosity	0.85
Catalyst diameter (m)	0.005
Catalyst density (kg/m^3)	1820
Inlet Steam to methane ratio	1:6
Inlet methane to oxygen ratio	3:2

⟹ Python scipy Runge-Kutta 45 solver used to solve for the simulation problem

Simulation results: ⟸

further used as initial guess to optimization problem
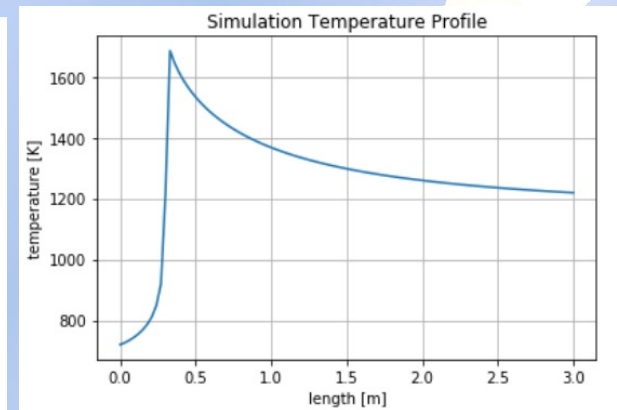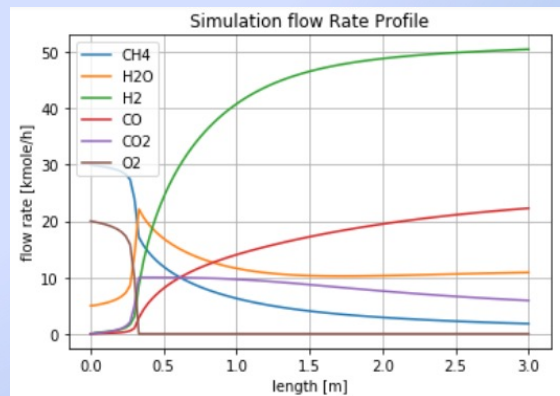
Reforming Optimization

PYOMO DAE collocation method with CONOPT solver is used to solve the optimization problem.

Objective Function:

Minimize Byproducts Concentration

Inlet and operating variables range:

$$600 < \text{inlet } T < 900 \, K$$

$$5.0 < \text{inlet } P < 15.0 \, \text{Bar}$$

$$0 < \text{inlet } O_2 \, \& \, H_2O \text{ Flowrate} < 30.0 \, kmol/hr$$

Outlet Conditions specification:

$$1.95 < H_2 : CO < 2.05$$

Optimization results:

- DAE Problem discretized to 100 finite elements with 3 point Lagrange-Radau collocation method containing 9335 constraints and 9337 variables is solved within 0.20 CPU s
- Byproducts are decreased from 20.4% to 7.2%, most is just water.
- Outlet 2:1 hydrogen to carbon dioxide ratio has been reached

Optimizaion Flow Rate Profile

Optimization Temperature Profile
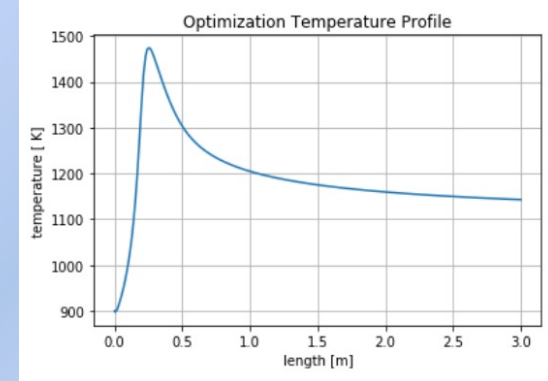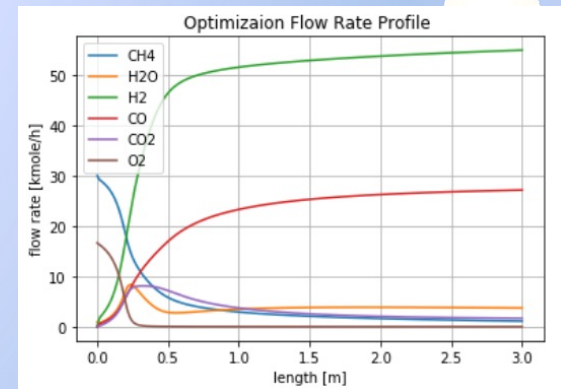
Membrane Steam Methane Reformer (MSMR)

Reactions:

- Steam reforming reactions:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \qquad \Delta H = 206.6 \; KJ/mol$$

$$CO + H_2O \rightarrow CO_2 + H_2 \qquad \Delta H = -41.1 \; KJ/mol$$

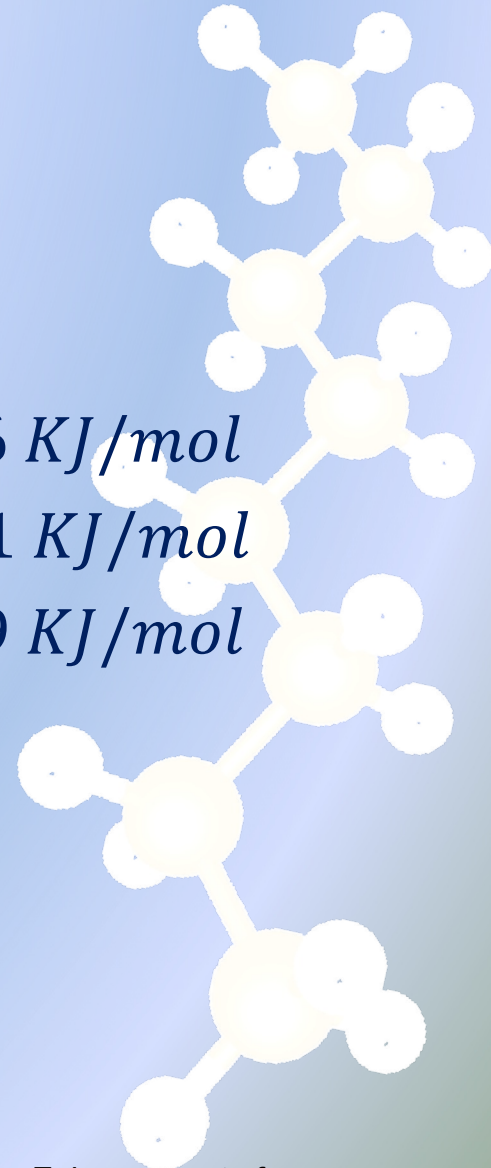$$CH_4 + 2H_2O \rightarrow CO_2 + 4H_2 \qquad \Delta H = 164.9 \; KJ/mol$$

Permeation:

mass flux

$$J_{H_2} = Q \; exp\left(\frac{-E_{mem}}{RT}\right)\frac{P_{H_2,pcl}^{0.5} - P_{H_2,oc}^{0.5}}{d_{mem}}$$
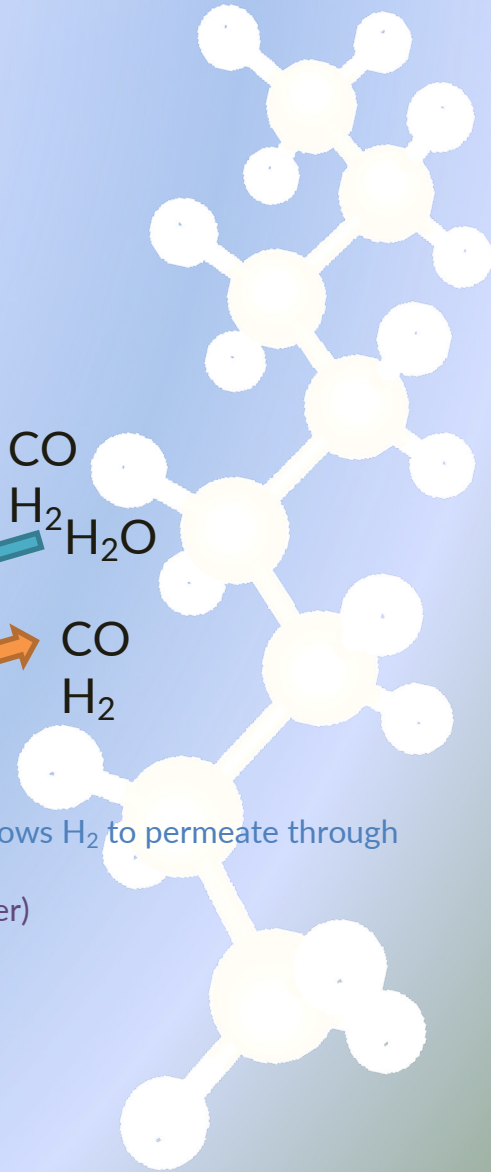
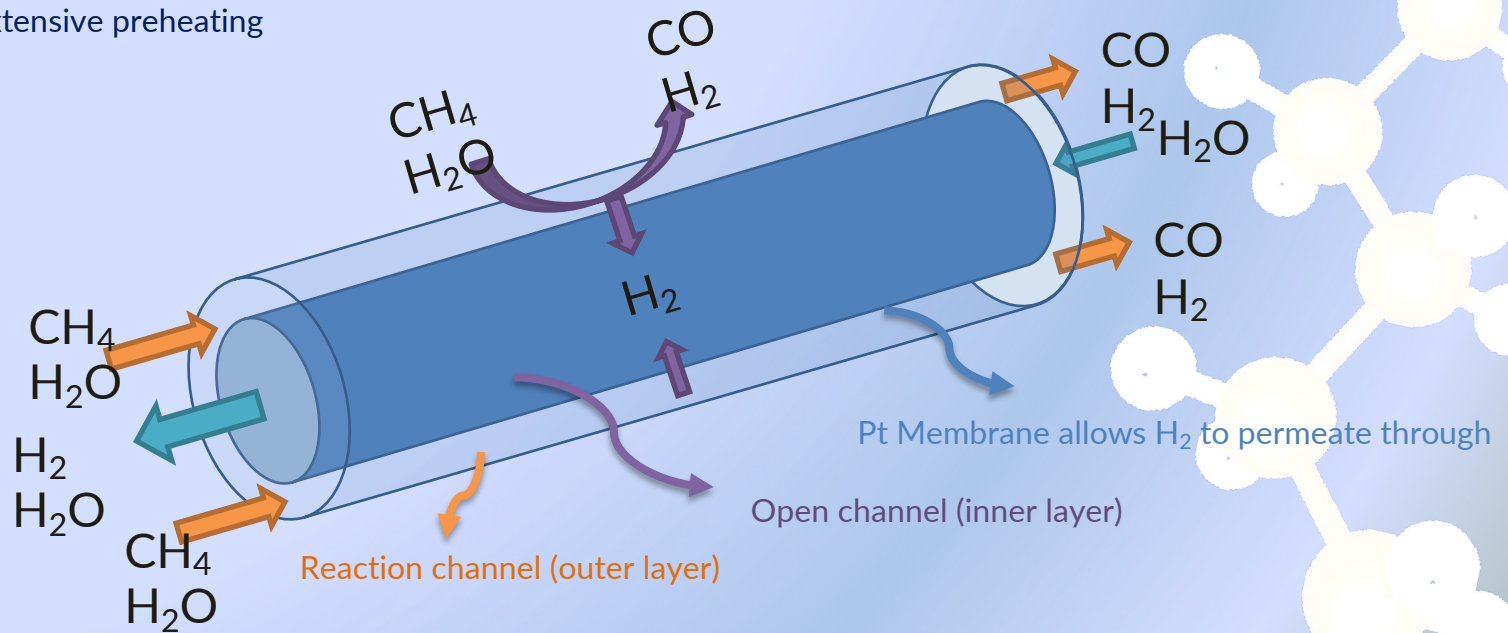(Flux parameters from Kyriakides et al, 2014)

A.-S. Kyriakides, L. Rodriguez-Garcia, S. Voutetakis, D. Ipsakis, P. Seferlis, S. Papadopoulou, Enhancement of pure hydrogen production through the use of a membrane reactor, *Int. J. Hydrog. Energy 39* (2014) 4749–4760.

Membrane Steam-Methane Reformer (MSMR)

Strength:
- Removing extra generated hydrogen will shift equilibrium of steam methane reaction
- More product can be produced thus less extreme operating condition is allowed
- H_2 can then be fed to other units in GTL process

Weakness:
- Requires extensive preheating

CO
H_2

CH_4
H_2O

CO
H_2 H_2O

CO
H_2

H_2

CH_4
H_2O

H_2
H_2O

CH_4
H_2O

Pt Membrane allows H_2 to permeate through

Open channel (inner layer)

Reaction channel (outer layer)

MSMR Modeling assumptions

Ignore radial concentration and temperature distribution

Assume both layer of stream are preheated to a high

    temperature

Concentration profile:

Reaction channel:
$$\frac{dF_j}{dZ} = \sum_{i=1}^{3} R_i \nu_{i,j} \rho_{cata} (1 - \varepsilon) A_c \quad j = CH_4, H_2O, H_2, CO, CO_2$$

Open Channel:
$$\frac{dF_{H_2,oc}}{dZ} = -2\pi r_1 J_{H_2}$$

Pressure drop profile:

Reaction channel:
$$\frac{dP}{dZ} = \frac{G\mu}{\rho d_p^2} \frac{(1 - \varepsilon)^2}{\varepsilon^3} \left(150 + 1.75 \frac{Re_p}{1 - \varepsilon}\right)$$

Open Channel:
$$\frac{dP_{oc}}{dZ} = \frac{4\mu_{oc} u_{oc}}{r_2^2}$$

Temperature profile:

Reaction channel :
$$\frac{dT_{rc}}{dZ} = \frac{1}{u\rho Cp} \left(2\pi r_2 h(T_{mem} - T_{rc}) + \sum_{i=1}^{3} A_c (1 - \varepsilon) \rho_{cata} \Delta H_i R_i\right)$$

Open Channel:
$$\frac{dT_{oc}}{dZ} = -\frac{1}{u_{oc} \rho_{oc} Cp_{oc}} \left(2\pi r_2 h(T_{mem} - T_{oc})\right)$$

MSMR Simulation

MSMR Inlet & Operating Conditions:

Term	value
Length of reactor (L)	1m
Radius of OC (r_1)	0.007m
Radius of reactor system (r_2)	0.021m
Thickness of semi-permeable membrane (d_{mem})	5×10^{-6}m
Reaction tube inlet pressure (bar)	10
Flue tube inlet pressure (bar)	1
Reaction tube inlet temperature (K)	823
Flue tube inlet temperature (K)	823
Catalyst porosity	0.85
Catalyst diameter (m)	0.005
Inlet CH_4 flow rate	0.01 kmol/hr
Inlet H_2O flow rate	0.01 kmol/hr

- Model: Boundary Value problem

- Method: Single shooting method

- Approach: Python with Scipy ODE solver

MSMR Simulation results:
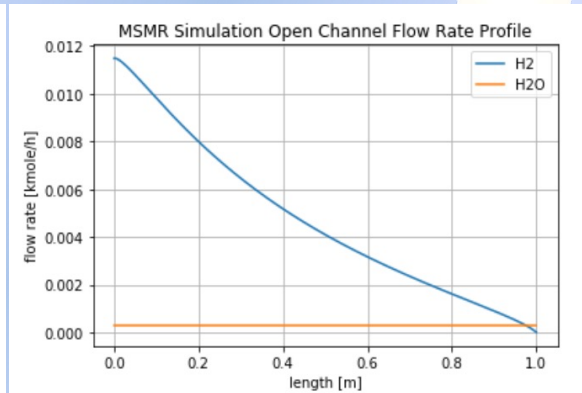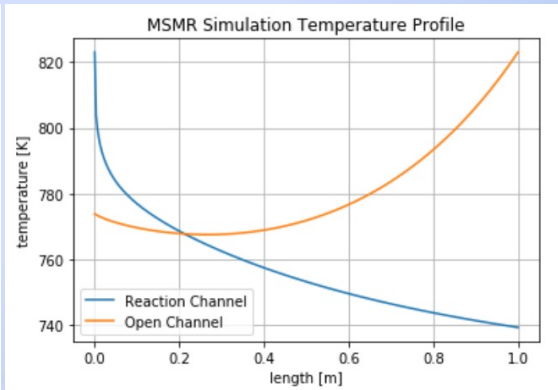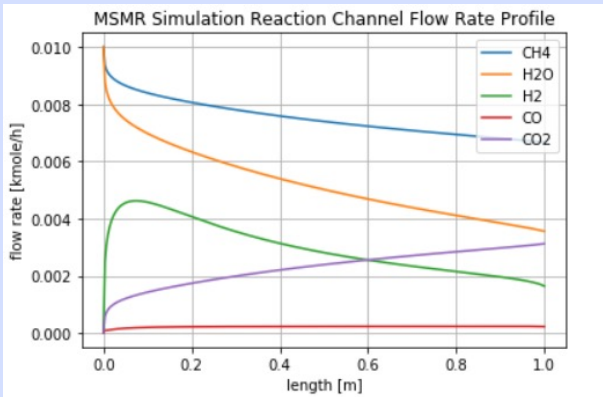
MSMR Optimization Conditions

Objective Function:

Minimize Byproducts Concentration

PYOMO DAE collocation method with CONOPT solver is used to solve the optimization problem.

Inlet and Operating Parameters:

Reactor Length L:	$1 m$
Radius of Open Channel (r_1):	$0.007 m$
Semi $-$ permeable membrane thickness(d_{mem}):	5×10^{-6}
Radius of Reaction Channel (r_2):	$0.021 m$
Inlet CH_4 Flowrate: (F_{CH_4}):	$0.01 \ kmol/hr$

Inlet and operating variables range:

$$600 < \text{inlet } T < 1200 \ K$$
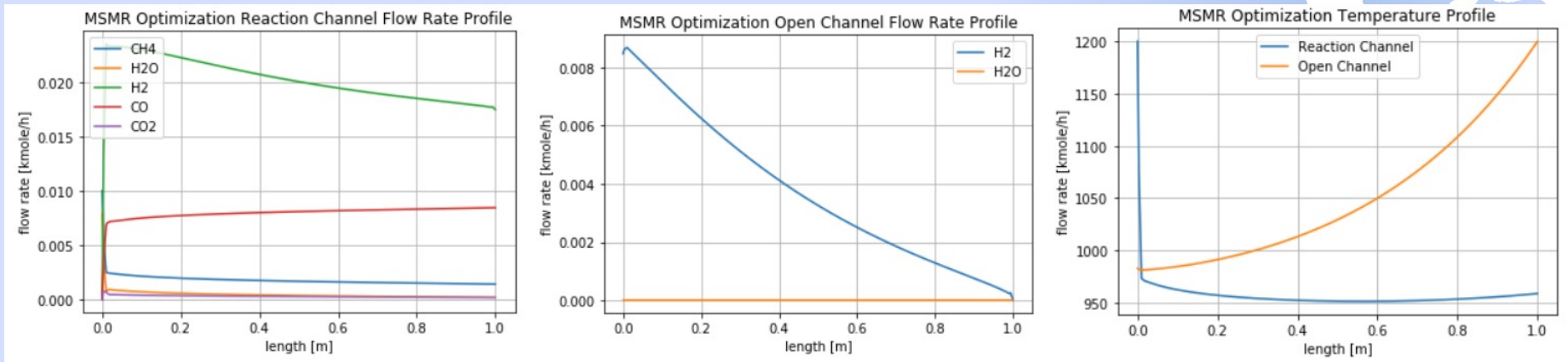
$$5.0 < \text{inlet } P_{rc} < 20.0 \ \text{Bar}$$

$$1.0 < \text{inlet } P_{oc} < 3.0 \ \text{Bar}$$

$$0 < \text{inlet } H_2O \text{ Flowrate} < 1.0 \ kmol/hr$$

Outlet Conditions specification:

$$1.95 < H_2 : CO < 2.05$$

MSMR Optimization Results

Optimization results:
- DAE Problem discretized to 100 finite elements with 3 point Lagrange-Radau collocation method containing 9335 constraints and 9339 variables is solved within 0.70 CPU s
- Byproducts are decreased to 6.4%
- Outlet 2:1 hydrogen to carbon dioxide ratio has been reached
- Inlet temperature reached upper bound which is 1200 K. It suggests that extensive preheating is required for this process

Y. Xing, C.M. Masuku, L.T. Biegler, Modular gas-to-liquids process with membrane steam-methane reformer and Fischer–Tropsch reactive distillation, *AIChE J. 67* (2021) e17467.

Modeling the Reaction Kinetics

- We previously used simplified kinetics (pseudo-homogeneous) to design the Membrane SMR reactor (Xing et. al., 2021).

- In the pseudo-homogeneous model, the reactor bed is viewed as a single, continuous phase,

- With the assumption that the temperatures of the solid and fluid are identical at a given location in the reactor.

- This disregards intra-particle diffusion limitations on the reaction as well as inter-particle mass and heat transfer resistances.

Y. Xing, C.M. Masuku, L.T. Biegler, Modular gas-to-liquids process with membrane steam-methane reformer and Fischer–Tropsch reactive distillation, *AIChE J. 67* (2021) e17467.

Pseudo-Homogeneous vs. Heterogeneous Models

- To compensate for the diffusion resistance within the pellet's external film (external diffusion resistance), and its internal pores (internal diffusion resistance), the reaction rates are multiplied by an effectiveness factor to account for the overall diffusion effects on the intrinsic reaction rates.

- Using a constant effectiveness factor is an acceptable simplification, having a great effect on computational time.

- On the other hand, heterogeneous models consider the solid and fluid phases individually, incorporating distinct mass and energy transport equations for the fluid phase in the tube and the catalyst particles.

- The equations for the fluid and solid are interlinked through the mass and heat transfer coefficient.

- This enables the representation of temperature and concentration differences between the fluid and solid phases.
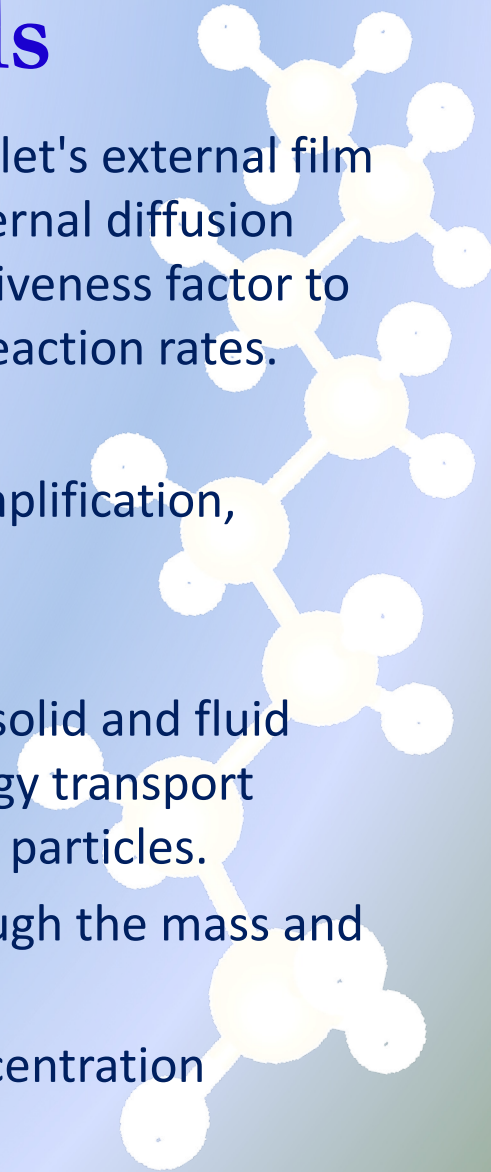
Dynamic Model with Axial Mixing

- Ideal plug flow may not be realized in many packed-bed reactors, there is always a degree of axial mixing in a tubular flow reactor, although the amount of this mixing may be small.

- Therefore, we developed a second model (M2) that allows for deviations from plug flow by considering axial mixing, and its comparison with the first model (M1) is of interest.

- The mixing in the axial direction, which is due to the turbulence and the presence of packing, is accounted for by superimposing an "effective" transport mechanism on the overall transport by plug flow.

- The flux due to this mechanism is described by a formula analogous to Fick's law for mass transfer or Fourier's law for heat transfer by conduction.

- The proportionality constants are effective diffusivities and conductivities.
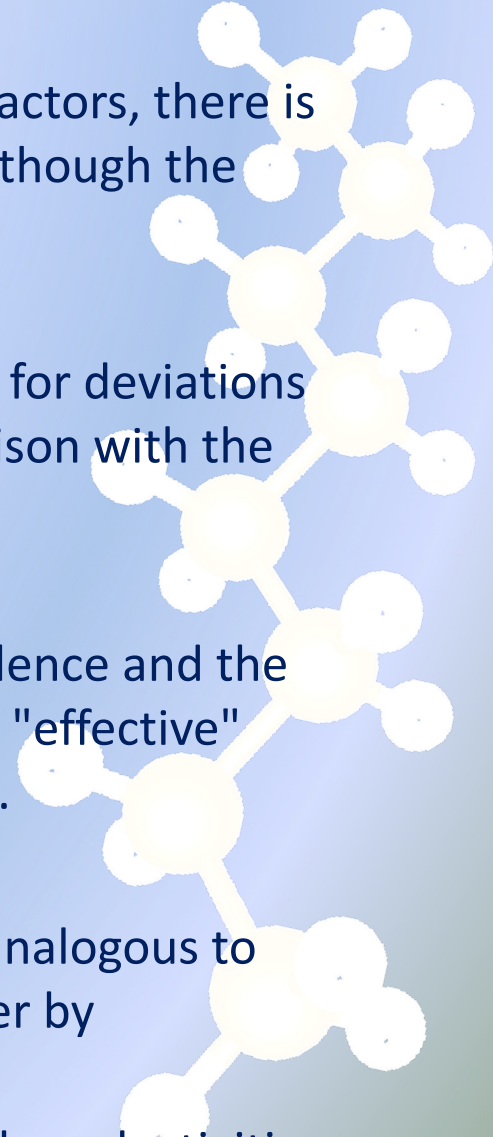
Dynamic Model with Axial Mixing

- Considering the axial mixing, the material balance equation is:

$$\varepsilon_b \frac{\partial C_i(z)}{\partial t} + \frac{\partial [C_i(z)U_z(z)]}{\partial z} = \varepsilon_b \mathcal{D}_{i,z}^e(z) \frac{\partial^2 C_i(z)}{\partial z^2} + (1 - \varepsilon_b)\rho_p \sum \eta_j R_j(z)\nu_{ij}$$
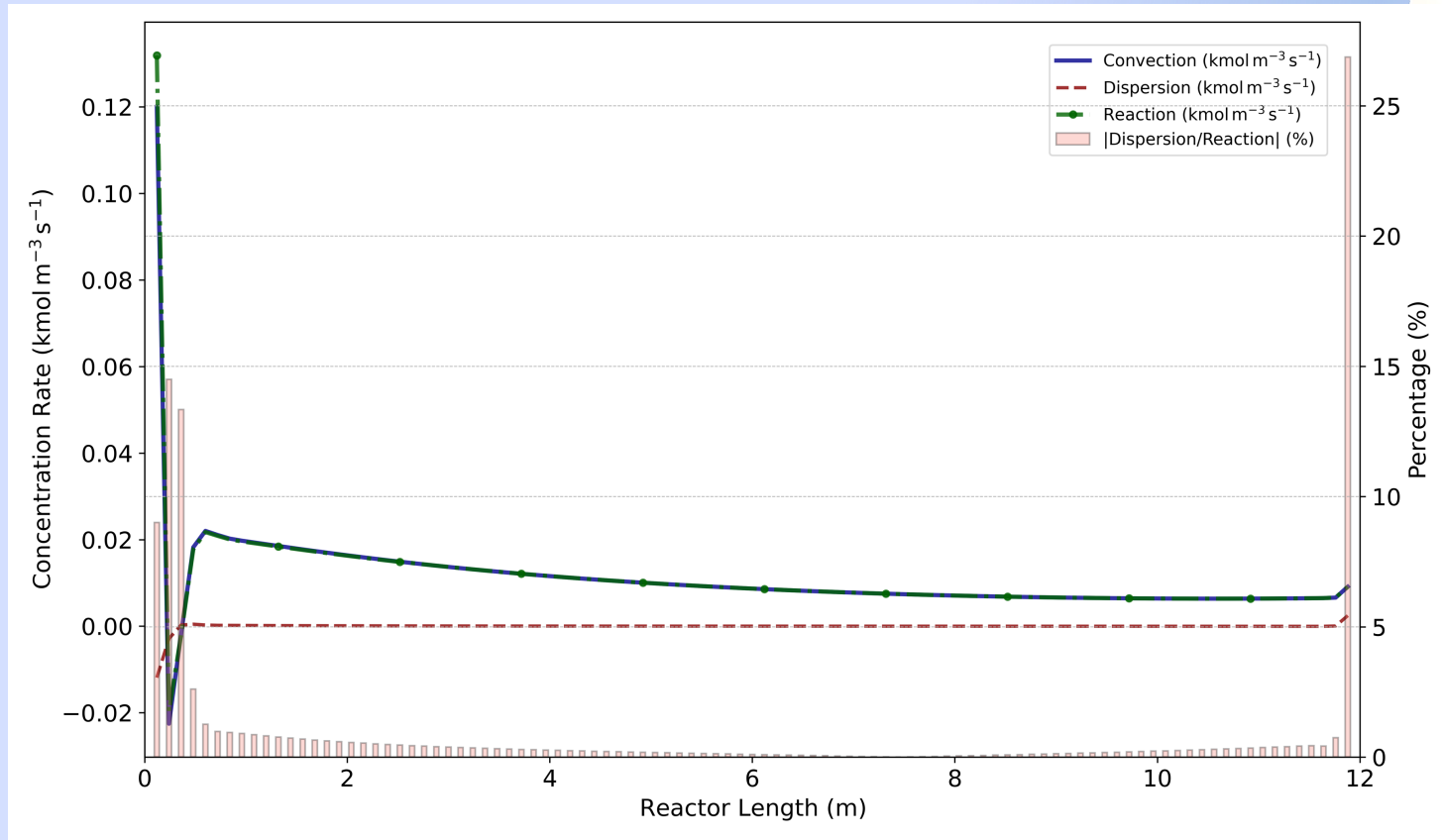
$$\forall z \in (0, L)$$

- The energy balance equation is:

$$\left[(1 - \varepsilon_b)\rho_p C_{p,p} + \varepsilon_b \rho_g(z)C_{p,g}(z)\right] \frac{\partial T(z)}{\partial t} + \rho_g(z)C_{p,g}(z)U_z(z)\frac{\partial T(z)}{\partial z}$$

$$= \frac{4U(z)}{d_t}[T_w(z) - T(z)] + \sum \left[\varepsilon_b \mathcal{D}_{i,z}^e(z)\frac{\partial C_i(z)}{\partial z} C_{p,i}\right]\frac{\partial T(z)}{\partial z} + \lambda_z^e(z)\frac{\partial^2 T(z)}{\partial z^2}$$

$$+ (1 - \varepsilon_b)\rho_p \sum \eta_j R_j(z)\left[-\Delta H_{rxn,j}(z)\right] \qquad \forall z \in (0, L)$$

Concentration Profiles along the Reactor Length with Axial Mixing


The introduction of axial mass dispersion in the mass balance equation and incorporating heat conduction and dispersion terms in the energy equation have limited influences on the overall performance.

# Concentration Profiles along the Reactor Length with Axial Mixing

- In the concentration rate profiles, the convention term, $\frac{\partial[C_i(z)U_z(z)]}{\partial z}$, and the reaction term, $(1 - \varepsilon_b)\rho_p \sum \eta_j R_j(z)v_{ij}$, coincide.

- This is no surprise as the dispersion (axial mixing) term, $\varepsilon_b \mathcal{D}_{i,z}^e(z)\frac{\partial^2 C_i(z)}{\partial z^2}$, is relatively small and usually ignored in literature.

- Except for the reactor entrance and exit, the proportion of dispersion/reaction is less than 1%.
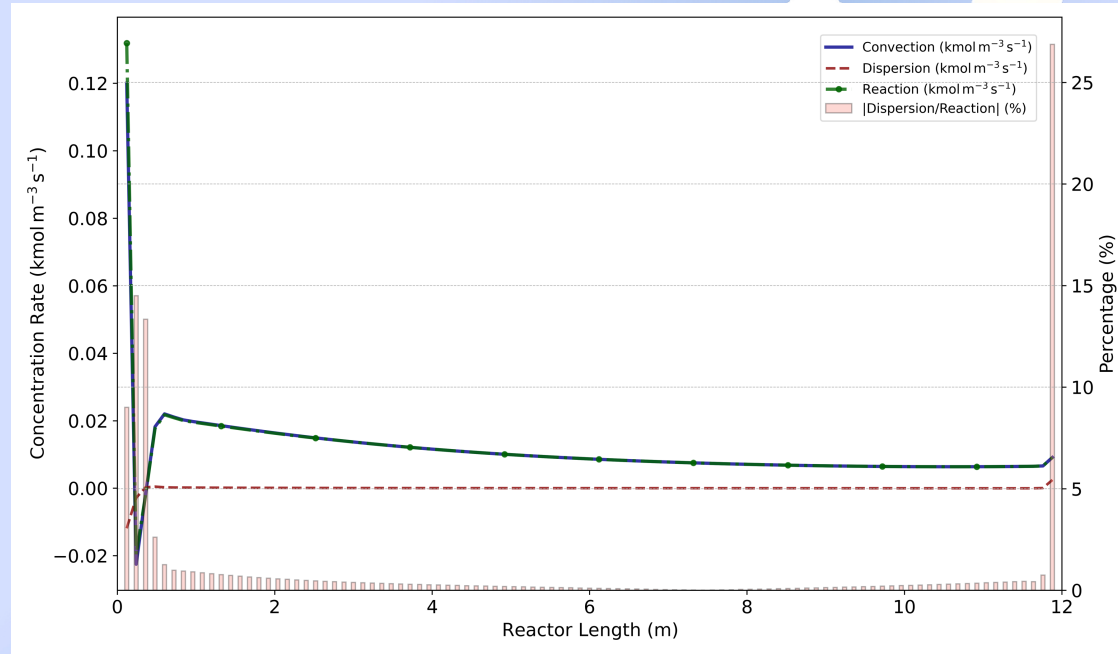
Energy Profile along the Reactor Length
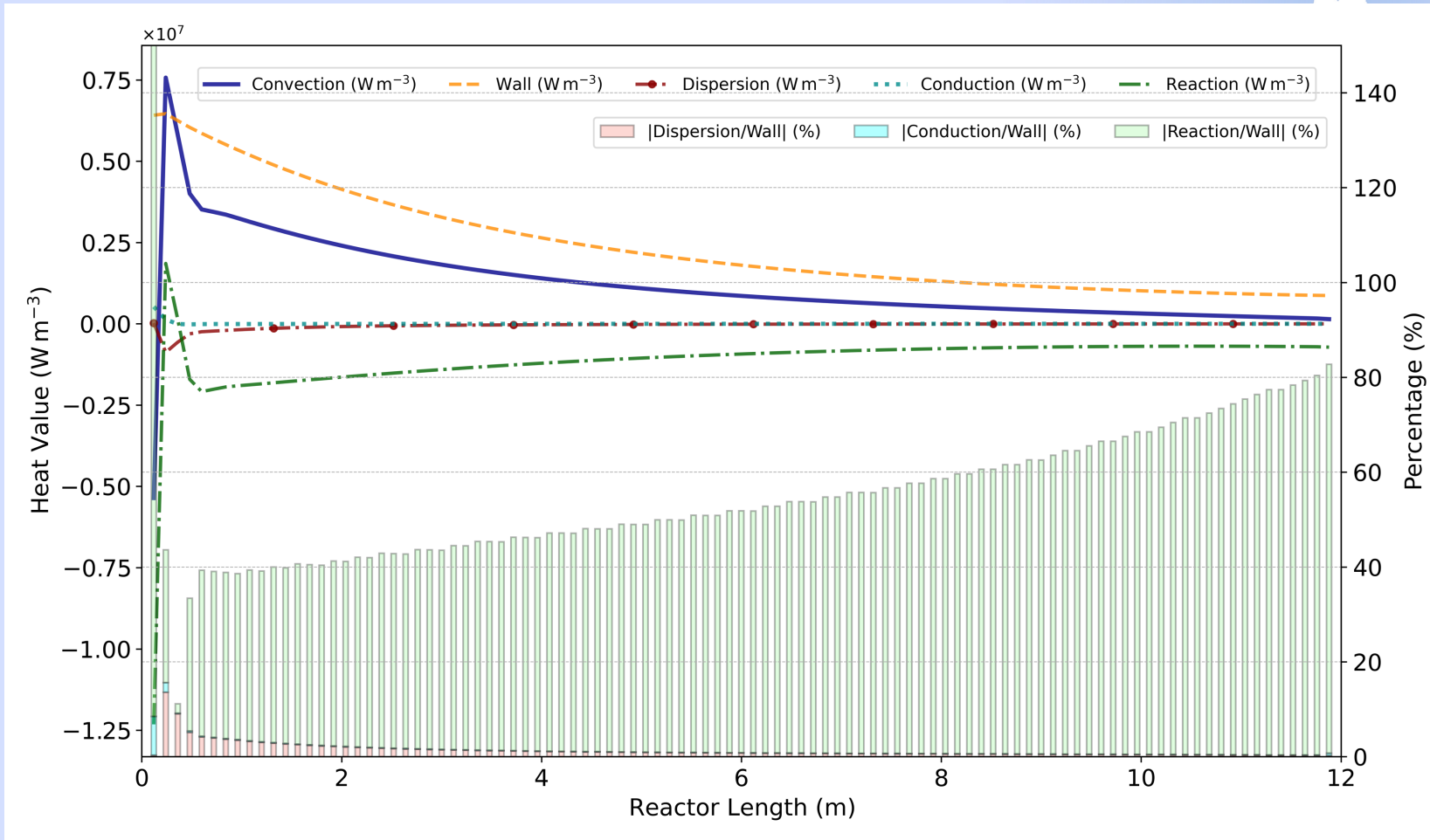
We examined the energy profile of model 2, and the heat dispersion, $\sum \left[\varepsilon_b \mathcal{D}_{i,z}^e(z) \frac{\partial C_i(z)}{\partial z} C_{p,i} \right] \frac{\partial T(z)}{\partial z}$, and the conduction, $\lambda_z^e(z) \frac{\partial^2 T(z)}{\partial z^2}$, terms are relatively small compared to the heat input from wall.

Energy Profile along the Reactor Length

- Also, it is interesting to note that the overall reaction heat is positive at the tube entrance, which means the overall reaction is exothermic.

- This can be attributed to the exothermic WGS reaction.

- This validates the assumption that axial mixing is relatively small in the SMR reactor which is operated in a turbulent flow.
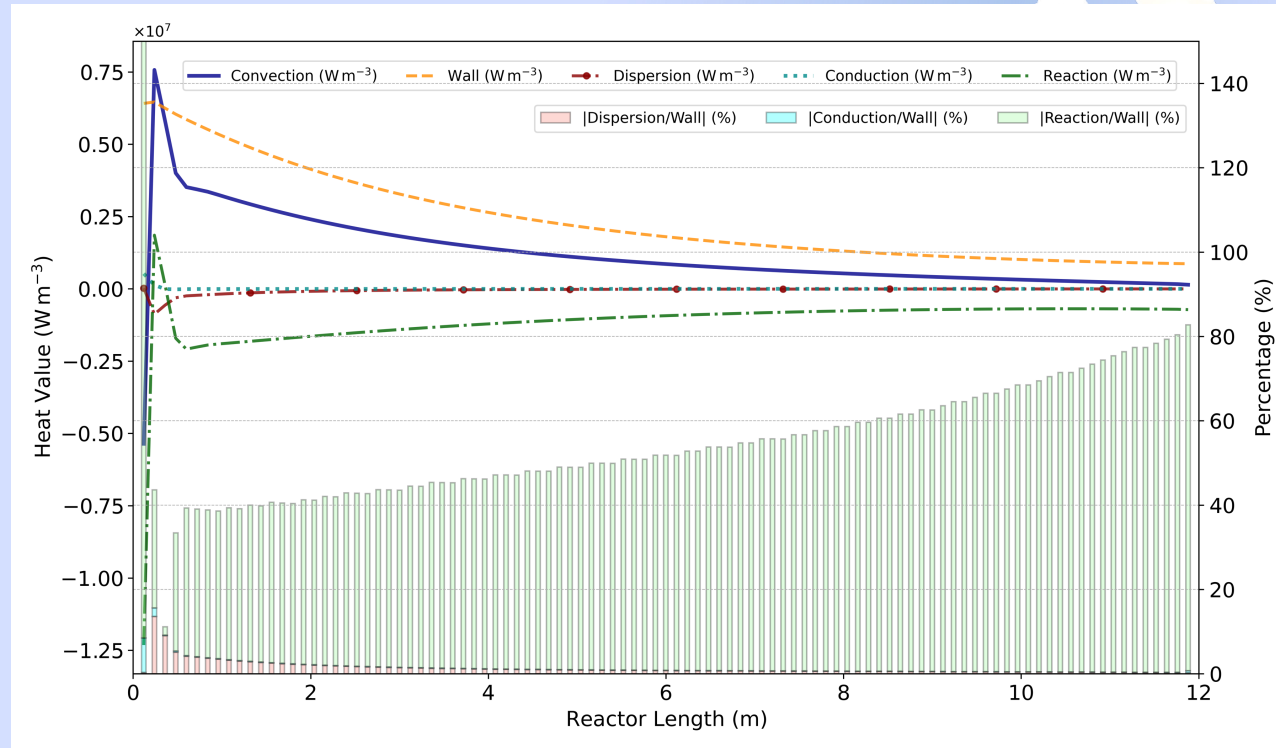
Dynamic Model of an Industrial Heterogeneous Catalytic Reactor

- The next step is to develop a packed-bed model incorporating transport properties to the catalyst.

- We plan to incorporate the bulk gas – catalyst particle mass and heat transfer coefficients.

- To calculate the axial mixing, it is necessary to have values of the axial dispersion coefficients.

- We plan to use correlations by Foumeny et. al. (1992) to estimate the effective mass dispersion coefficients in the packed-bed reactor.

- We plan to approximate molecular diffusion coefficients following the approach of Ghouse and Adams (2013).

E.A. Foumeny, M.A. Chowdhury, C. McGreavy, J.A.A. Castro, Estimation of dispersion coefficients in packed beds, *Chem. Eng. Technol. 15* (1992) 168–181.
J.H. Ghouse, T.A. Adams, A multi-scale dynamic two-dimensional heterogeneous model for catalytic steam methane reforming reactors, *Int. J. Hydrog. Energy 38* (2013) 9984–9999.

Electric Heating of Reactors, Boilers, Reboilers, etc.

- Resistance heating,

- Induction heating,

- Radiation heating,

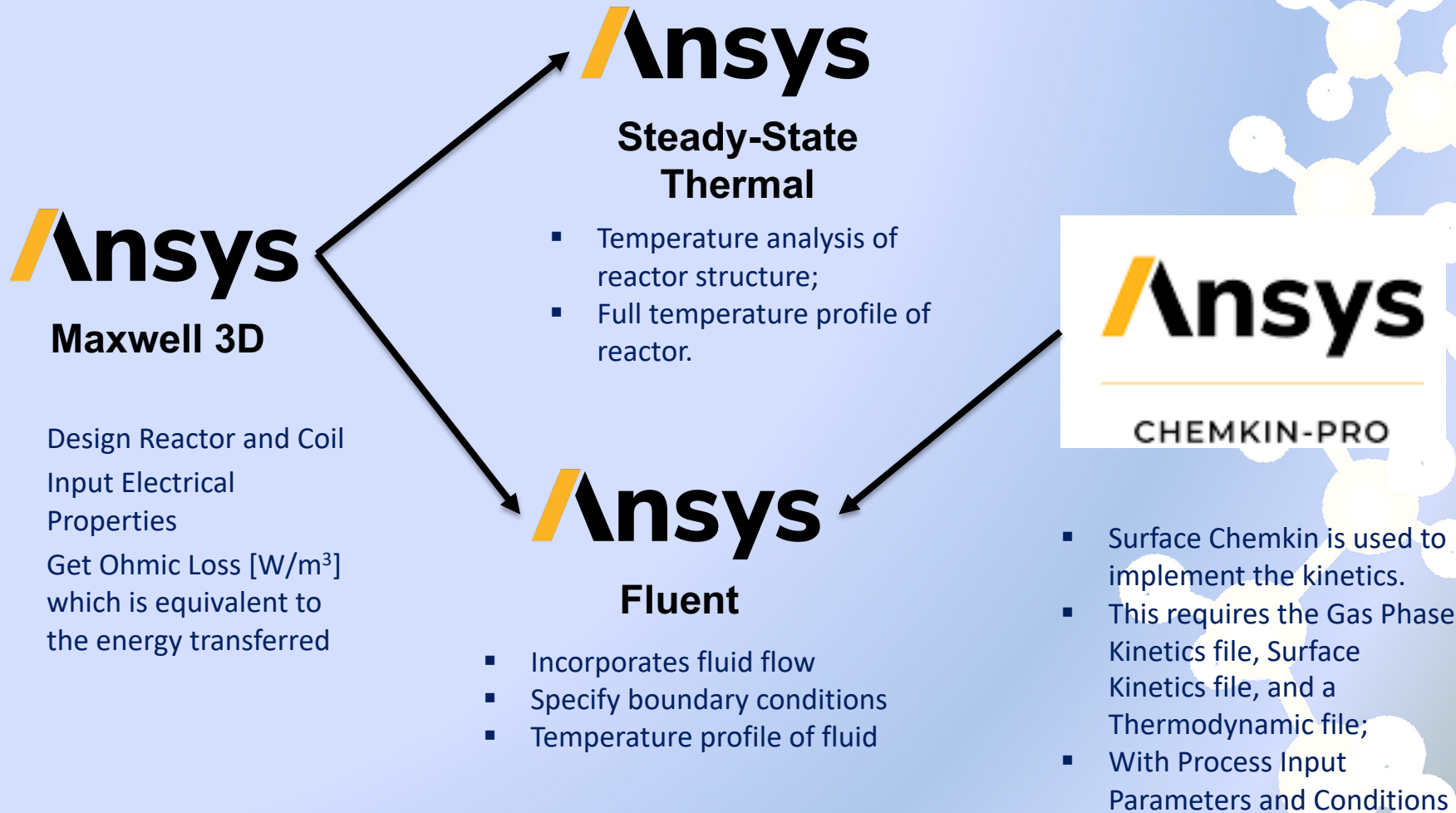- Controlled frequency electromagnetic heating,

- Ultrasonic vaporization.

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Induction Heating of Endothermic Reactors

Reactants and Inert Species →

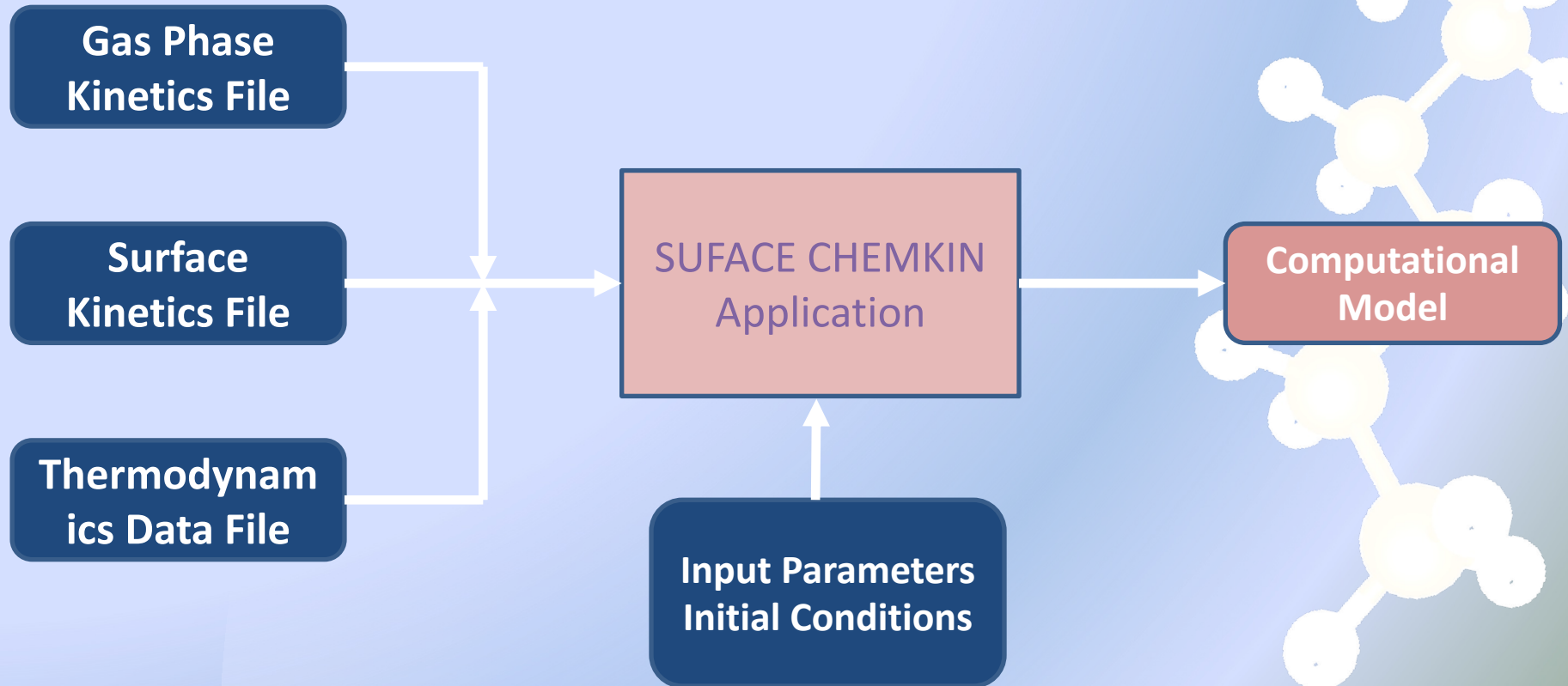Products →

Electrical Input from Renewable Grid ↑
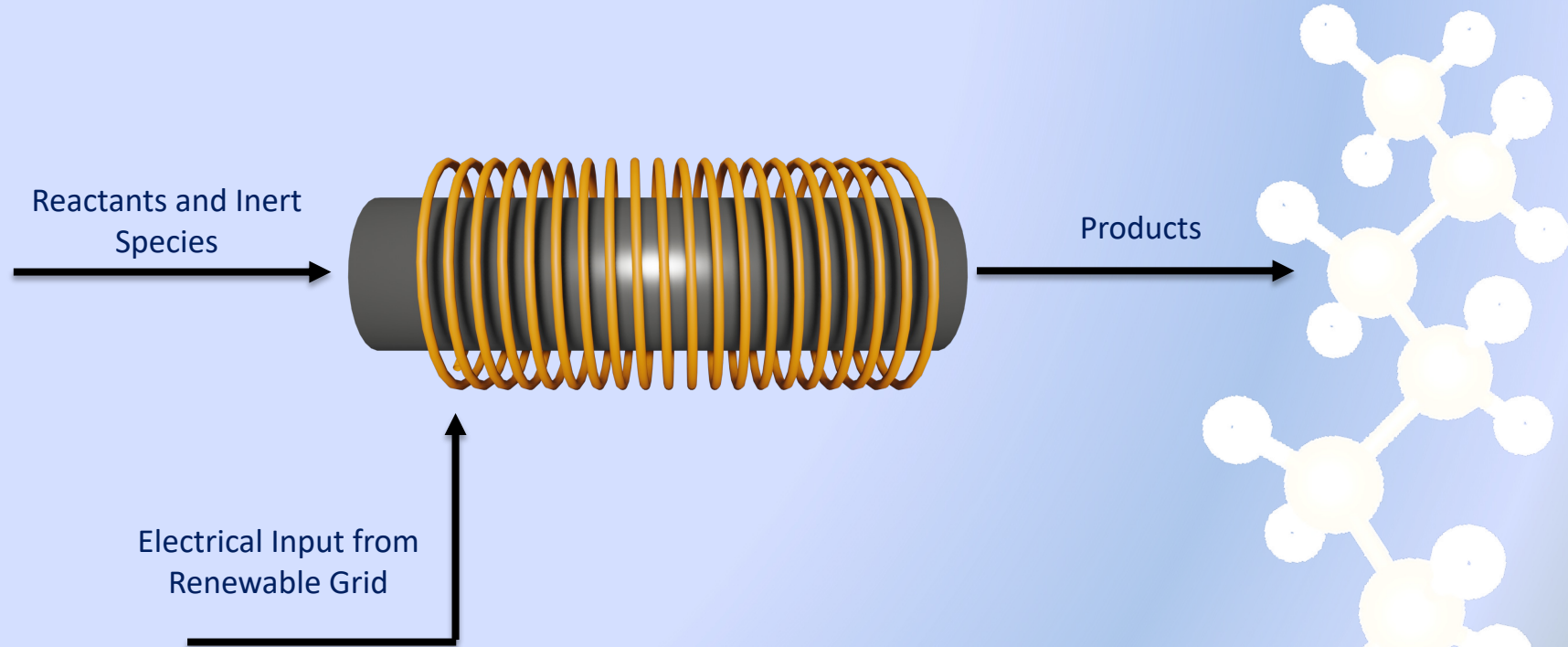
- Magnetic Field from Coil
  - Induces Eddy Currents
  - Material Resistivity
  - Heat Generation

- Electricity Input
  - Sourced from renewable power grid
  - Eliminates reliance on GHG producing fuel

Current Modeling with ANSYS Packages

∧nsys
Maxwell 3D

- Design Reactor and Coil
- Input Electrical Properties
- Get Ohmic Loss [W/m^3] which is equivalent to the energy transferred

∧nsys
Steady-State Thermal

- Temperature analysis of reactor structure;
- Full temperature profile of reactor.

∧nsys
Fluent

- Incorporates fluid flow
- Specify boundary conditions
- Temperature profile of fluid

∧nsys
CHEMKIN-PRO

- Surface Chemkin is used to implement the kinetics.
- This requires the Gas Phase Kinetics file, Surface Kinetics file, and a Thermodynamic file;
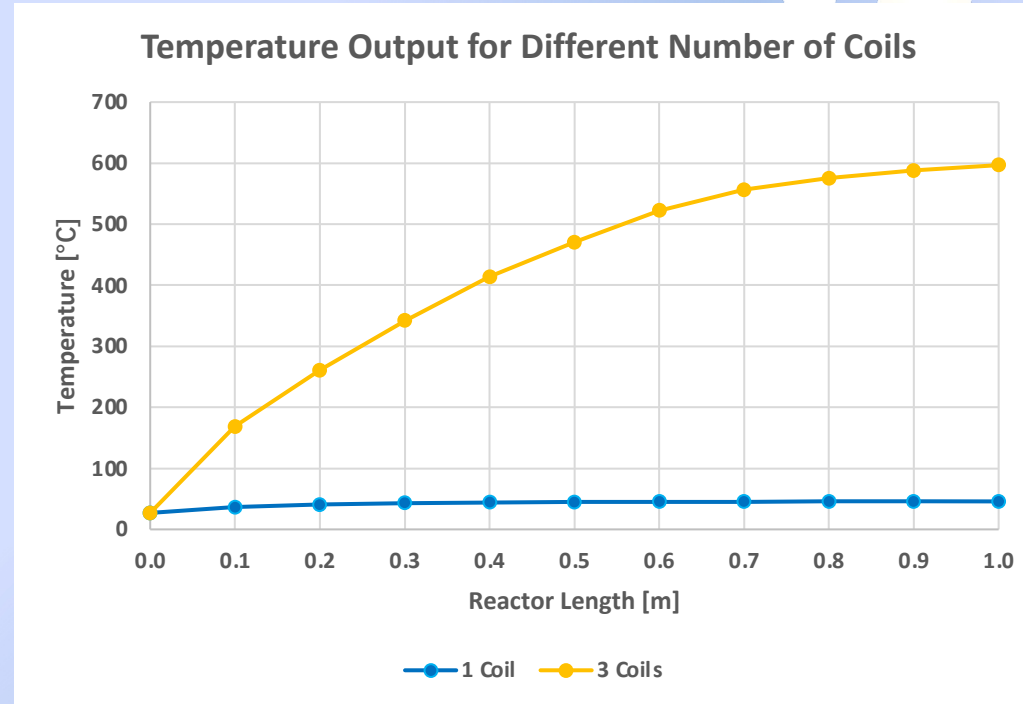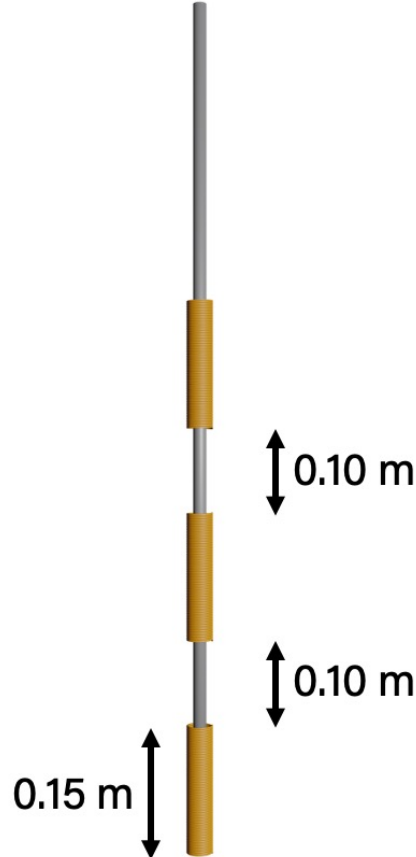- With Process Input Parameters and Conditions

Kinetic Description implemented in ANSYS CHEMKIN-PRO

- Three files are required for Surface Chemkin-Pro to model the reaction kinetics.

Gas Phase Kinetics File		
Surface Kinetics File	SUFACE CHEMKIN Application	Computational Model
Thermodynamics Data File		
	Input Parameters Initial Conditions	

Precise Temperature Control by Number of Coils

Temperature Output for Different Number of Coils

- Maximum outlet temperature (1 coil) = $46°C$
- Maximum outlet temperature (3 coil) = $597°C$

Outlet Temperature is a Function of Current and Frequency

For a Frequency of 10 kHz

For a Current of 10 A
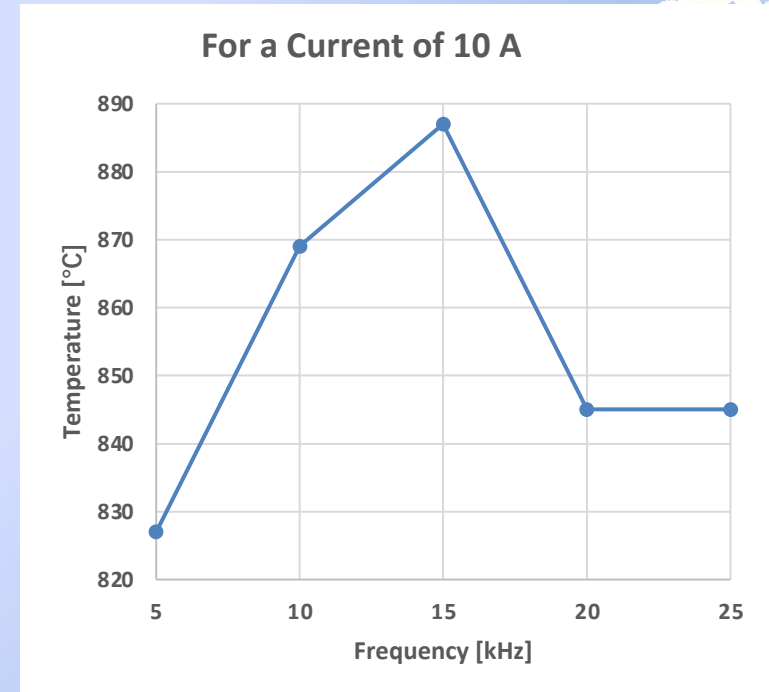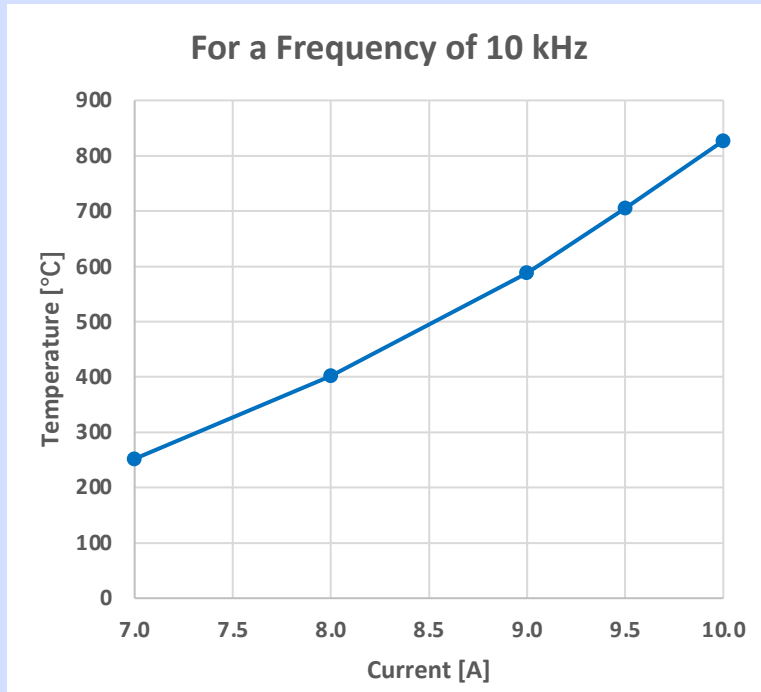
- For a given geometry at a fixed frequency, the increase in current increases the output temperature.

- However, an increase in the Frequency has a maximum. This is attributed to the Skin Depth Penetration Effect (high frequencies have thin penetration depths, therefore, there's an optimum frequency).

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Current Reactor Design

- Feed Stream Conditions:
  - Mass flow rate = 0.15 g/s
  - Inlet volumetric flow rate = 7.5 L/min

- Coil Conditions:
  - Current = 10 A
  - Frequency = 10 kHz
  - Coil radius = 1.0 mm
  - Coil height = 0.15 m

- Reactor Dimensions:
  - Inner radius = 4.1 mm
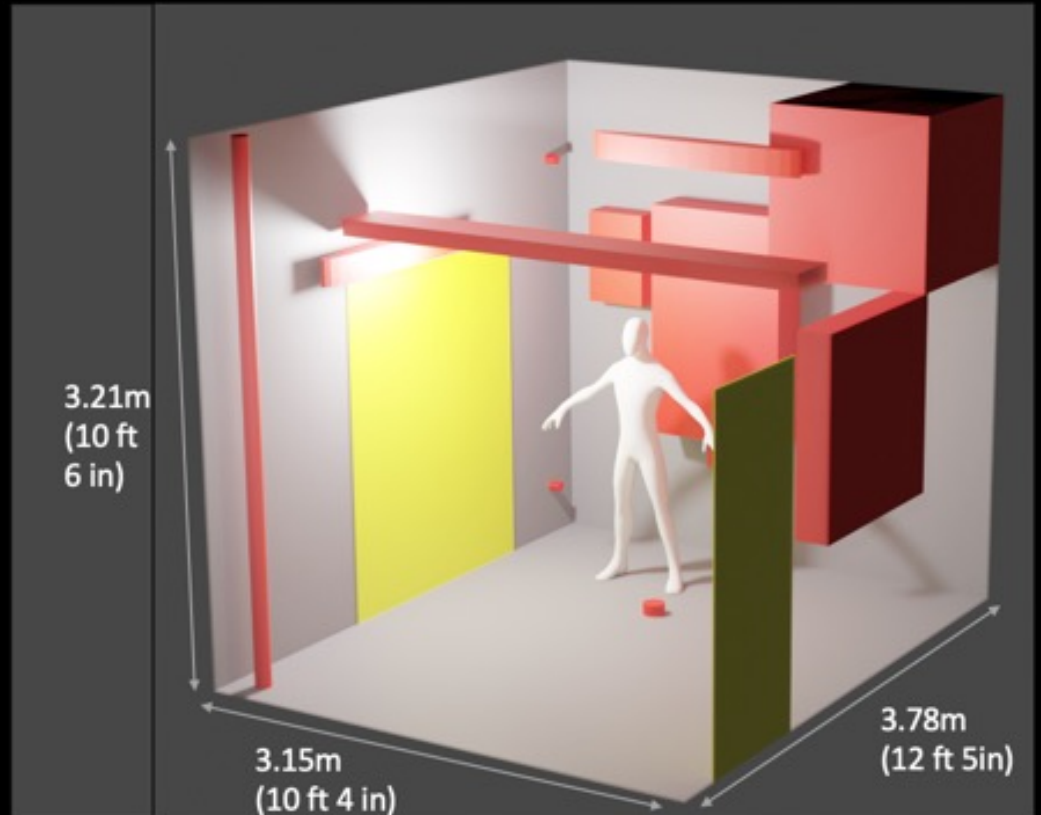 - Outer radius = 7.8 mm
  - Length = 1 m.
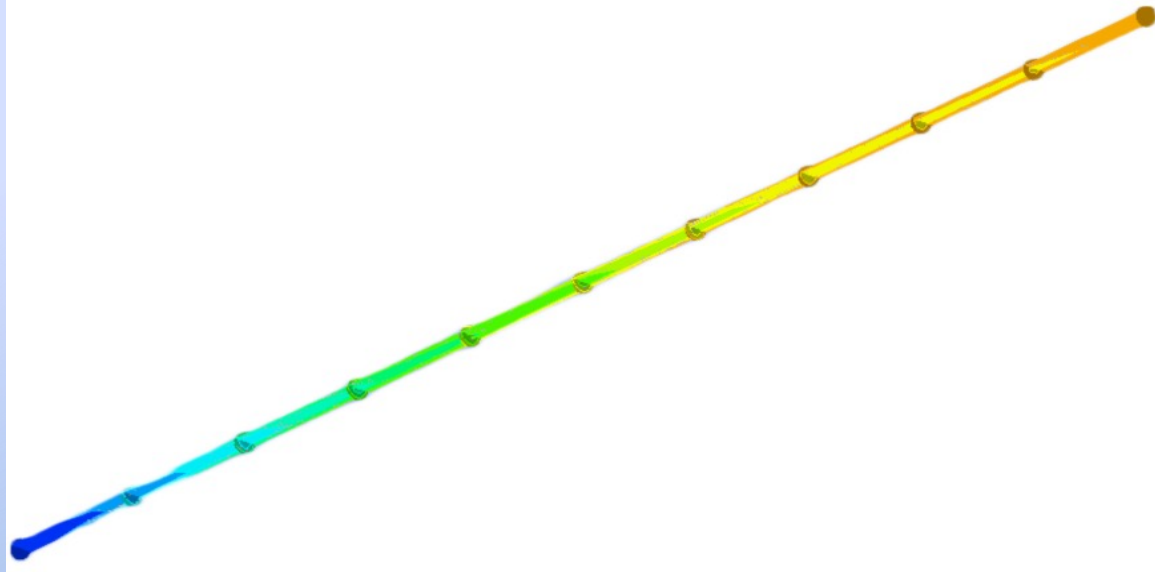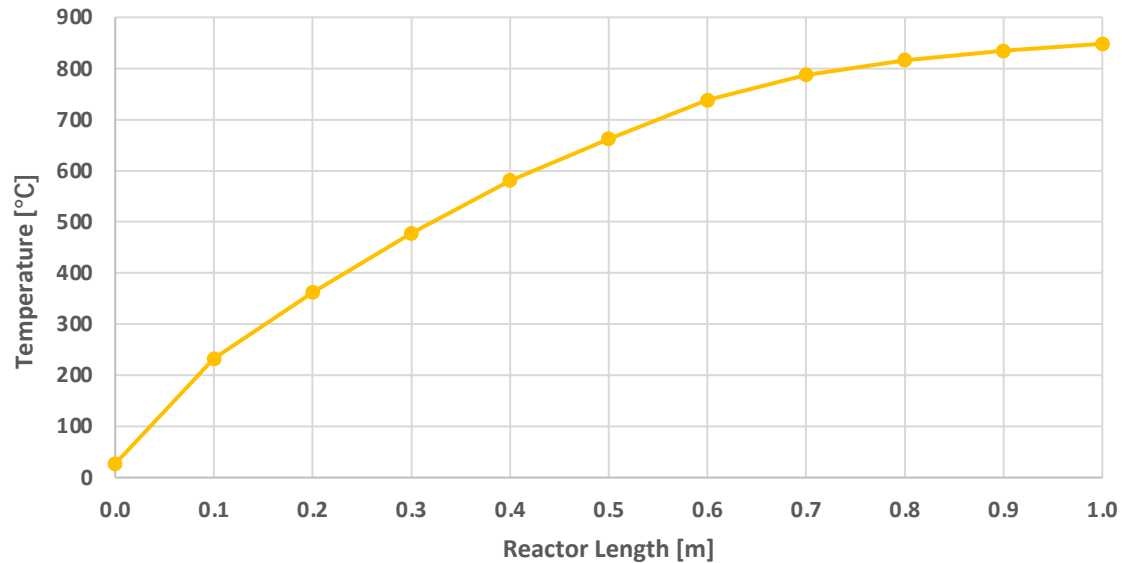
Laboratory Technology Demonstration

Zucrow Lab

- 3D model of the room that is going to house the reactor
  - Red objects are immovable objects
  - Yellow objects are doors
  - The humanoid object is 5 foot 10 inches tall
- The 3D model was created in Blender

3.21m (10 ft 6 in)

3.15m (10 ft 4 in)

3.78m (12 ft 5in)

The Future of the Chemical Industry in a Net-Zero CO_2 World

- Industrial Decarbonization is critical to our efforts to achieve a carbon-neutral economy by 2050.

- As carbon-free electricity becomes more available and lower in cost,

- Process electrification especially for heating and for electrolysis will prove to be an economically competitive decarbonization strategy.

- Endothermic Reaction Heat Supply does not need to be limited to resistive heating as an electrifying mechanism. Inductive heating is also available as a potential solution.

Thank You

- Questions?