On-Demand Reactivity Enhancement to Enable Advanced Low Temperature Natural Gas Internal Combustion Engines

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Project ID: ft086

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Project Overview

Timeline

Project Start Date: 5/1/2018

Project End Date: 6/30/2021

Percent Complete: 25%

Budget

Total Project Funding:

DOE Share = $1,102,367

Contractor = $294,358

FY 2018 Funding = $25,334

FY 2019 Funding* = $173,599

* through 5/1/2019

Barriers

Lean/Dilute Natural Gas Combustion:

- Methane is a highly stable fuel - requires high ignition energy in lean/dilute conditions
- Cannot be easily used in any advanced compression ignition modes
- Poor low temperature oxidation of methane in existing exhaust aftertreatment catalysts

Partners

University of Minnesota

Johnson Matthey
Inspiring science, enhancing life

Carnegie Mellon University
Pittsburgh Pennsylvania 1900

Relevance/Objectives

- **Overall Objectives**

  Demonstrate ≥10% indicated efficiency improvement compared to state-of-the-art lean-burn Natural Gas (NG) dedicated spark ignition (DSI) engine by using fuel pretreatment by oxidative coupling of methane (OCM)

- **Objectives in this Period**

  Experiments and modeling to demonstrate capability of the short contact time reactor to achieve desired OCM conversion and selectivity

Milestones

Tasks/Milestones	Budget Period 1					Budget Period 2				Budget Period 3			
	FY18		FY19				FY20				FY21		
	Q1	Q2	Q3	Q4	Q5	Q6	Q7	Q8	Q9	Q10	Q11	Q12	Q13
1. Perform Bench-Scale Short-Contact Reactor Studies (UMN)		M1.1			M1.2, D1.1							M1.3	
2. Develop Highly Effective OCM Catalysts and Wash-Coat Technology (JM)			M2.1				M2.2						
3. Design and Characterize Single Cylinder Engine and Reactor System (All)							M3.1	M3.2	M3.3, D3.1		M3.4		M3.5
4. Use CFD Modeling to Guide Reactor Design and LTCI Engine Combustion (CMU)			M4.1		M4.2			M4.3			M4.4	M4.5	
5. Prove NG Engine Efficiency Gains Through 1-D Modeling											M5.1		M5.2

Completed | To be Completed | Current

Approach

- Oxidative coupling of methane introduction
  - Proposed industrially to produce ethylene from natural gas
  - Low conversion and potential for unselective oxidation
  - Oxygen utilization is a challenge - leads to unselective oxidation
  - Could be used to enhance the reactivity of natural gas for engines

<u>Selective oxidation</u>

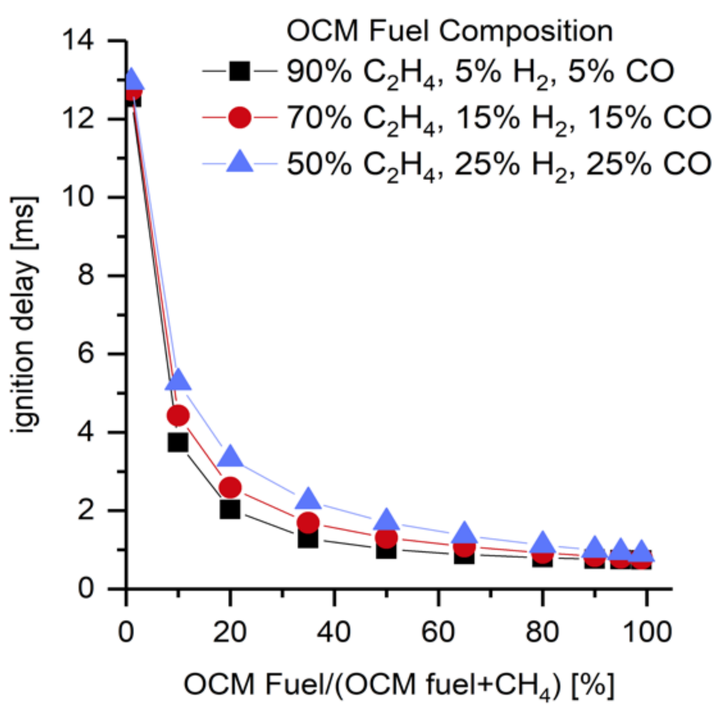
$$2CH_4 + \frac{1}{2}O_2 \rightarrow C_2H_6 + H_2O$$

$$C_2H_6 + \frac{1}{2}O_2 \rightarrow C_2H_4 + H_2O$$

<u>Unselective oxidation</u>

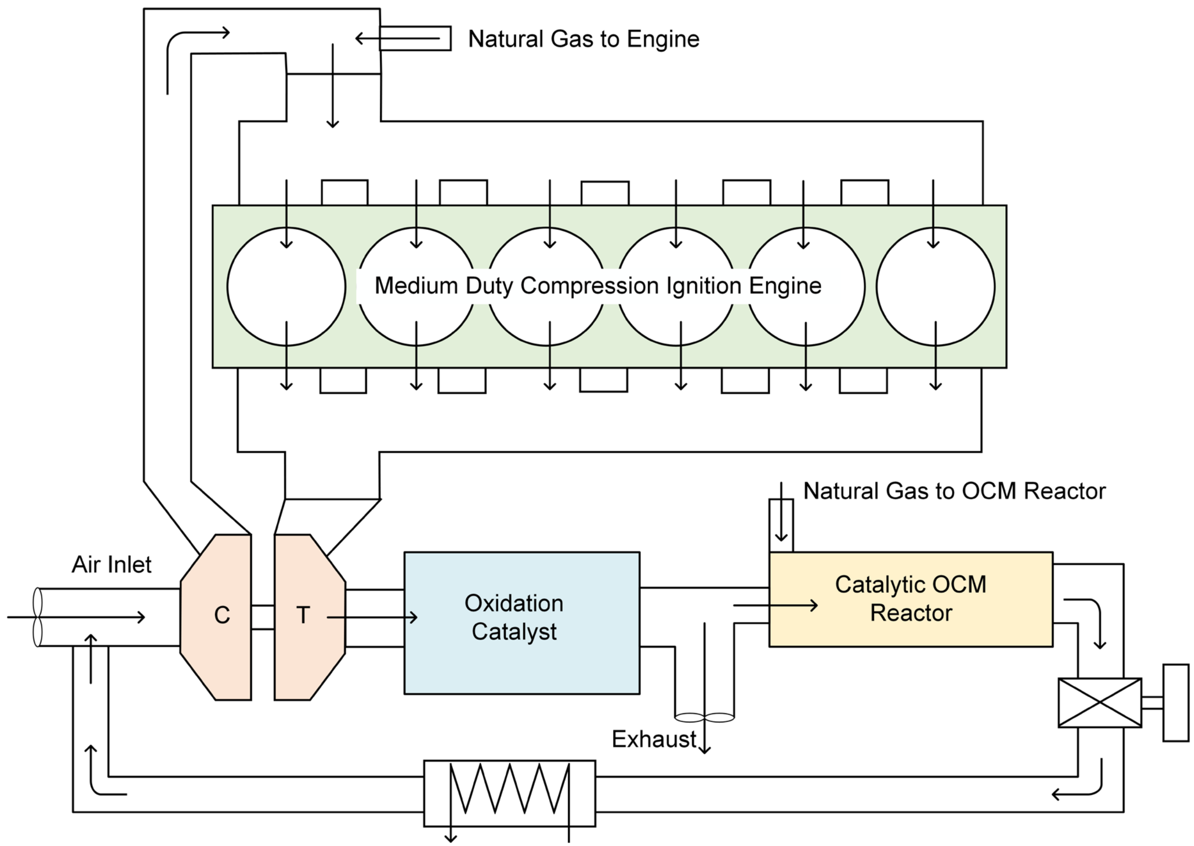
$$CH_4 + O_2 \rightarrow CO + H_2O + H_2$$

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O$$



Approach

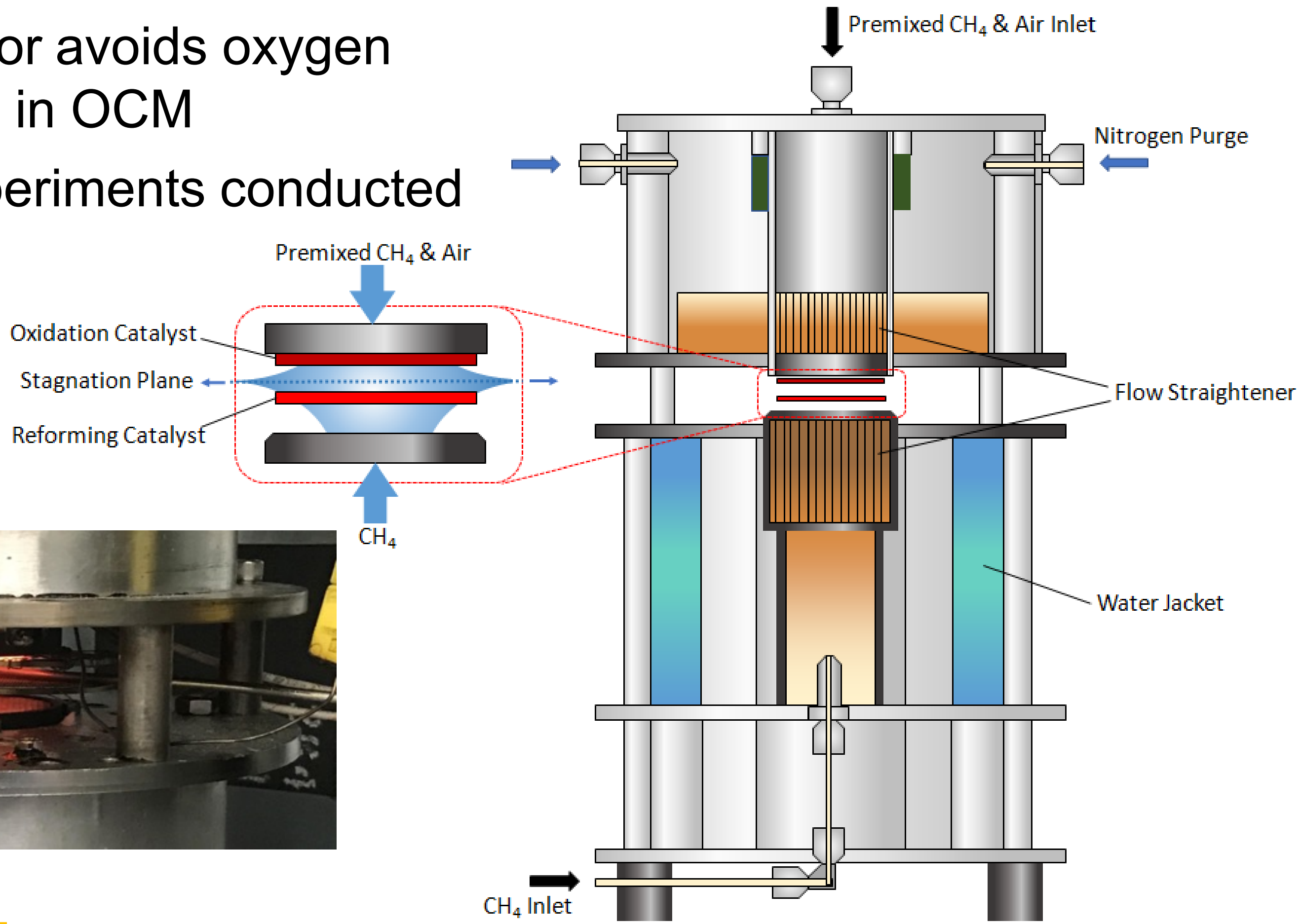
- Integrate OCM into a reformed exhaust gas recirculation scheme to enable advanced dilute combustion modes
- Use new short-contact time reactor to avoid oxygen utilization problems found in large-scale reactors



Technical Accomplishments

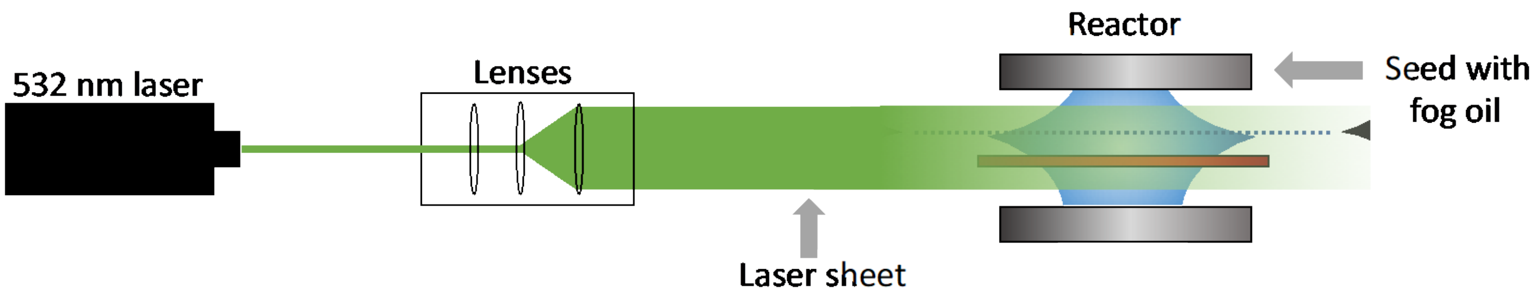
Task 1: Bench scale reactor completed and partial oxidation reforming catalyst tested

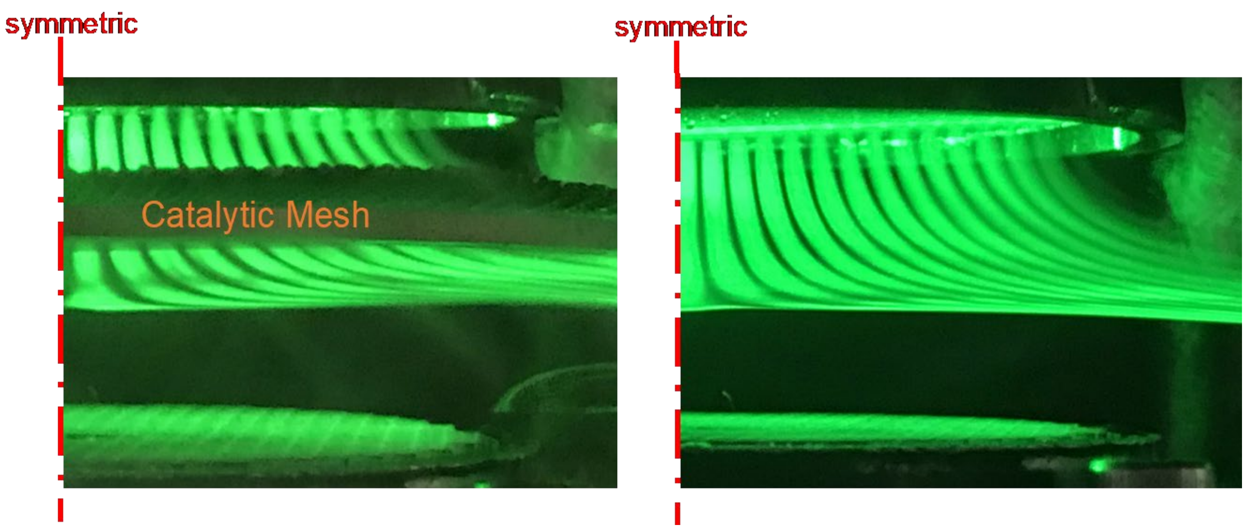
- Derived from previous work under NSF project
- Opposed flow reactor avoids oxygen distribution problem in OCM
- Partial oxidation experiments conducted



Technical Accomplishments

Task 1: Flow visualized to determine pressure drop through catalytic mesh





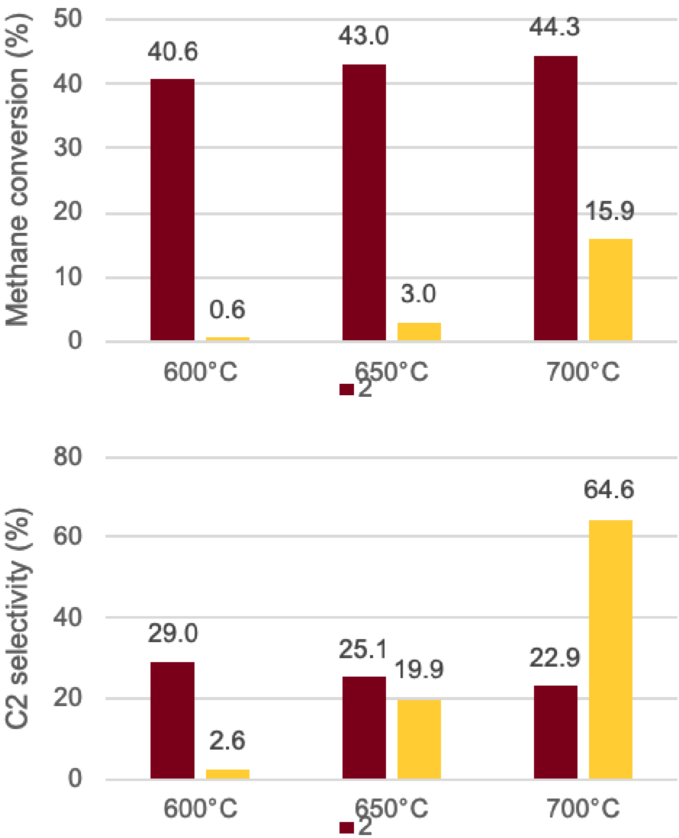
- Mesh did not affect the flow in the center region
- The flow direction was changed at the edge by the mesh

Technical Accomplishments

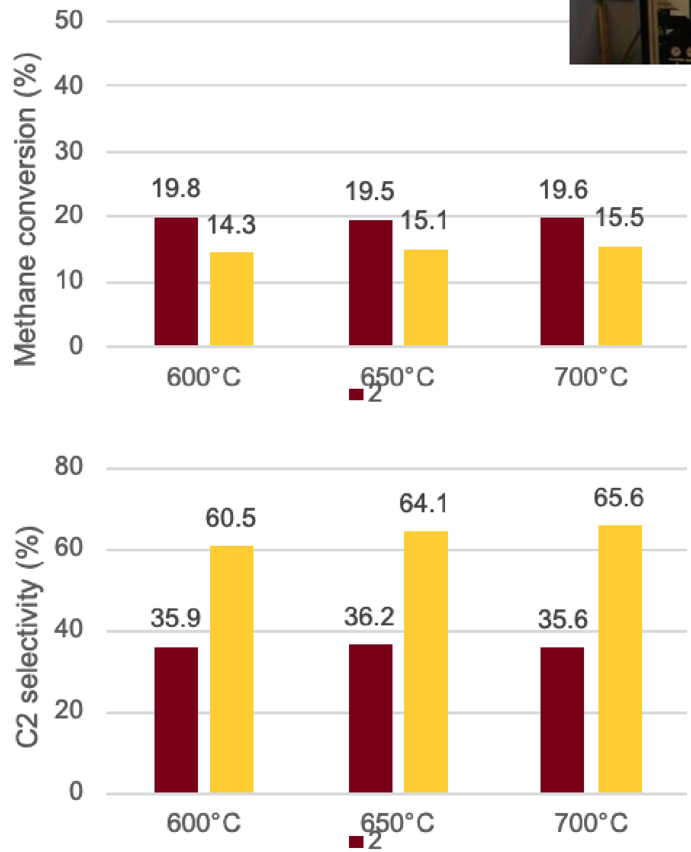
Task 2: Catalysts evaluated: integral micro-reactor

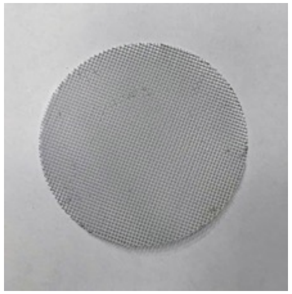
- Sr/La_2O_3 catalyst: better low temperature conversion
- Coated meshes provided for opposed flow reactor

LaSr/CaO



Sr/La_2O_3





Sr/La_2O_3 Mesh

Testing conditions
Space velocity 50,000 to 100,000 h^{-1}
CH_4/O_2 ratio from 2:1 to 6:1 molar
Temperature 600-700°C

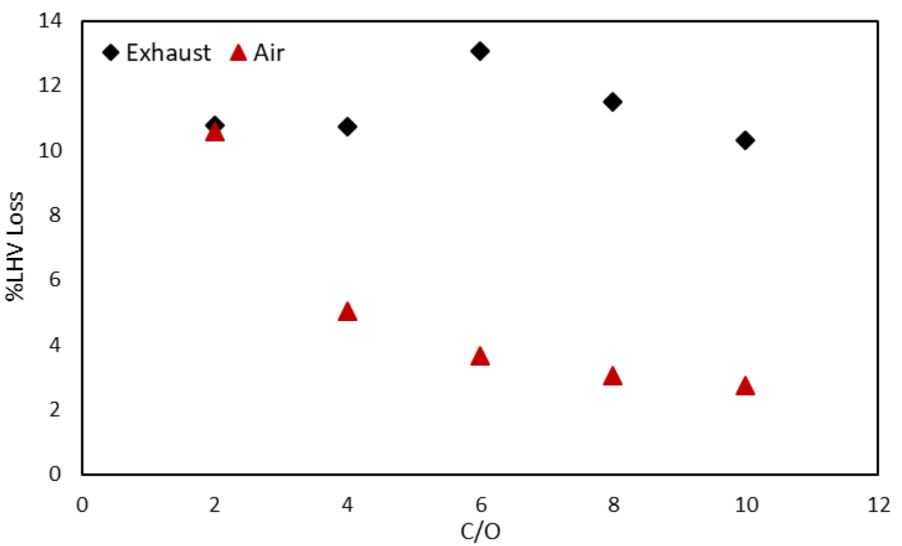
Presented here
Space velocity 50,000 h^{-1}
CH_4/O_2 ratio 2:1 or 6:1 molar
Temperature 600-700°C

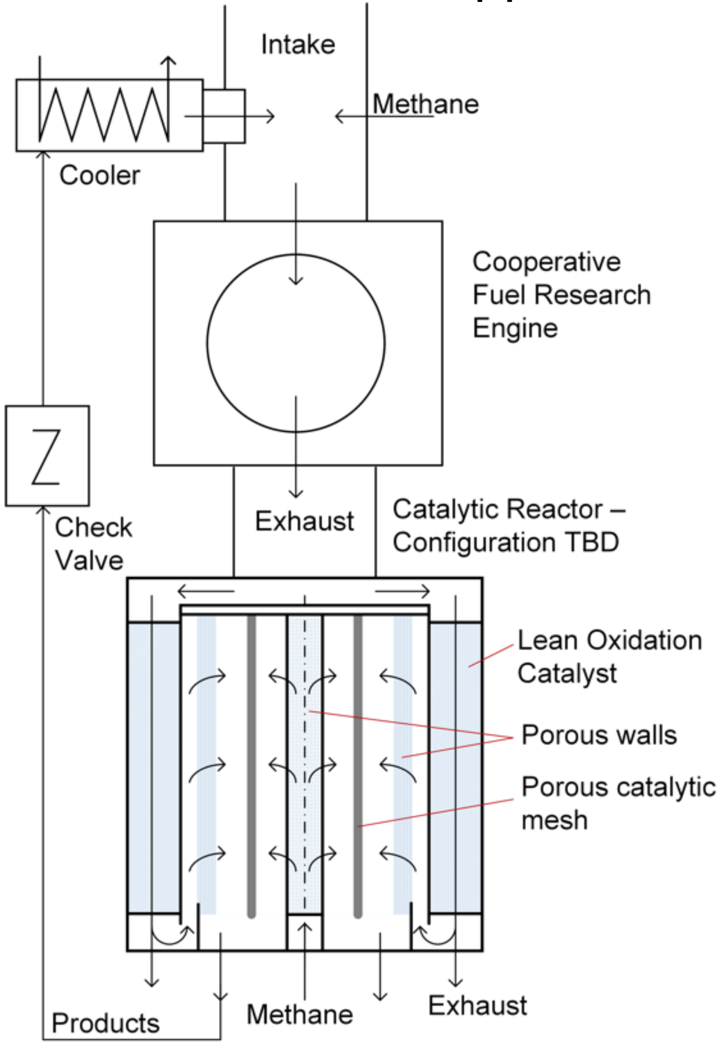
2:1 CH_4/O_2 lean
6:1 CH_4/O_2 rich

Technical Accomplishments

Task 3: Two approaches for engine scale reactor considered: opposed flow and traditional monolith reactors

- CFR reactor set up to study autoignition
- Short contact, better O_2 utilization
- Residence time may not be sufficient
- Rich reaction with fresh air may be required to mitigate LHV loss



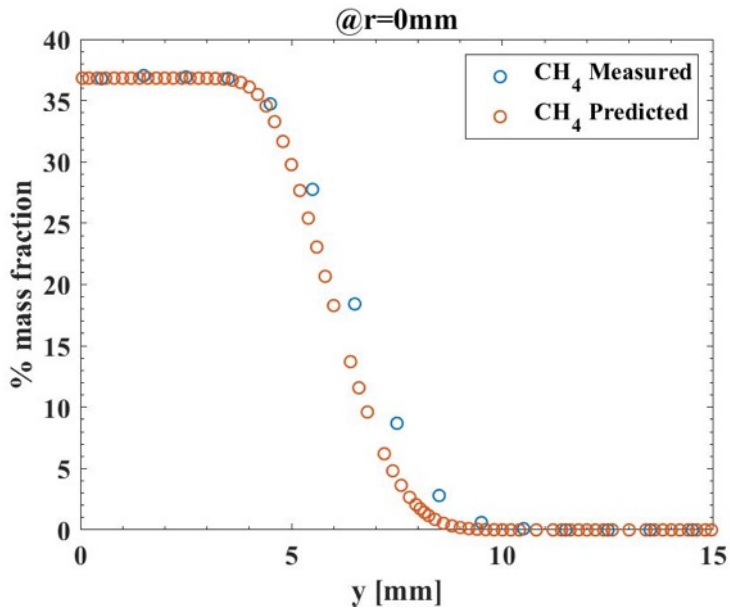


Technical Accomplishments

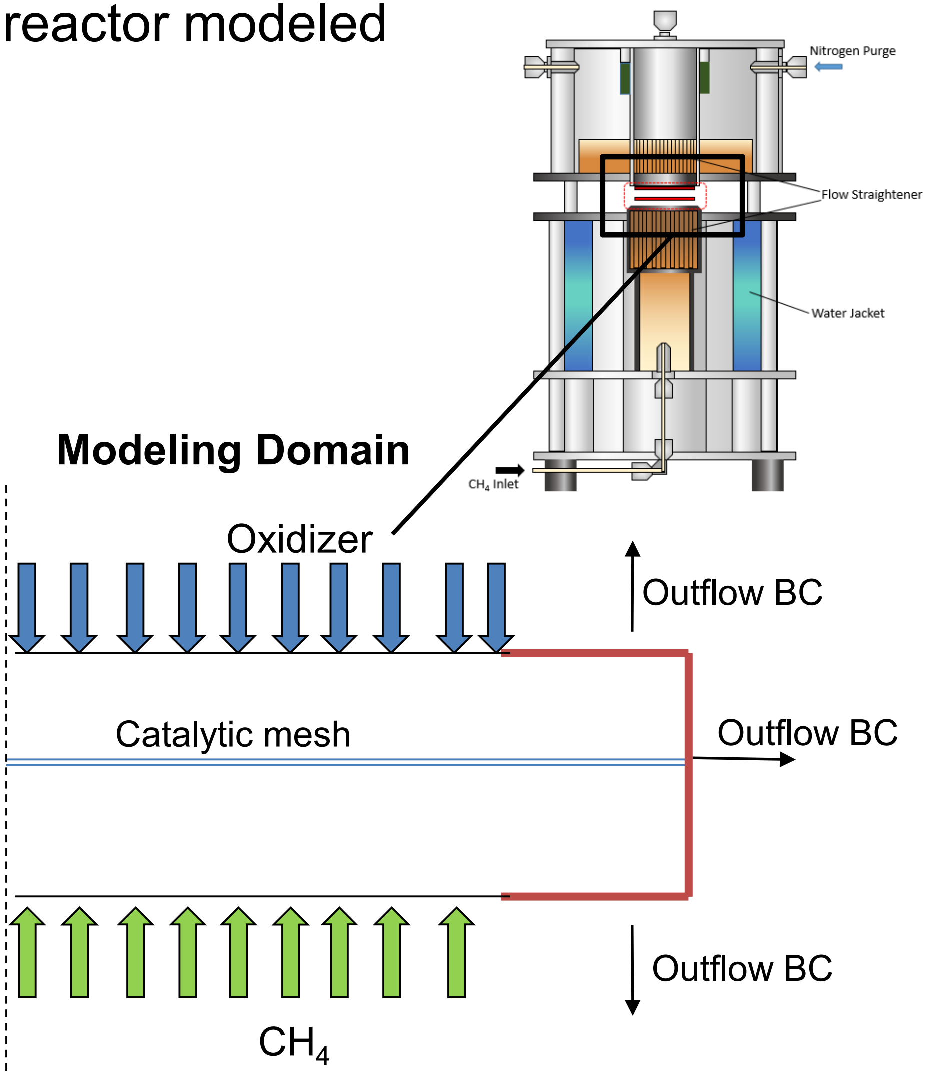
Task 4: Bench-scale short contact time reactor modeled in Fluent and Converge

- Reacting and non-reacting cases modeled
- Reacting cases use literature partial oxidation mechanism

Non-Reacting CH_4 Comparison



Modeling Domain

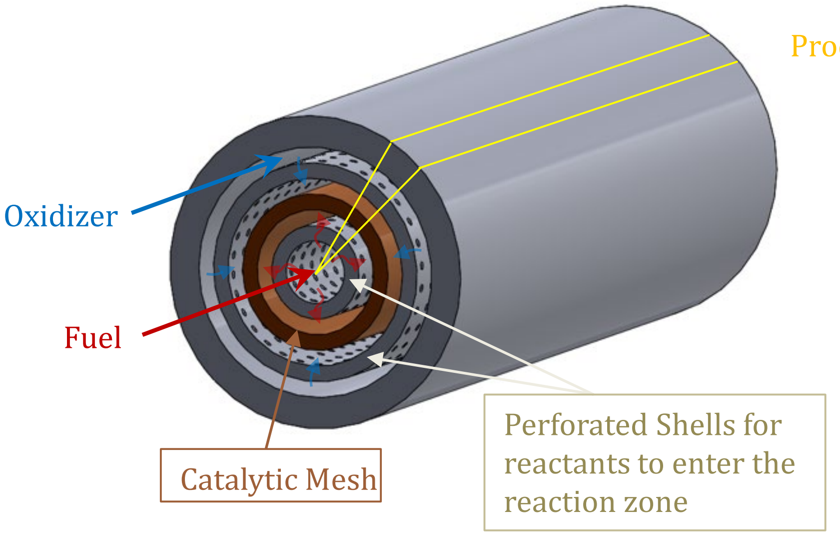


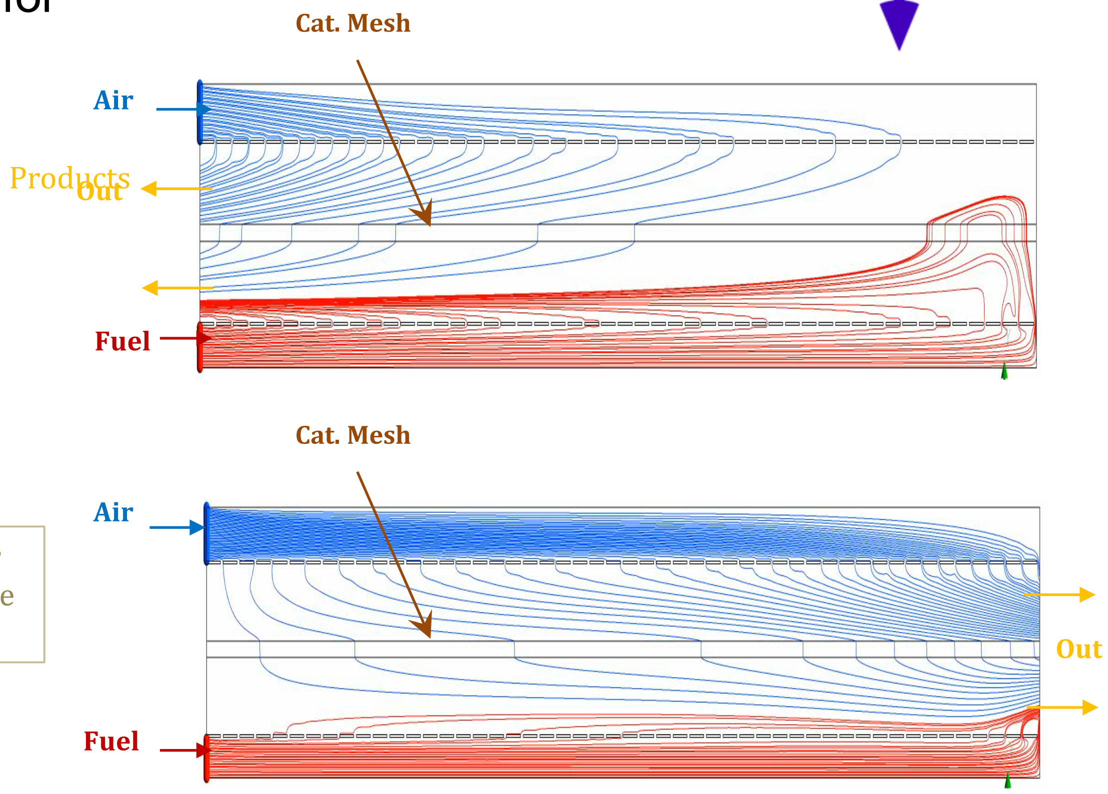
Technical Accomplishments

Task 4: Engine-scale CFD model constructed in Converge

- Gas distribution studied
- Completed non-reacting cases
- Waiting for validation of kinetics for Sr/La_2O_3 catalyst

Computational Domain





Response to Previous Comments

This is the first year the project has been presented

Partnerships/Collaborations

Partners

Johnson Matthey
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- OCM catalyst research and development
- Catalyst wash-coating for bench and engine-scale reactors

Carnegie Mellon University
Pittsburgh Pennsylvania 1900

- Computational fluid dynamics of reactors and engines
- Supporting low-dimensional engine modeling efforts

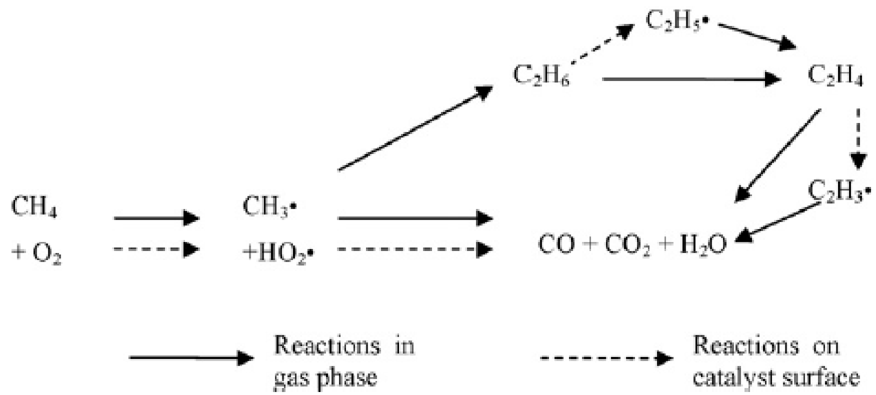
Collaborators

Ghent University

- Provided OCM kinetics for modeling efforts

Remaining Challenges and Barriers

- Achieving acceptable conversion and selectivity over provided OCM catalysts using bench-scale reactor
- OCM reaction is more difficult than reforming:
  - Deactivation at T > 900ºC
  - Low activity at T < 600ºC
- Determine appropriate OCM stoichiometry to limit heating value loss – balance with engine efficiency gain
- Short contact time may not achieve sufficient conversion of methane – OCM consists of series of gas + surface phase reactions[1]



[1] Sun, Jianjun, Joris W Thybaut, and Guy B Marin. "Microkinetics of Methane Oxidative Coupling." *Catalysis Today* 137 (2008): 90–102.

Proposed Future Research

To be completed in FY19/FY20(Q1):

- Characterize OCM in bench scale reactor, achieving >10% conversion and >20% selectivity to C_2 products
- Determine direction for engine-scale reactor design, short contact or monolith
- Configure single-cylinder engine test cell to begin engine reactivity studies

Future*:

- Should project be successful, the project team will seek opportunities to integrate with full scale natural gas engine

*Any proposed future work is subject to change based on funding levels

Thank You

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