



Electromagnetic Energy-Assisted Approaches to Convert Fossil Fuels to Low Cost Hydrogen

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PI: Johannes G. van der Watt (University of North Dakota)

PM: Heather Hunter (DOE/NETL)

Program Overview

Opportunity: Produce H₂ from methane rich sources **without CO₂** formation

Problem: Carbon deposition and catalyst deactivation

- $\text{CH}_4(\text{g}) \xrightarrow{\text{catalyst}} \text{C}(\text{s}) + 2\text{H}_2(\text{g})$
- Catalyst loss – either reactivity or physical loss

Goal: Extend H₂ production through targeted catalyst regeneration

Solution approach: Use an in-situ electromagnetic (EM) energy-assisted mechanism to regenerate catalysts

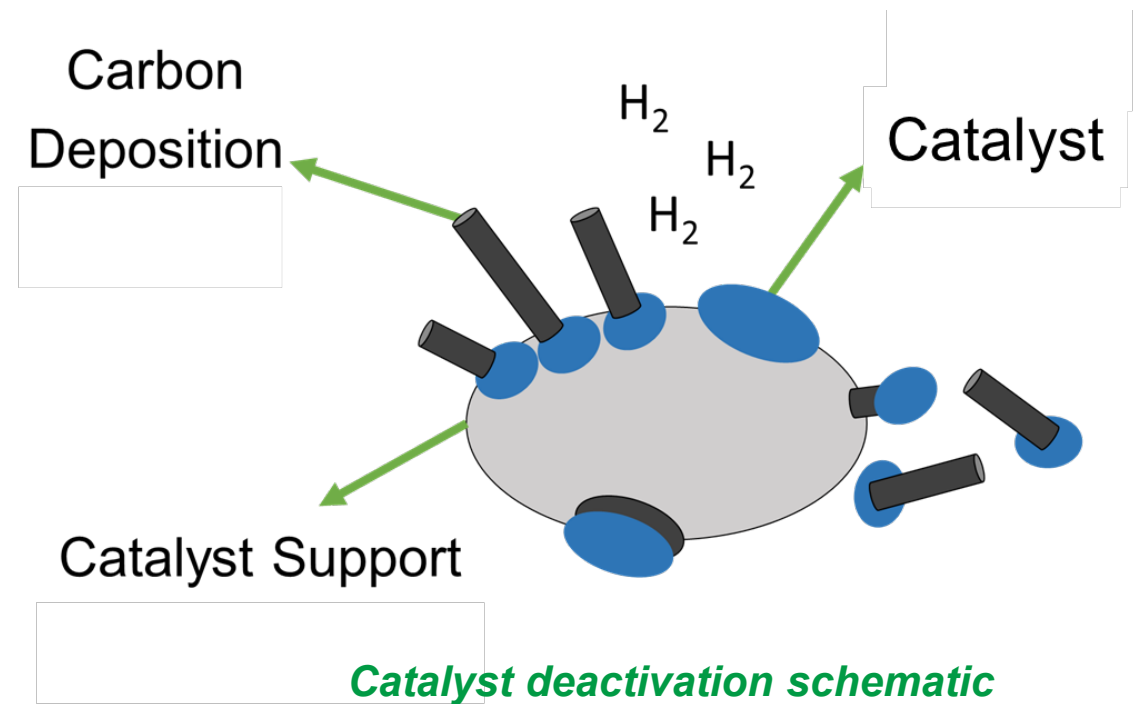
Potential Significance – Catalyst Regeneration

- **Reduce** overall catalyst replenishment cost
- **Purer** carbon products – less catalyst poisoning
- **Low-cost & CO₂-free** process for H₂ production
- Enable **wider adoption** of H₂-related technologies from fossil resources
- **Applicable** to other catalyst-based processes – coking issues

Background – CH₄ Decomposition

Thermo-catalytic decomposition of CH₄

- $\text{CH}_4(\text{g}) \rightarrow \text{C}(\text{s}) + 2\text{H}_2(\text{g}) \Delta H_{\text{rxn}} = 37 \text{ kJ/mol-H}_2$
 - Electrolysis **285 kJ/mol-H₂**
 - Steam methane reforming (SMR) **41 kJ/mol-H₂**
- Typically conducted at **500 – 800°C**
 - Typical catalysts include: Transition metals, even C
 - Without catalyst: 1200°C requirement



Background – Catalyst “Cleaning” Approaches

- Physical removal – attrition
 - C remains same
 - Potential catalyst loss & impure catalysts with C
- Chemical removal – Combust/Gasify carbonaceous deposits
 - *Combustion: $O_2 + C \rightarrow CO_2$*
 - *Gasification: $H_2 + C \rightarrow \text{“}CH_4\text{”}$ and $H_2O + C \rightarrow CO + H_2$*
 - *Chemical – Acid leaching to remove and recover metal catalysts*

Approach

Technique:

- In-situ carbon removal using an Electromagnet (EM) Energy-Assisted Thermocatalytic Process
- Mild process applicable to catalyzed reaction temperatures
- Target periodic cleaning
- Mechanism – chemical and physical C removal

How:

- *Conduct proof-of-concept experiments*
- *Supplement with computational modeling*
- *Assess large-scale applicability of approach (integrated setups)*



Technical Summary

Task 1 – PMP and Technology Maturation Plan

Task 2 – Catalyst Preparation and Performance Testing

Task 3 – Computational Fluid Dynamics (CFD) Modeling of Conversion System

Task 4 – Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

Task 5 – Component Identification for Future Work

Task 6 – PMP (North Dakota Industrial Commission - NDIC)

Task 7 – Evaluation and Characterization of Carbon Products

Task 8 – Process Model of Greener H₂ Production System



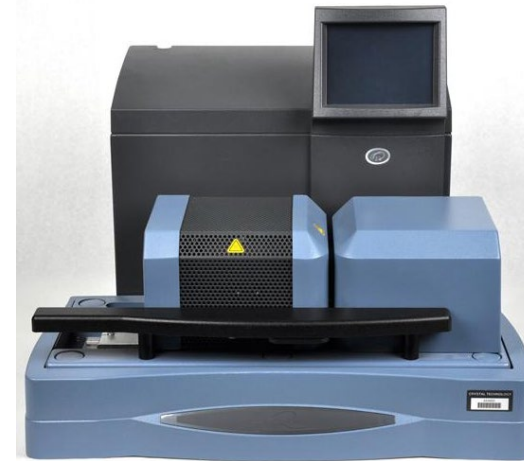
***Extended
Scope***



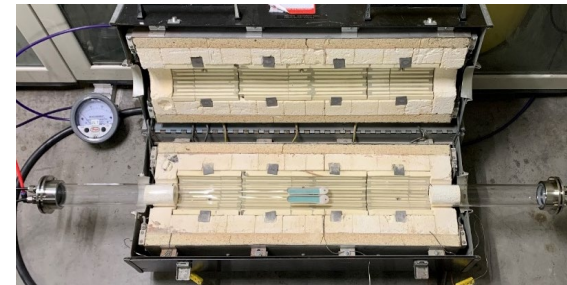
Task 2: Catalyst Preparation and Performance Testing

Subtask 2.1 – Catalyst Preparation

- Catalyst/Support structures identification
- Combinations of different supports tested
 - SiO_2 , C, Al_2O_3 , and Aerogel Supports
- Catalysts
 - Ni, C and Fe
- Identified promising Ni- SiO_2 catalyst
 - Added Cu-promotor – disperses Ni more evenly



TGA Tests (TA SDT Q600)



2-inch
tube furnace



Fluidized/Fixed bed
testing in 3/8-inch
tube reactor

Task 2: Catalyst Preparation and Performance Testing

Kinetics for CFD

- Conduct kinetic study using TGA
- Ensure sufficient gas flow – mitigate external mass transfer limitations (**initially**)
- Initial tests – determine optimum flow rate & sample mass
 - 500-650 °C & 30%-50% CH₄ (N₂ balance)
 - Eventually use CFD to develop geometries complementing EM-assisted process



Pre- and post-test with
Ni-SiO₂ catalyst

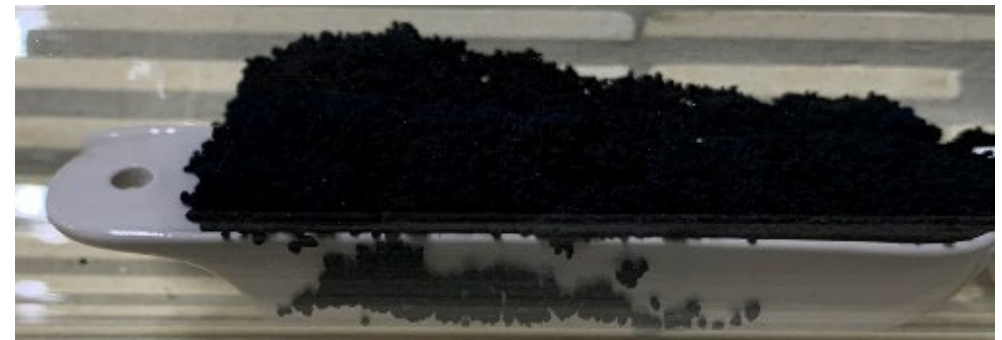
Task 2: Catalyst Preparation and Performance Testing

Subtask 2.2 – Initial High Temperature Catalyst Performance Testing

- TGA testing scaled up to tube furnace
- Tube furnace – easy integration EM Energy-assisted thermocatalytic process
- Just under 70% CH₄ conversion at 650°C
- Key result - moldable catalyst with suitable performance



Pre-test material



Post-test material with carbon buildup

Task 2: Catalyst Preparation and Performance Testing

- Structured catalyst testing
- Required flat surface for C-deposition and C-removal
- Decided on disk-shape (compatible with tube furnace)
- Placed perpendicular to gas flow
- Run short durations for C-growth
- Excessive run time – carbon growth destroys surface



Tube Furnace Testing - Structured

Task 3: Task 3 – Computational Fluid Dynamics (CFD) Modeling of Conversion System

Subtask 3.1 – Establish Baseline CFD and Kinetic Model

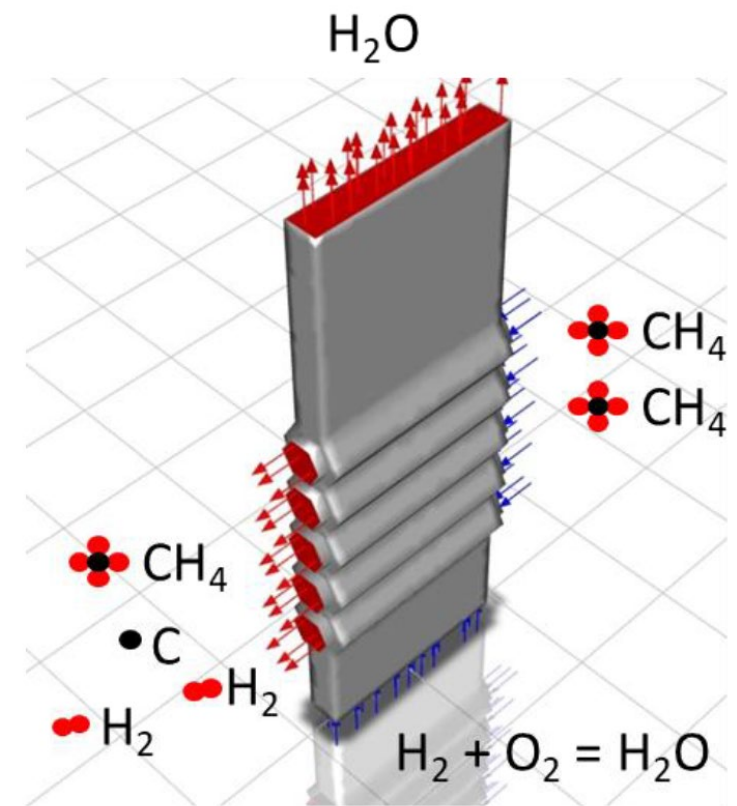
- Capture decomposition reaction behavior

Subtask 3.2 – Update CFD and Kinetic Model to Include Effects of the Electromagnetic Energy-Assisted Mechanism

- Assess geometries/setups enhancing removal technique

Results: 2D & 3D models

- Depicting zones of high C-deposition
- Temperature profiles (heat addition points)
- Locational aspects of EM Energy-Assisted Mechanism



Reaction Modeling

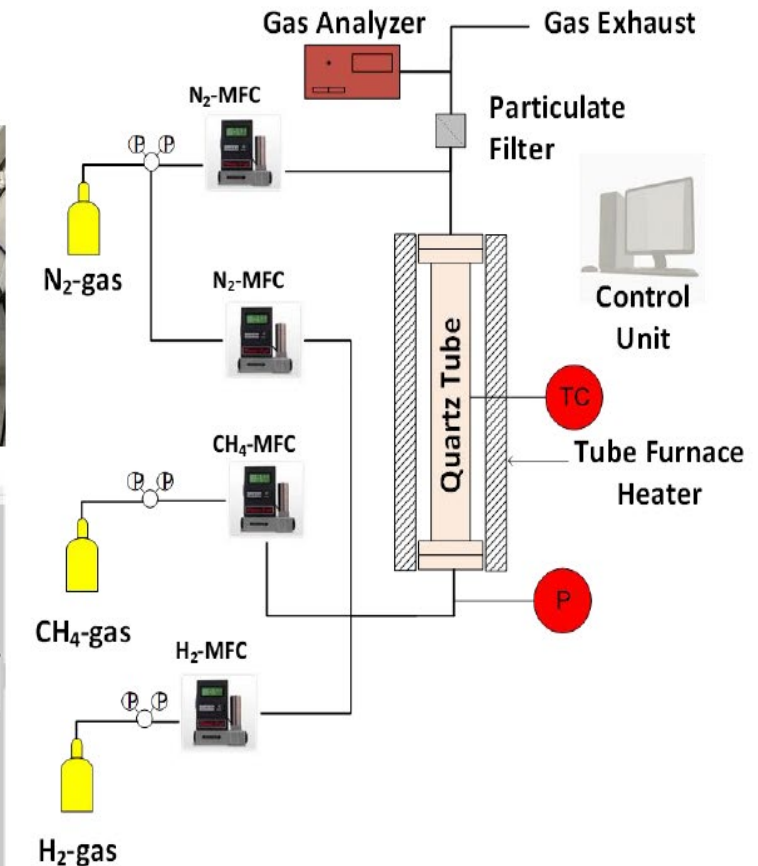
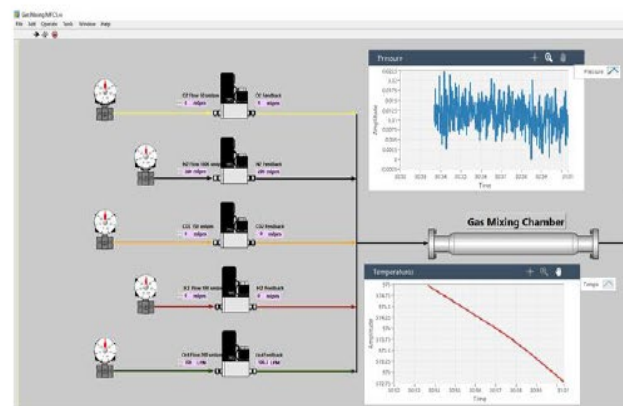
Task 4: Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

Series of experiments devoted to

- Decomposition testing
- Surface regeneration

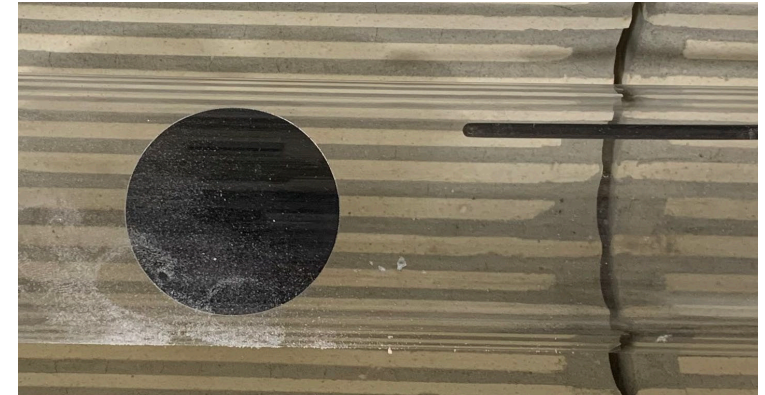
Assembled integrated test unit

- 2"-ID glass reactor
- Mass flow controllers
- Gas analyzer
- EM Energy-Assisted Module (not shown)



Task 4: Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

- Structured catalyst/support testing
- Initial tests failed (inset images)
 - Aggressive CH_4 decomposition (too long)
 - Refined binding technique
- Process refined: samples subjected to ~10 minutes of CH_4 decomposition
- Thin layer approach – lines up with CFD and viable C-removal technique



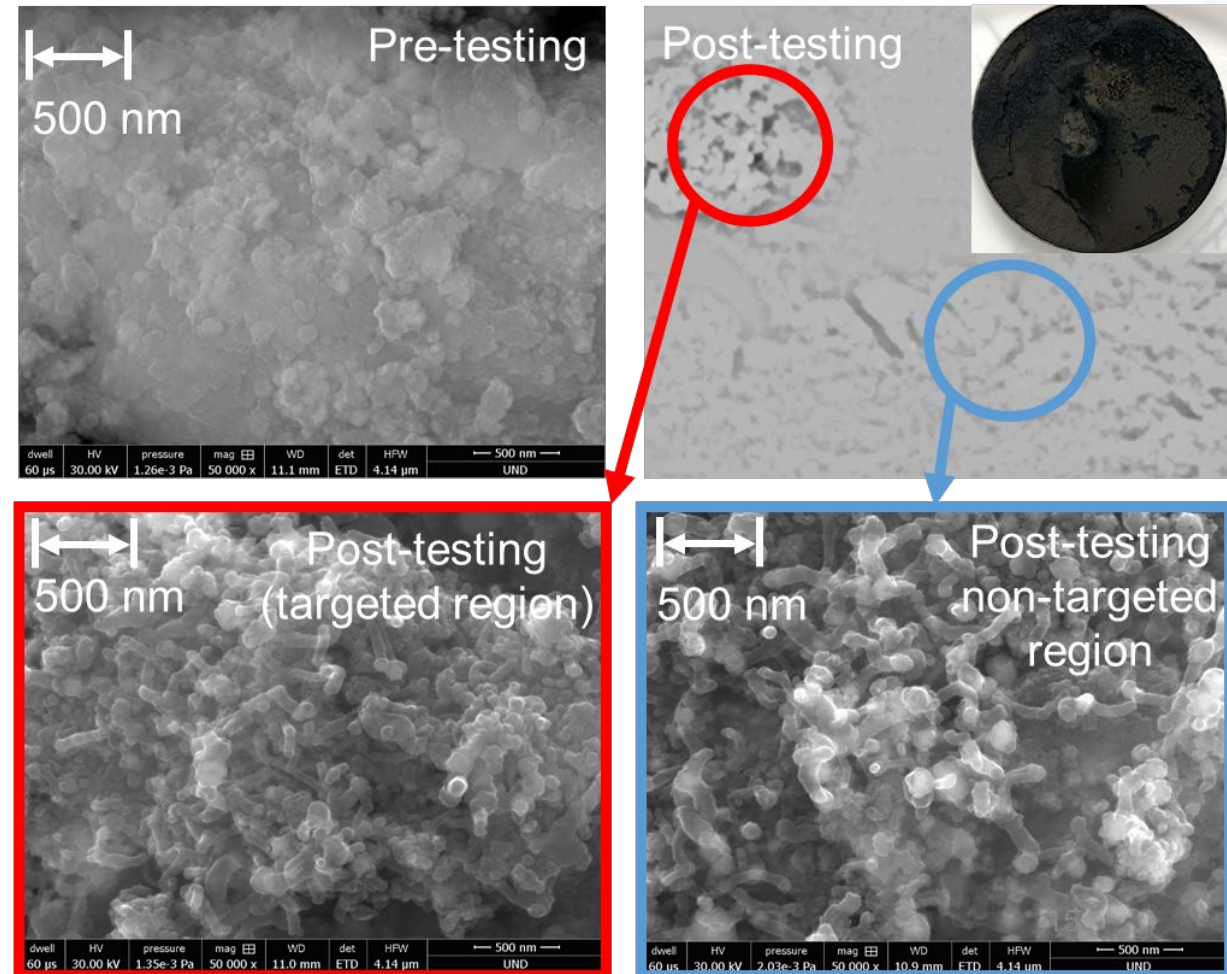
Pre-decomposition structured catalyst/support disk



Post-decomposition structured catalyst/support disk

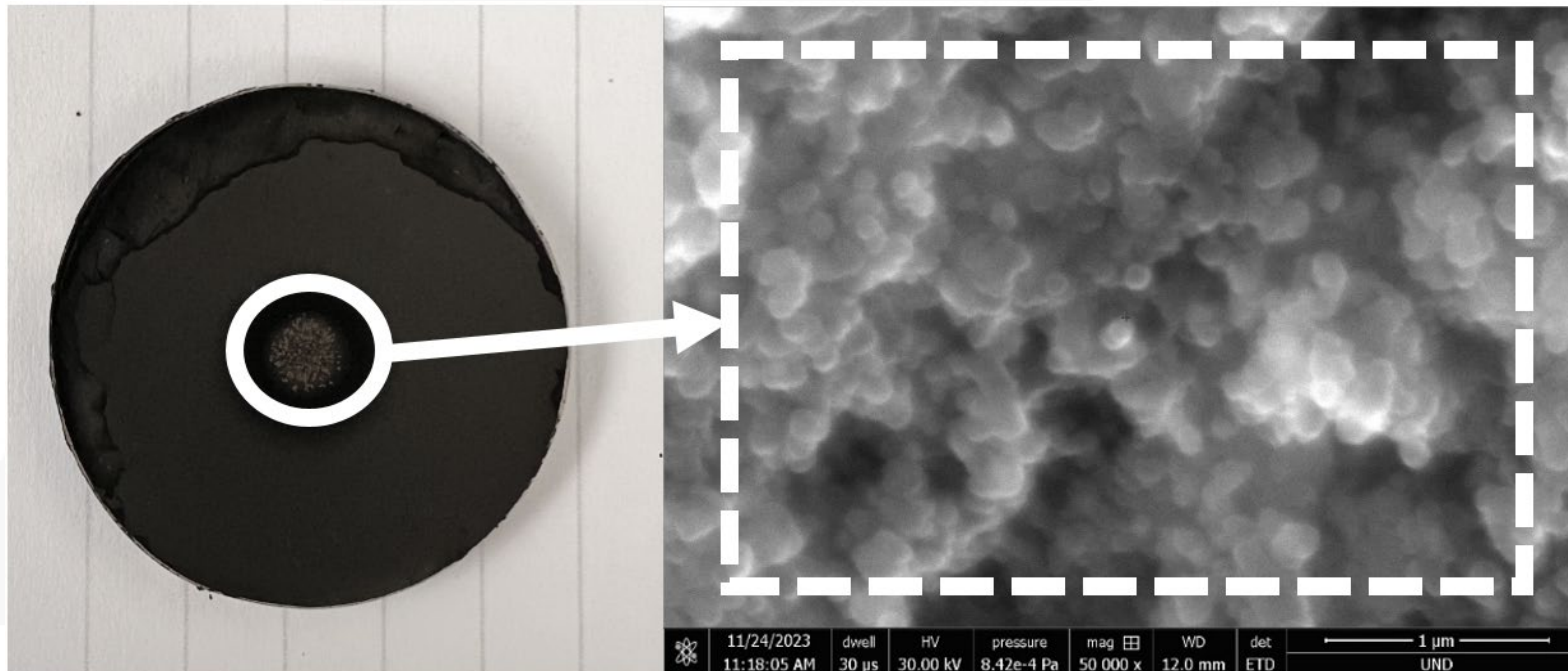
Task 4: Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

- EM-assisted mechanism on structured catalyst
- Ambient testing – no effect
- At 650°C visible surface changes
- SEM-EDX on removal/non-removal area
- Tubular-shaped C visually detected both cases
- EDX inconclusive (rough surface)



Task 4: Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

- Increased regeneration intensity following CH₄ decomposition
- Post cleaning surface indicating lack of carbonaceous material
- Excessive cleaning → catalyst inactivity



Catalyst surface after decomposition, regeneration and additional decomposition cycle

Task 4: Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

Change operating regime

- Alter regeneration intensity (<5 min.)

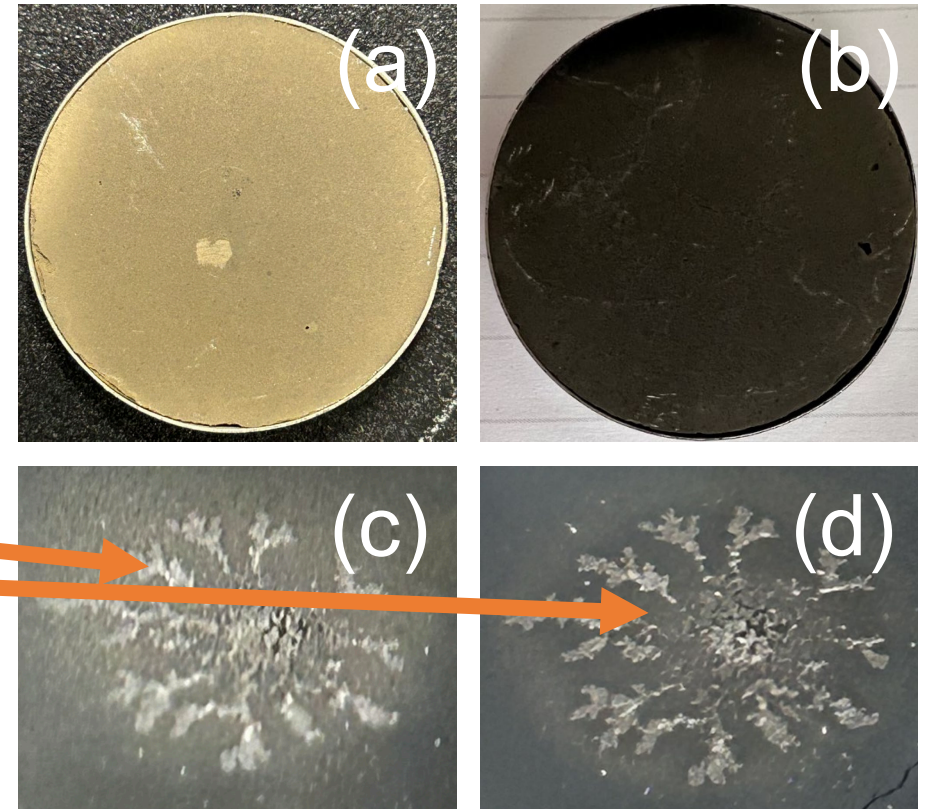
Start with:

- Structured catalyst (a)
- Decompose methane (b)
- Surface after catalyst regeneration (c)
- Surface after additional decomposition stage (d)

High intensity regions no activity

- Surrounding areas darkened (carbon deposition)

Ongoing: Assess surfaces for carbon deposition post-regeneration (TGA, SEM, TC/TIC)

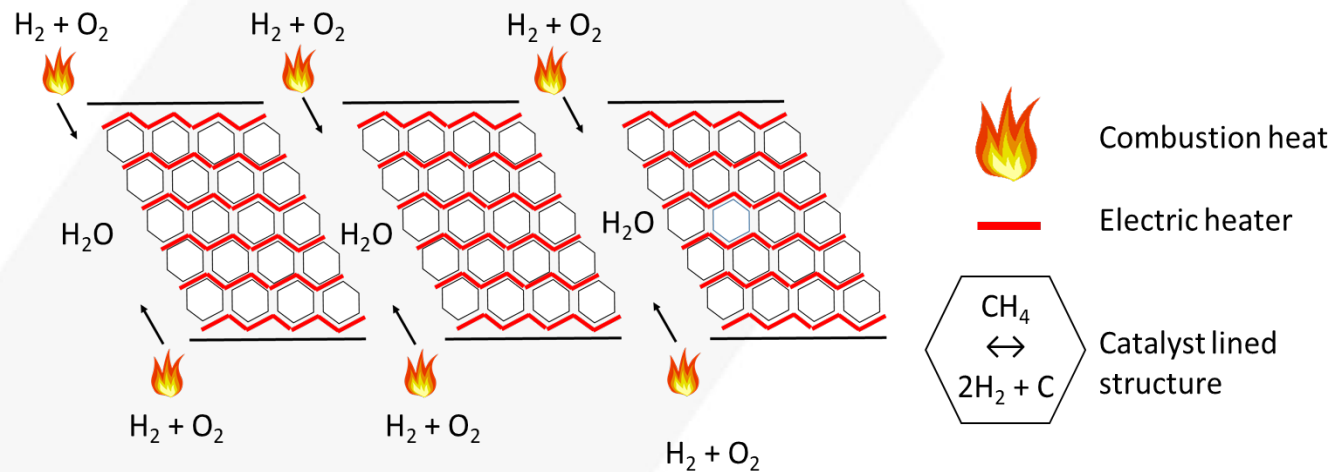


Carbon buildup and removal sequences

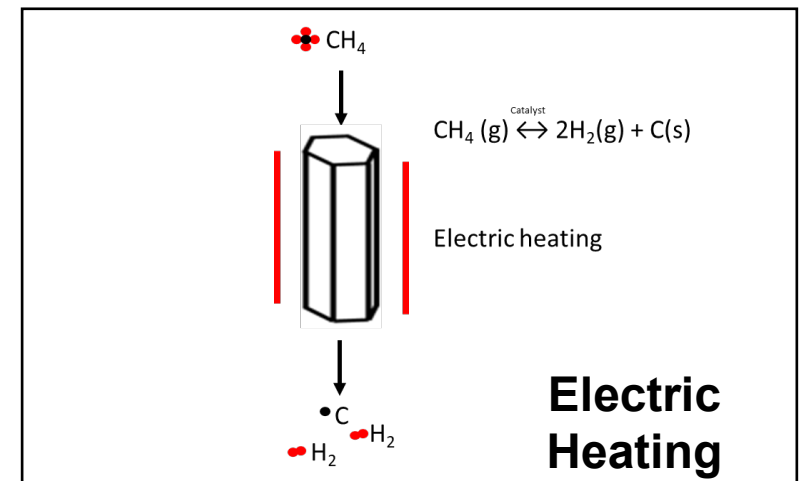
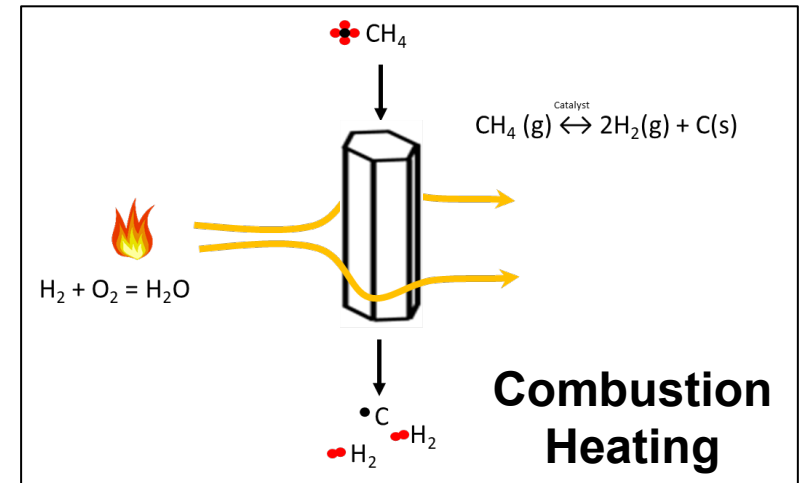
Task 5: Component Identification for Future Work

System Design:

- Design large-scale system for CDM process
- CO₂-free process (with renewable electricity)
- 1 tonne H₂ per hour



CO₂-free Production route for H₂



Extended Scope

- Cost-share provided by North Dakota Industrial Commission (NDIC)
- Scope extended to:
 - Examining uses for carbonaceous products
 - Implementing strategy for producing H₂ using process at large scale
 - Focus on H₂ production without CO₂ formation
 - Project end date: 07/31/24 (DOE and NDIC)
- Added Tasks 6, 7 and 8 to project
- Task 6 – PMP for NDIC



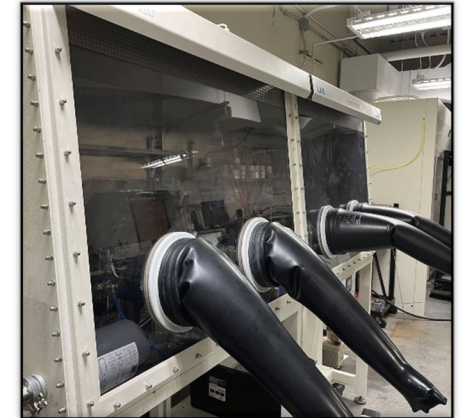
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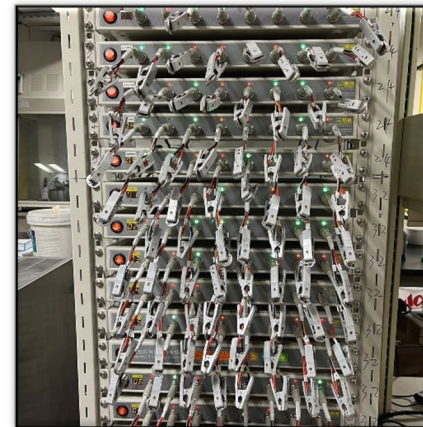
- Produced bulk carbon sample
- Tested carbon purity
- Acid leached carbon
- Prepared coin cells and testing
- Compare to commercially procured carbon



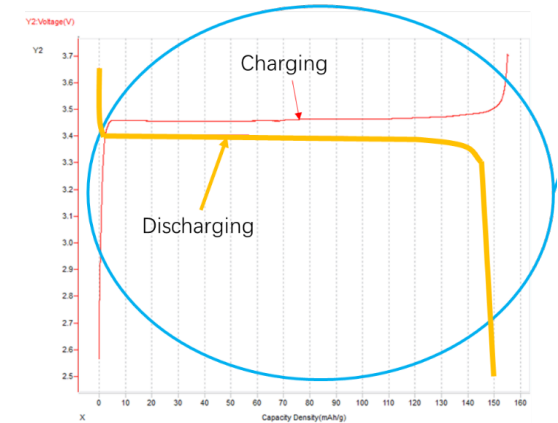
Bulk carbon production



Coin cell production



Coin cell testing



Results

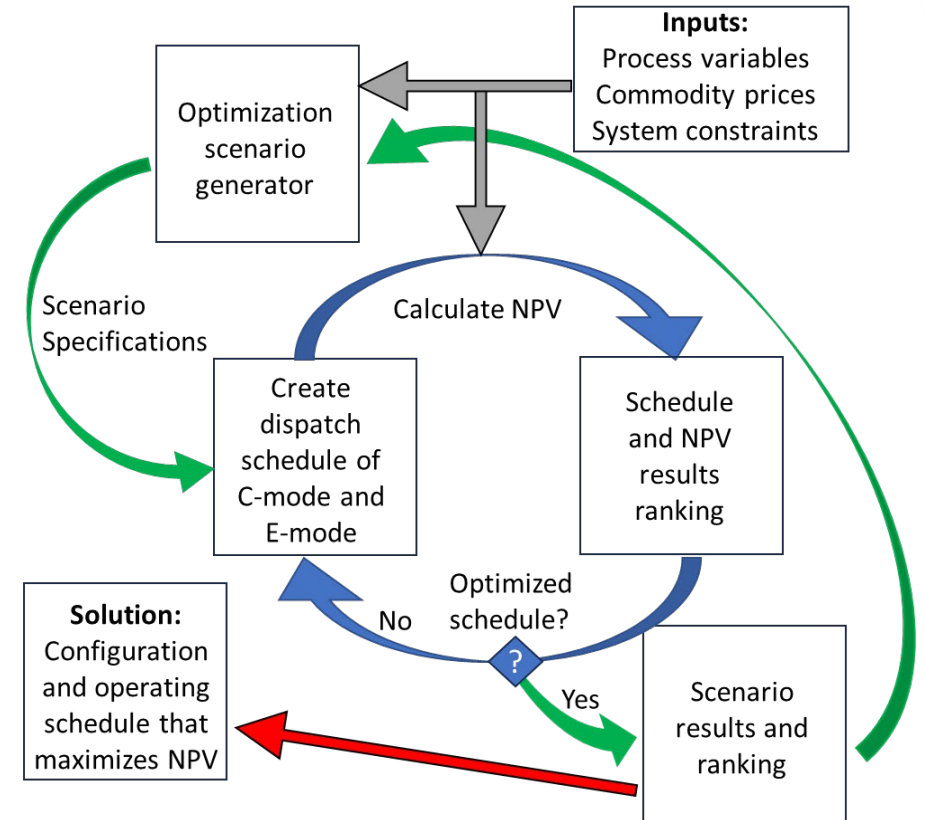
Task 8: Process Model of Greener H₂ Production System

Approach

- CH₄ decomposition for H₂
- C-mode and E-mode for production flexibility
- CO₂-free process (with renewable electricity)

Key Questions

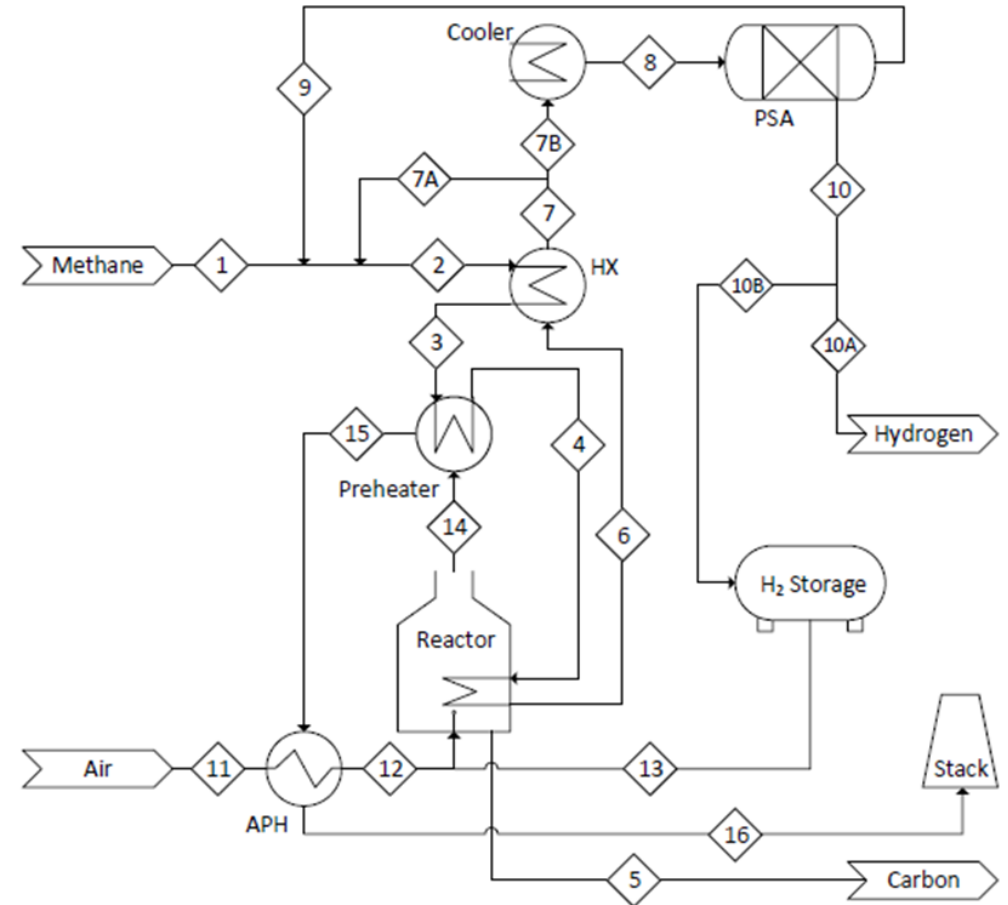
- Time ratio in each mode
- Impact of electricity and hydrogen prices
- Optimal hydrogen storage



Electric and Combustion Heating Loop Optimization Routine Depiction

Task 8: Process Model of Greener H₂ Production System

- H₂ Production Scenarios:
 - ND-MISO
 - NREL future system scenario analysis
 - H₂ supply by pipeline or liquid tankers
- Next steps:
 - Getting Plant CAPEX
 - Process optimization (sizing variations)
- Inset figure – general process layout
 - 1 tonne H₂ per hour



CO₂-free H₂ Production Pathway

Future Work

Task 1 – PMP and Technology Maturation Plan

Task 2 – Catalyst Preparation and Performance Testing

Task 3 – Computational Fluid Dynamics (CFD) Modeling of Conversion System

Task 4 – Hydrocarbon Conversion Testing using EM Energy-Assisted Thermocatalytic Process

- *Catalyst regeneration verification – SEM, TGA and TC/TIC*

Task 5 – Component Identification for Future Work

Task 6 – PMP (NDIC)

Task 7 – Evaluation and Characterization of Carbon Products

- *Compare lab-grown C in batteries to commercially procured C*

Task 8 – Process Model of Greener H₂ Production System

- *Update equipment costing from simulation results and input information for inner-outer loop approach for process optimization*



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Thank you Questions?

Dr. Johannes van der Watt

College of Engineering and Mines Research Institute (CEMRI)

University of North Dakota

johannes.vanderwatt@und.edu

701-777-5177

