U.S. DEPARTMENT OF ENERGY | Fossil Energy and Carbon Management

NATIONAL ENERGY TECHNOLOGY LABORATORY

FY22 FECM Spring R&D Project Review Meeting
DE-FE0032005

Reversible Methane Electrochemical Reactor as an Efficient Energy Storage for Fossil Power Generation

AOI 3 – Innovative Concepts and Technologies of DE-FOA-0002332 “Energy Storage for Fossil Power Generation.”

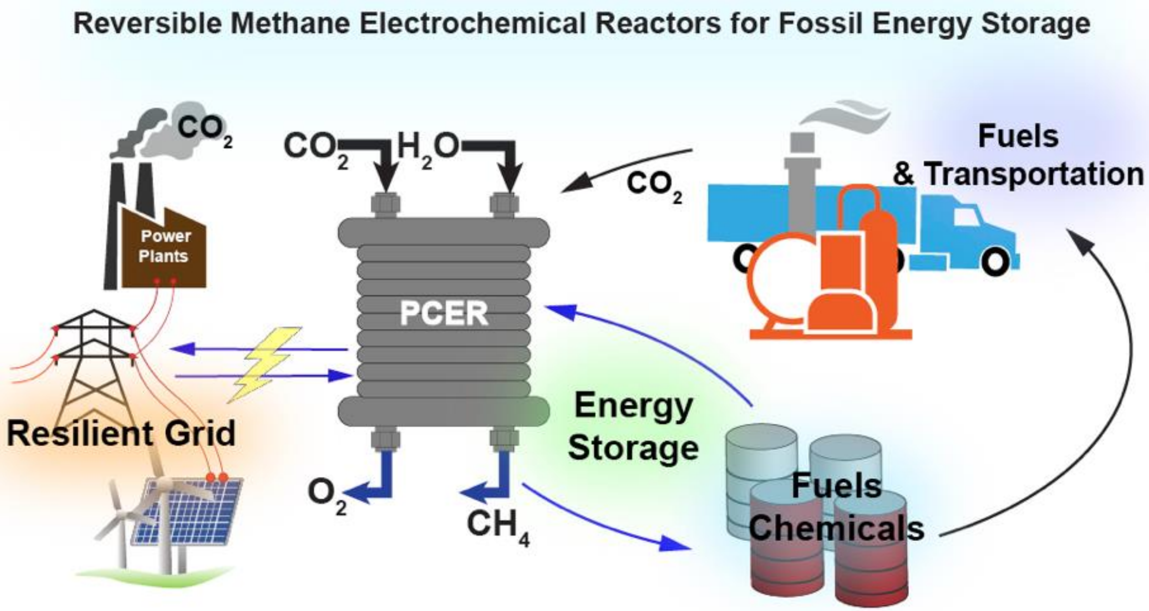
May, 2022

OU

KSU

AOI 3: Innovative Concepts & Technologies

"The RFI established that a substantial number of energy storage technologies are relatively early-stage in their development. …The technologies need additional R&D to clarify their current state, understand their suitability for future advancement and integration, and to advance their maturity through R&D."



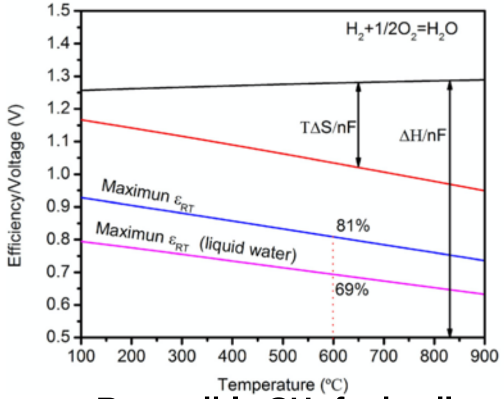
Project Objectives

Conduct a comprehensive R&D program to demonstrate the suitability and future advancement and integration of reversible methane electrochemical reactors as an Efficient Energy Storage (EES) with fossil fuel power plants.

Protonic ceramic electrochemical reactor for power generation and chemicals production

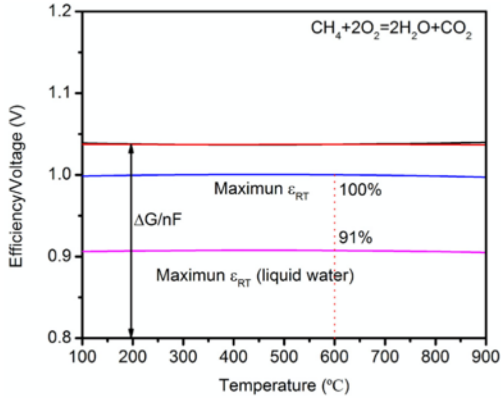
Reversible H_2 fuel cell

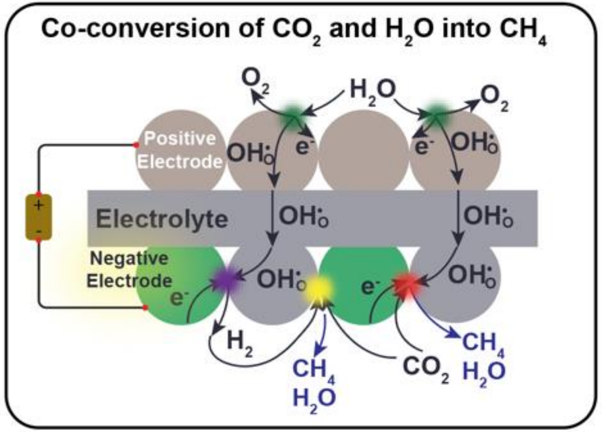
Round trip efficiency: 81%

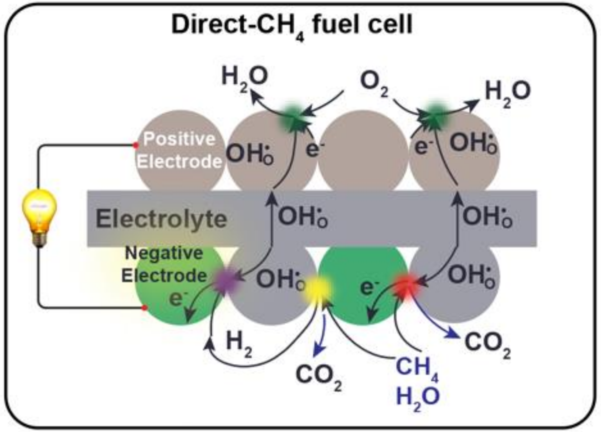


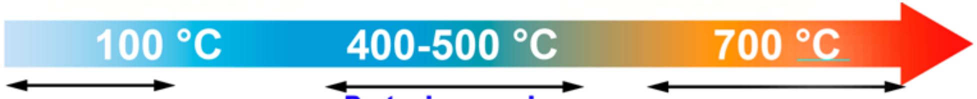
Reversible CH_4 fuel cell

Round trip efficiency: 100%









Polymer membrane Reactor

- Sluggish Kinetics
- High-cost catalyst
- Cannot process hydrocarbons,

Protonic ceramic electrochemical reactor

- High Performance
- Durable
- Can process hydrocarbons
- Thermodynamically favorable for methanation

Oxygen-ion solid oxide electrochemical reactor

- High Performance
- Poor Durability
- Coking
- Product is limited to CO or syngas

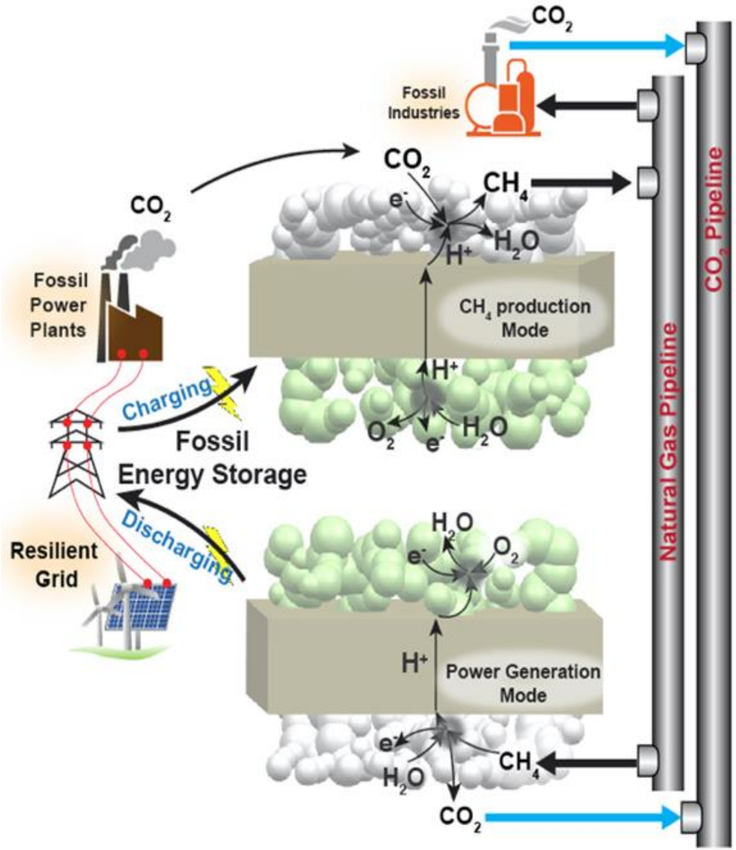
Distinguish feature of the proposed technology

1- Direct Integration with fossil assets including fossil power plants and fossil-fuel industrial applications

2- PCERs exhibit high H_2S tolerance and coking tolerance

3- Reversible methane electrochemical cells display a theoretical round-trip efficiency of up to 100%

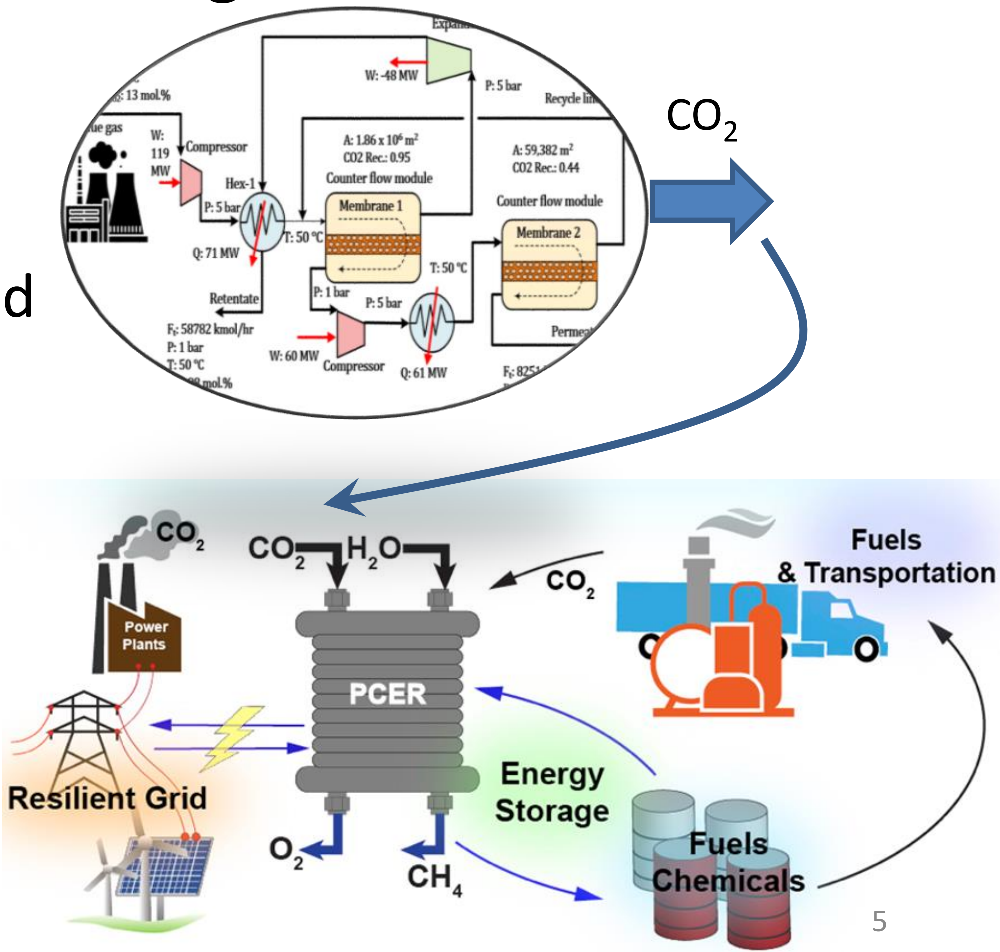
4- Reduced operating temperatures enable hybridization with a broader range of waste heat sources



Technology integration with existing or new fossil assets

Direct integration: the flue gas enters the system without additional complex separation and purification processes to capture CO_2, allowing significant cost reduction.

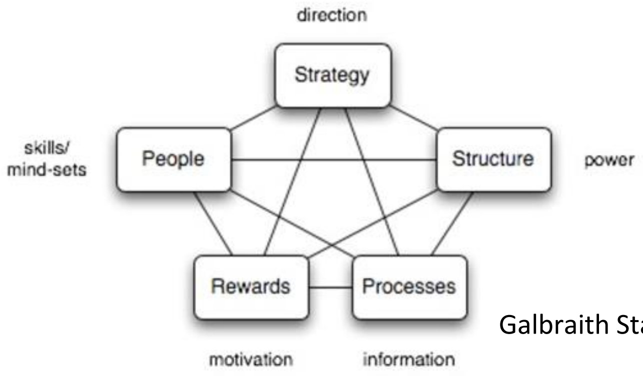
Indirect integration: CO_2 is captured and separated through an intermediate system before entering the reversible PCER.



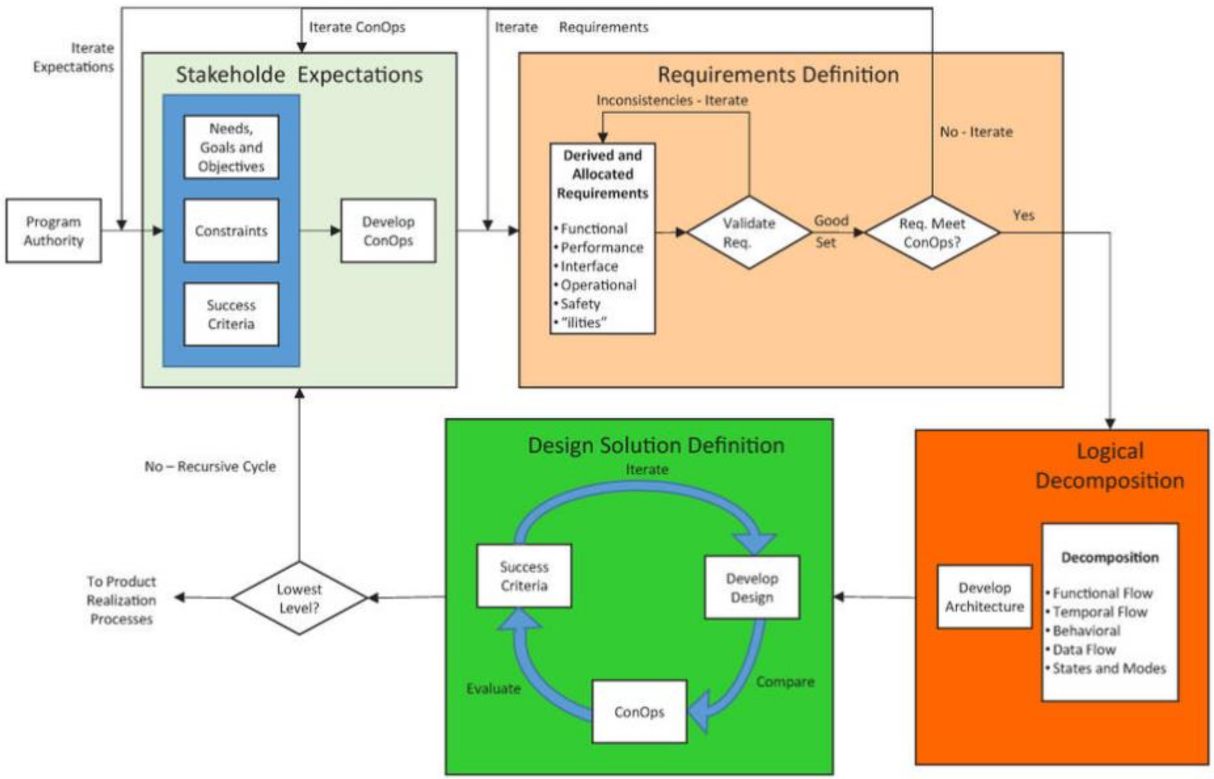
Define the Proposed Energy Storage Technology

Systems Design Processes

1. Stakeholders Expectations Definition
2. Technical Requirements Definition
3. Logical Decomposition
4. Design Solution Definition



Galbraith Star Model



[2] Interrelationships among the System Design Processes. NASA

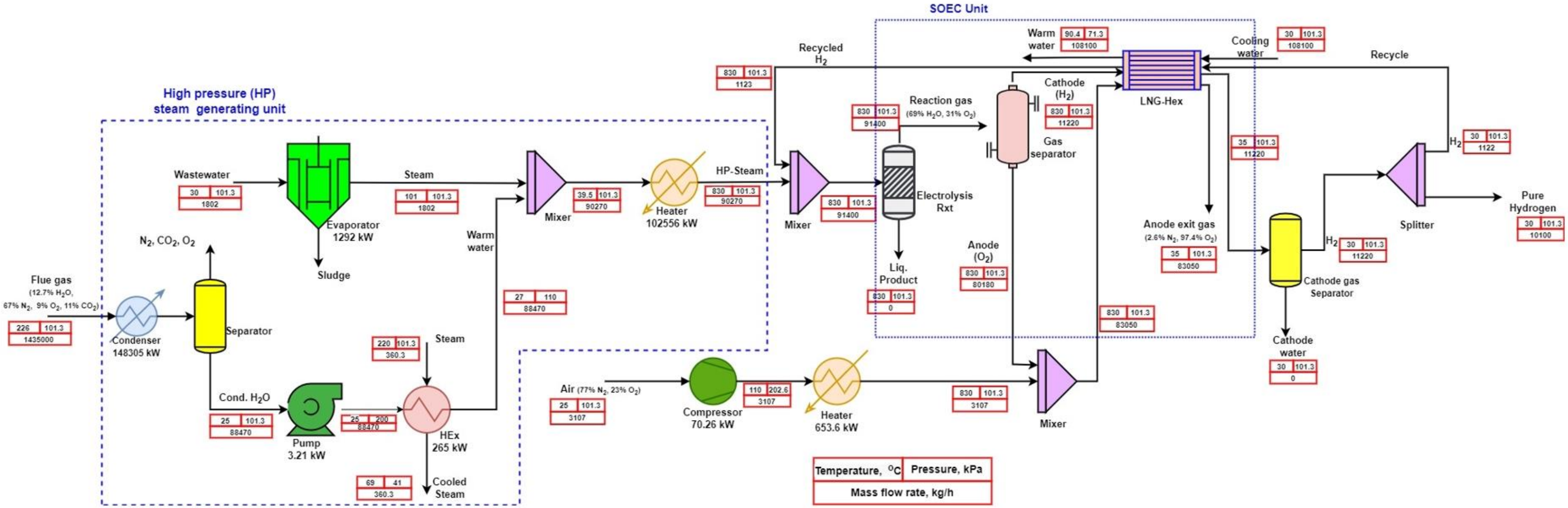
[1] https://pragmaticarchitect.wordpress.com/2013/05/14/how-to-build-a-roadmap-define-end-state/
[2] https://www.nasa.gov/seh/2-fundamentals

DOE Status and Targets for Reversible Solid Oxide Performance and Cost

- Stack cost target of $225/kW
- System cost target of $900/kW
- Less than 0.2% per 1,000 hours over an operating lifetime of 40,000 hours
- Efficiency of greater than 60% without carbon capture and storage

Metric	2018 Status	2020 Targets	2025/2030 Targets
System Cost ($/kWe)	>12,000	6,000	900
System Degradation (%/1000 hrs)	1-1.5	0.5-1.0	>0.2
Durability (hr)	<2000	5000	8000
Fuel	Natural gas	Natural gas Simulated syngas	Natural gas Coal-derived syngas
Demonstration Scale	50 kWe – 200 kWe	200 kWe – 1 MWe	10-50 MWe

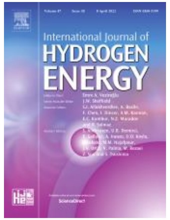
Comprehensive Techno-economic System Modeling of Reversible Methane PCER



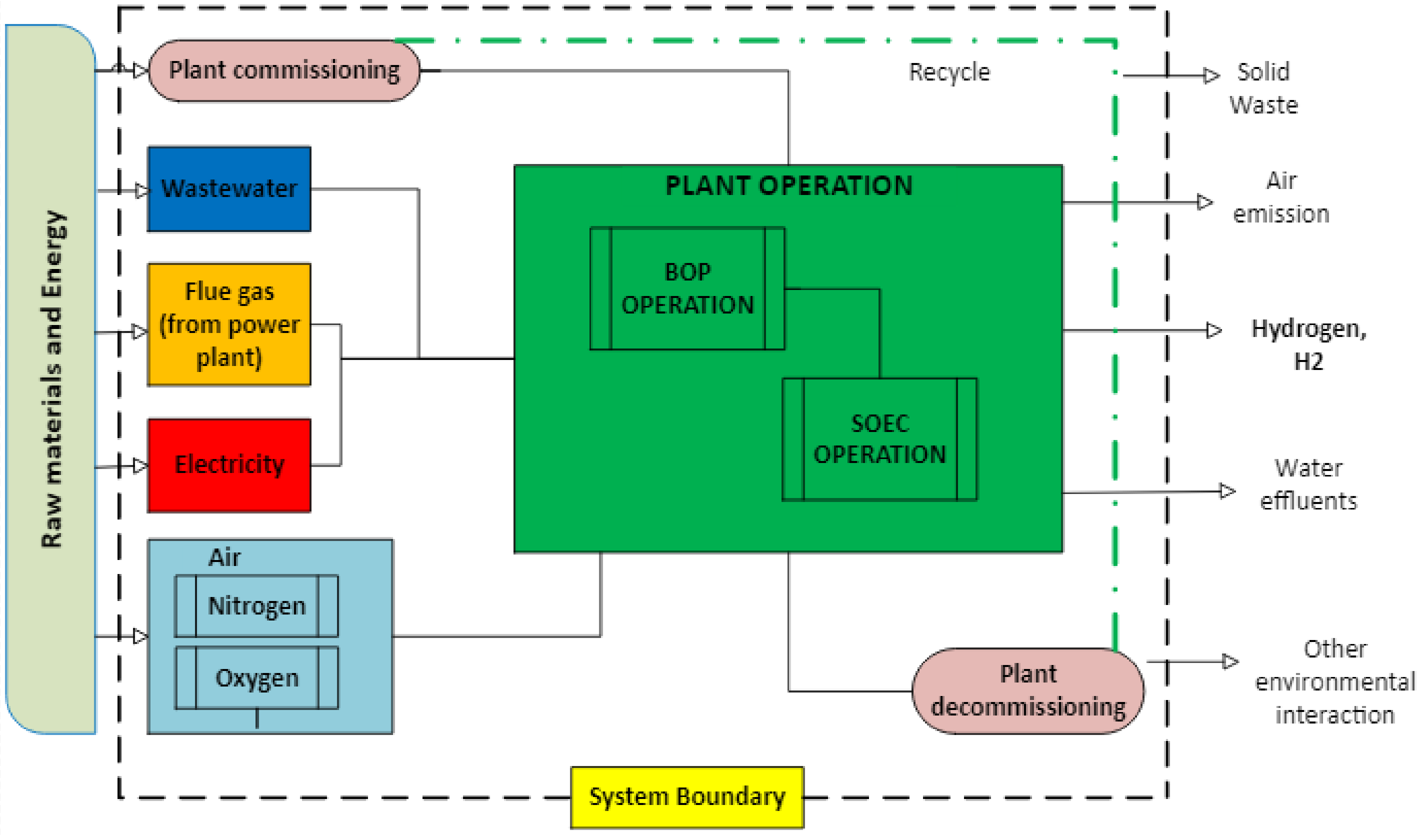
A Novel Hydrogen Economy based on Electrochemical Cells Using Water-Energy Nexus Framework (submitted to IJHE)

Proceedings of the ASME 2022 16th International Conference on Energy Sustainability ES2022
July 11-14, 2022, Virtual, Online

WHEC-2022
23rd World Hydrogen Energy Conference
onsite and online
June 26-30, 2022 Istanbul Congress Center, Istanbul, Turkey

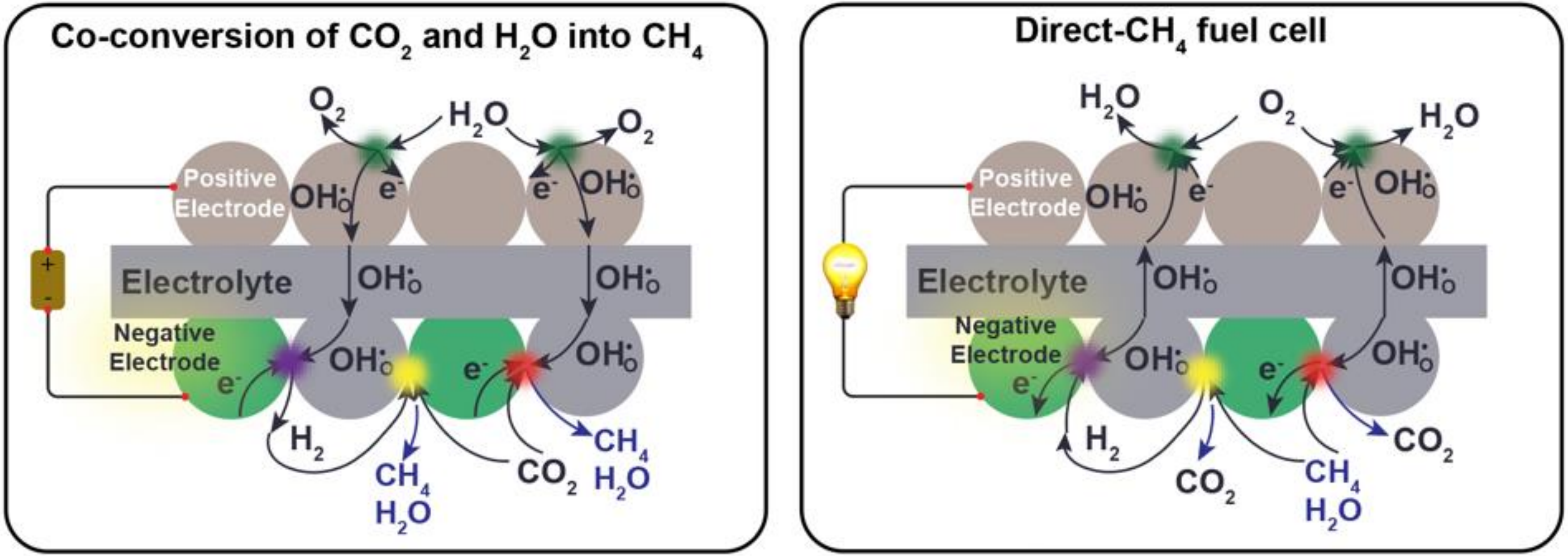


Generic LCA illustration of the water-energy nexus hydrogen production



Experimental Studies to Define Reversible Methane PCER Performance Parameters

Previous Protonic ceramic electrochemical reactor for power generation and chemicals production



High-performance protonic ceramic fuel cells (PCFCs)

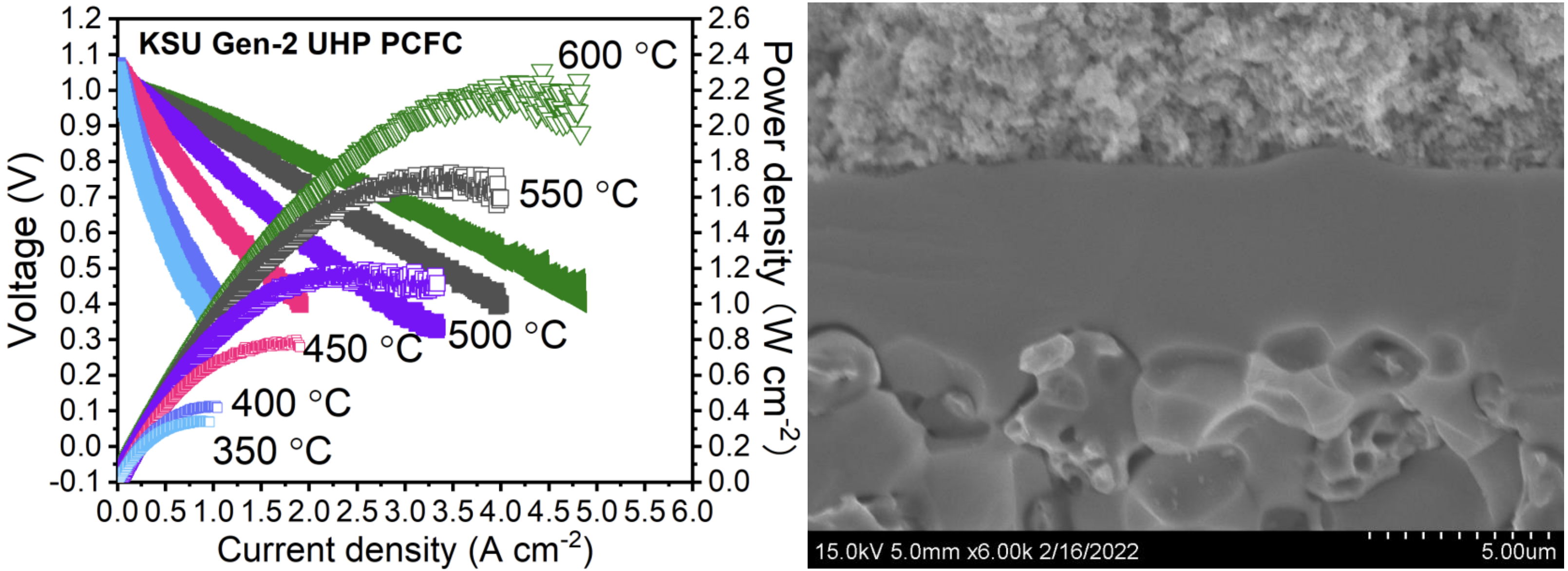
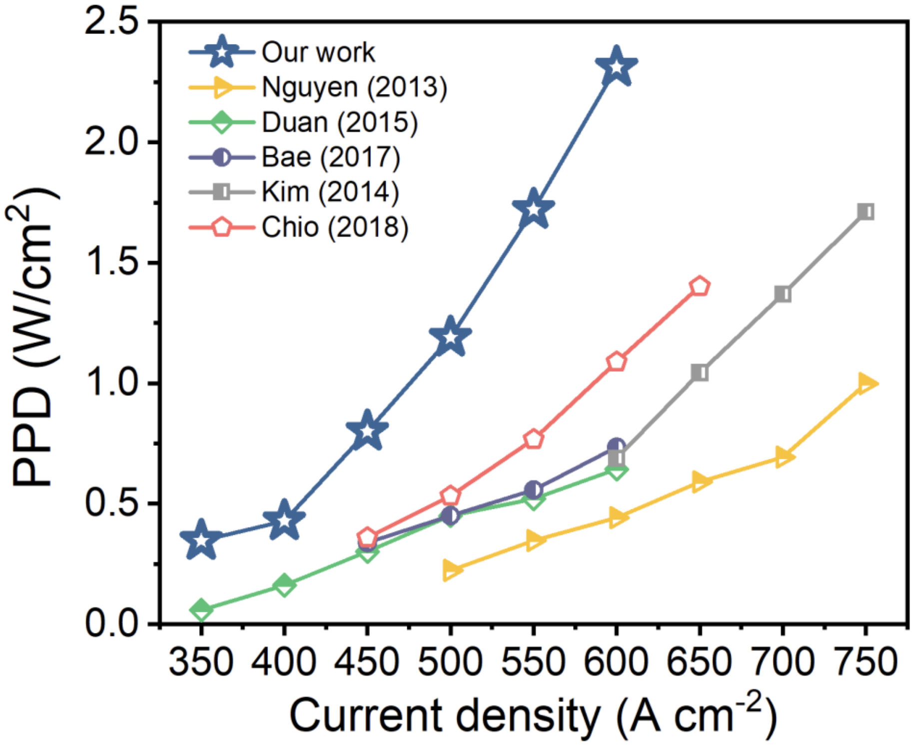


Figure 1. Ultra-high-performance KSU Gen-2 PCFC recently demonstrated at KSU.

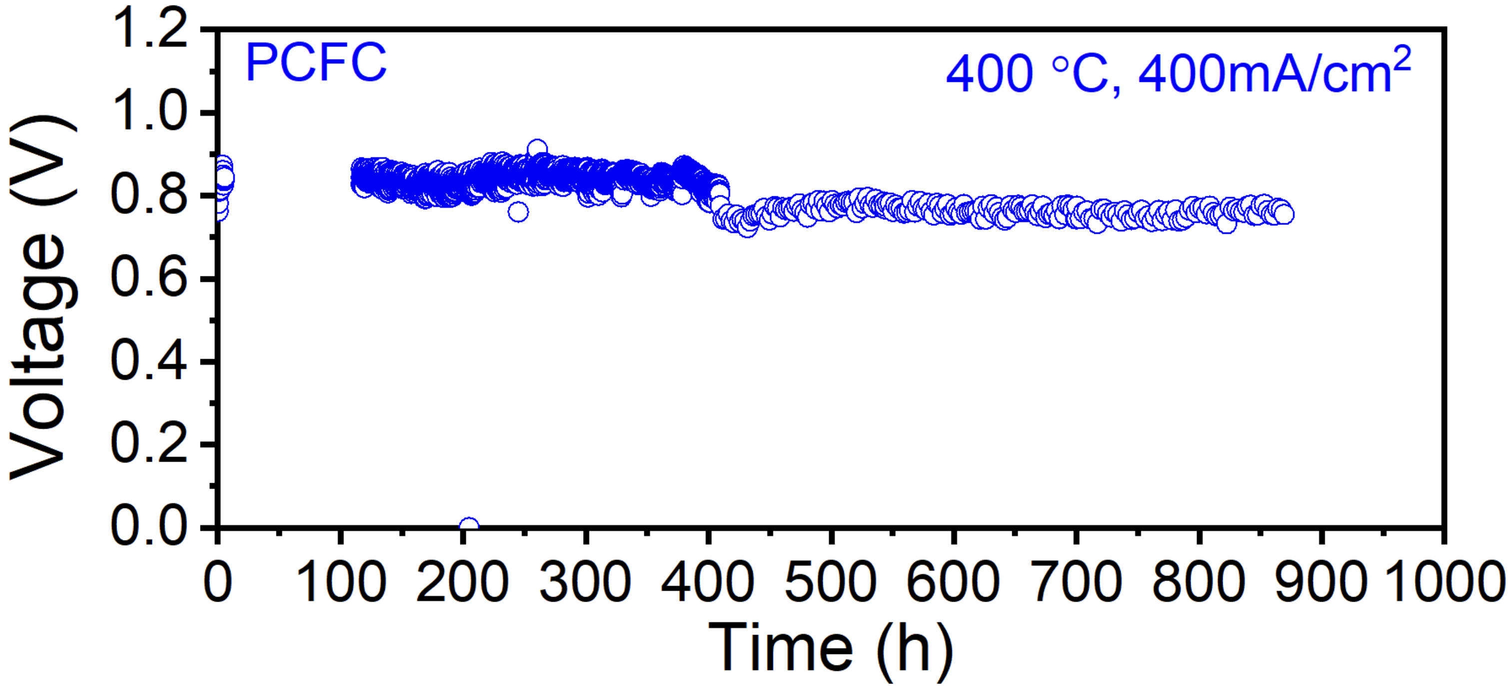
Word record PCFC performances

Research Activity 1 at KSU: high-performance protonic ceramic fuel cells (PCFCs)



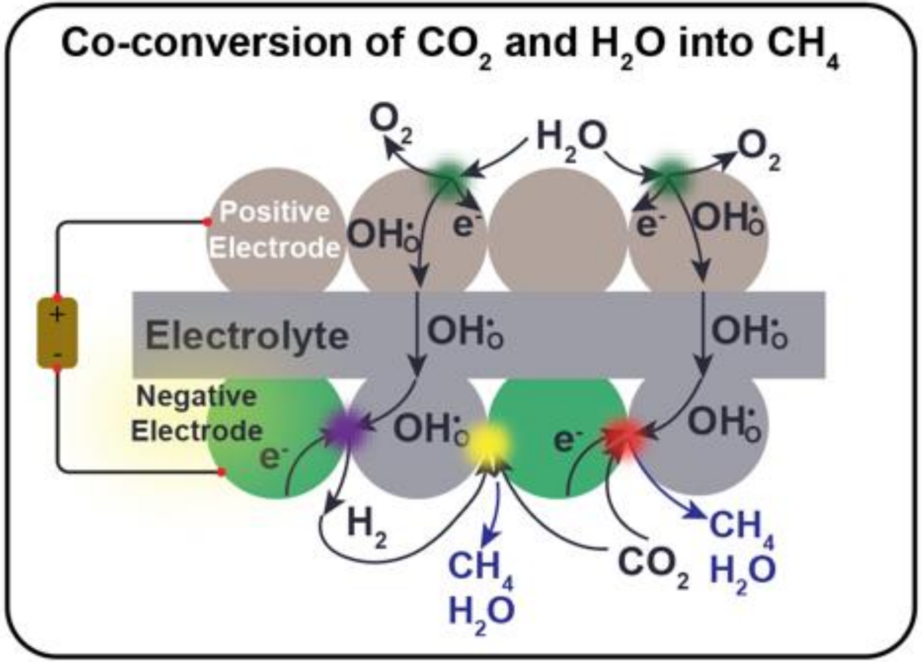
Word record PCFC performances

High-performance protonic ceramic fuel cells (PCFCs)



Durable PCFC performances at intermediate operating temperatures

Research Activity 2 at KSU: CO_2 methanation catalysts



ACS APPLIED MATERIALS & INTERFACES

www.acsami.org

Research Article

Enhanced CO_2 Methanation Activity of $Sm_{0.25}Ce_{0.75}O_{2-\delta}$–Ni by Modulating the Chelating Agents-to-Metal Cation Ratio and Tuning Metal–Support Interactions

Fan Liu, Yoo Sei Park, David Diercks, Pejman Kazempoor, and Chuancheng Duan*

Cite This: *ACS Appl. Mater. Interfaces* 2022, 14, 13295–13304

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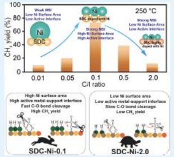
ABSTRACT: Highly active and selective CO_2 methanation catalysts are critical to CO_2 upgrading, synthetic natural gas production, and CO_2 emission reduction. Wet impregnation is widely used to synthesize oxide-supported metallic nanoparticles as the catalyst for CO_2 methanation. However, as the reagents cannot be homogeneously mixed at an atomic level, it is challenging to modulate the microstructure, crystal structure, chemical composition, and electronic structure of catalysts via wet impregnation. Herein, a scalable and straightforward catalyst fabrication approach has been designed and validated to produce $Sm_{0.25}Ce_{0.75}O_{2-\delta}$-supported Ni (SDC–Ni) as the CO_2 methanation catalyst. By varying the chelating agents-to-total metal cations ratio (C/I ratio) during the catalyst synthesis, we can readily and simultaneously modulate the microstructure, metallic surface area, crystal structure, chemical composition, and electronic structure of SDC–Ni, consequently fine-tuning the oxide–support interactions and CO_2 methanation activity. The optimal C/I ratio (0.1) leads to an SDC–Ni catalyst that facilitates C–O bond cleavage and significantly improves CO_2 conversion at 250 °C. A CO_2-to-CH_4 yield of >73% has been achieved at 250 °C. Furthermore, a stable operation of >1500 hours has been demonstrated, and no degradation is observed. Extensive characterizations were performed to fundamentally understand how to tune and enhance CO_2 methanation activity of SDC–Ni by modulating the C/I ratio. The correlation of physical, chemical, and catalytic properties of SDC–Ni with the C/I ratio is established and thoroughly elaborated in this work. This study could be applied to tune the oxide–support interactions of various catalysts for enhancing the catalytic activity.



KEYWORDS: *CO_2 methanation, SDC–Ni, oxide–support interaction, structure–property relationship, in situ operando DRIFTS*

1. INTRODUCTION

Converting CO_2 and renewable H_2 to CH_4 can produce sustainable natural gas, reduce the reliance on fossil fuels, and decrease greenhouse gases emissions, leading to substantial economic and environmental benefits.[1−4] CO_2 methanation is thermodynamically favorable at 200–300 °C. However, CO_2 molecules are very stable, and accordingly, high operating temperatures (>300 °C) are required to activate CO_2 molecules and achieve practically valuable CH_4 yield, necessitating the development of highly active catalysts for CO_2 conversion at <300 °C.[5,6] However, CO_2 methanation at high operating temperatures consumes extensive energy and inevitably favors CO_2-to-CO conversion, reduces the equilibrium CO_2 conversion, and decreases CH_4 yield and purity. Therefore, a CO_2 methanation catalyst that can attain a CH_4 selectivity of >99%, a CH_4 yield of >70%, and a long operation stability at 250 °C is essential for economic renewable natural gas production. Despite enormous efforts that have been devoted to designing and synthesizing advanced CO_2 methanation catalysts via novel approaches, such as noble metal-based catalysts,[7−10] metal–organic frameworks (MOF)-supported metallic nanoparticles,[11,12] and plasma treatment,[13] there are limited facial and scalable fabrication methods, which can readily fine-tune metal–support interactions to realize a CO_2-to-CH_4 yield of >70% with a long-term stable operation (>1000 hours) at 250 °C.

It has been recognized for a long time that synergistic interactions are exhibited between oxide support and metallic nanoparticles. These interactions typically relate to the microstructure of metallic nanoparticles and oxide support, chemical compositions and electronic structure of both oxides and metallic nanoparticles, charge transfer between oxides and metals, and interfacial active area, which play essential roles in activating and converting CO_2.[14−24] Modulating these metal–support interactions is therefore a promising approach to improving CO_2 conversion and tuning the CO_2 methanation

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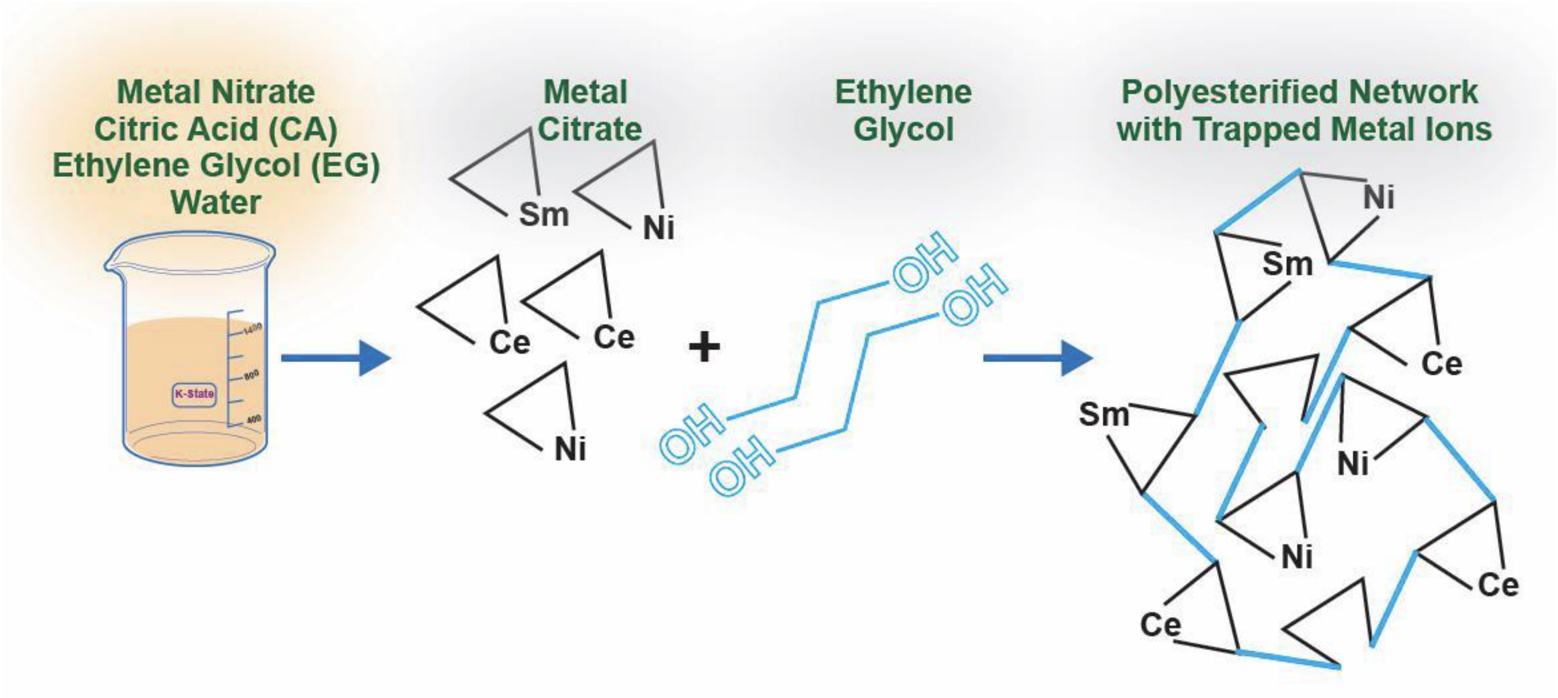
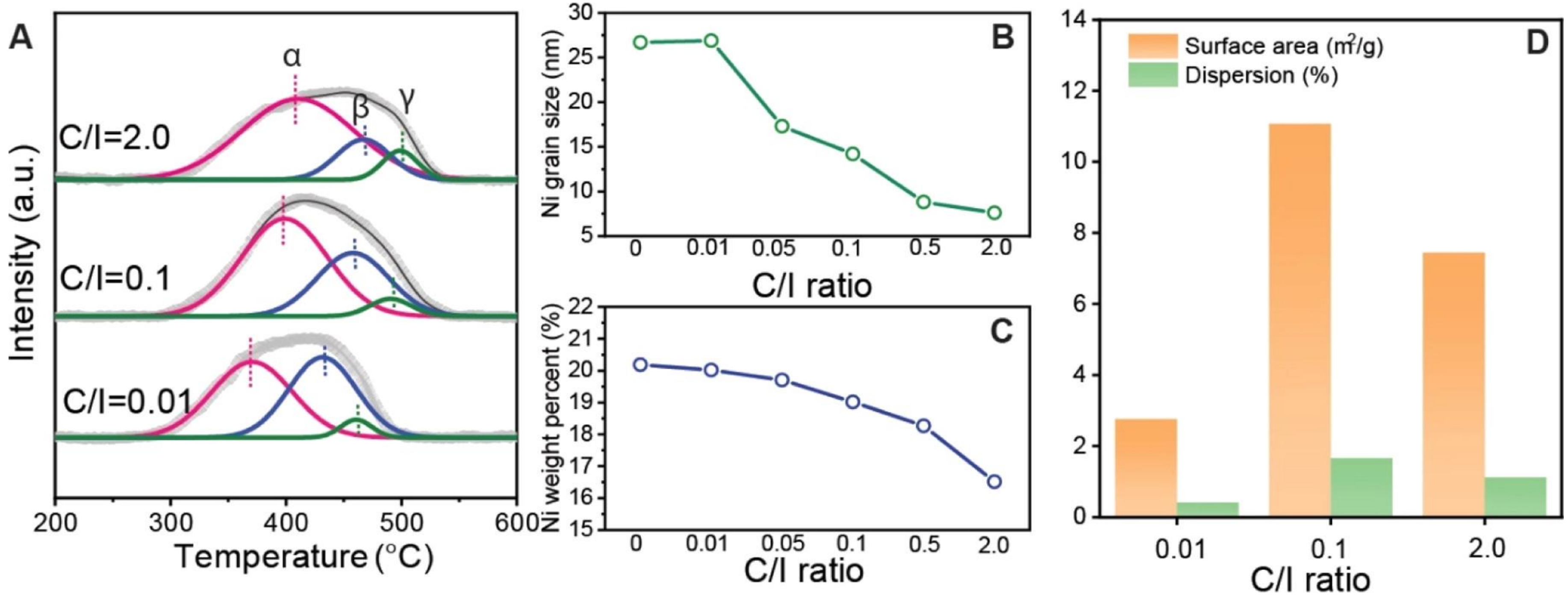
CO_2 methanation catalysts



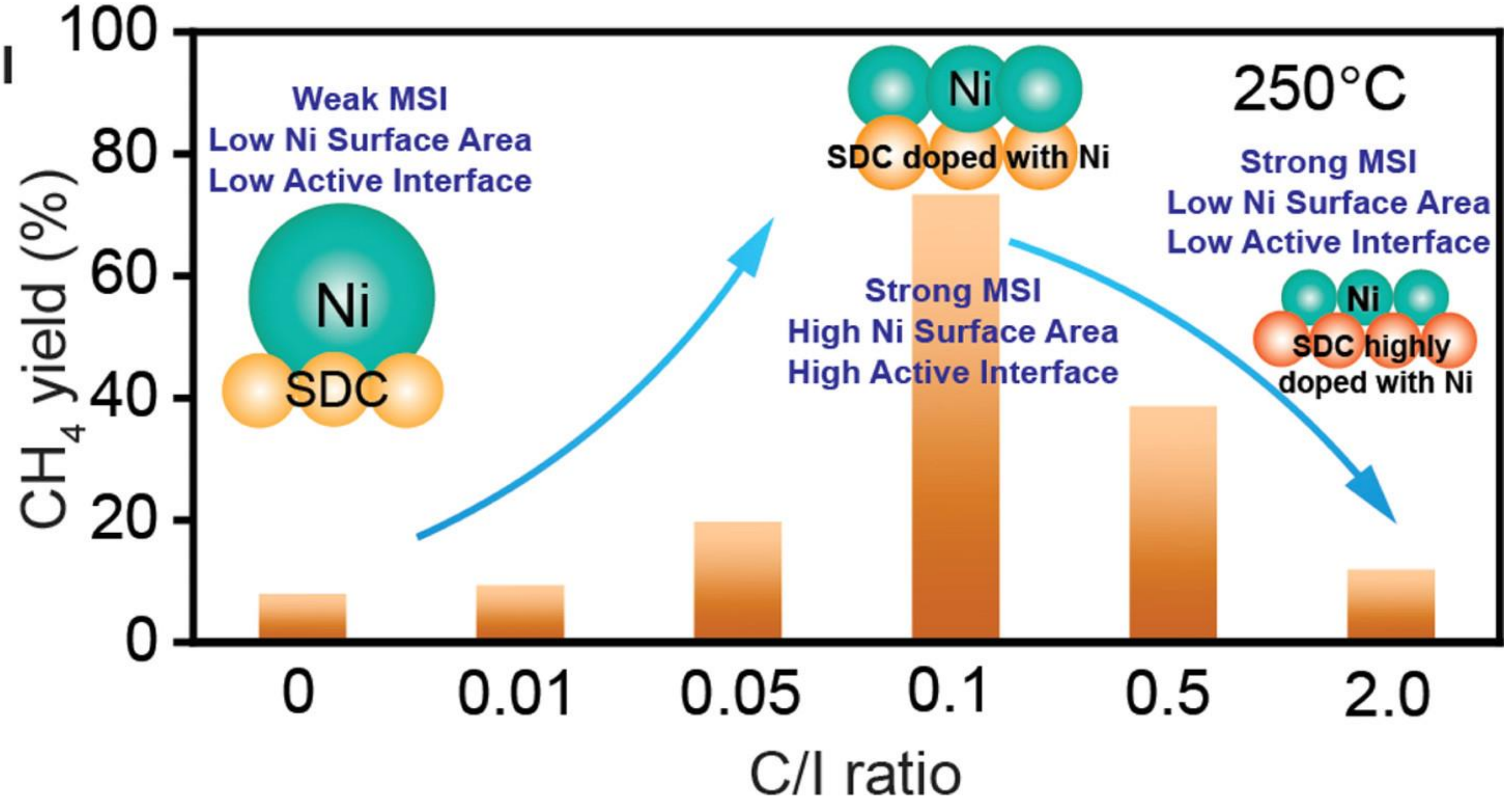
One facile CO_2 methanation catalyst synthesis approach by adjusting chelating agent/ion ratios (C/I)

Research Activity 2 at KSU: CO_2 methanation catalysts

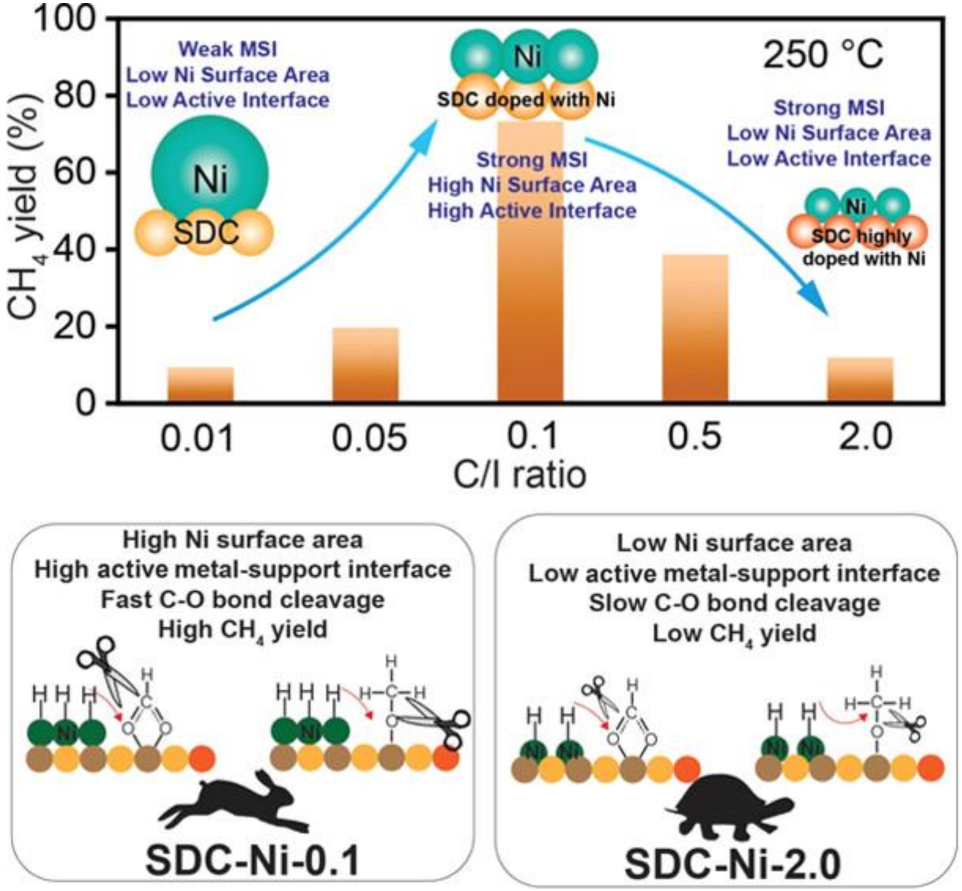


The C/I ratio affects the oxide-Ni interactions and microstructure

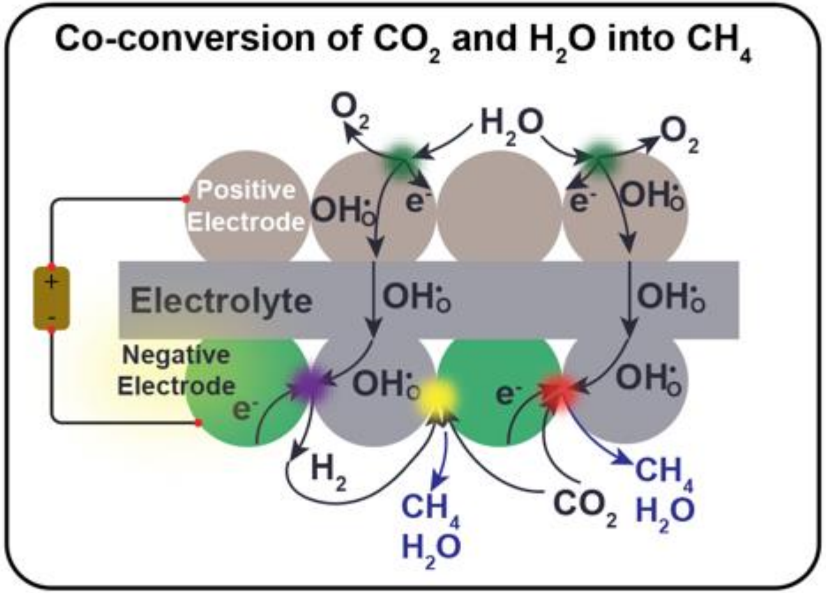
CO_2 methanation catalysts

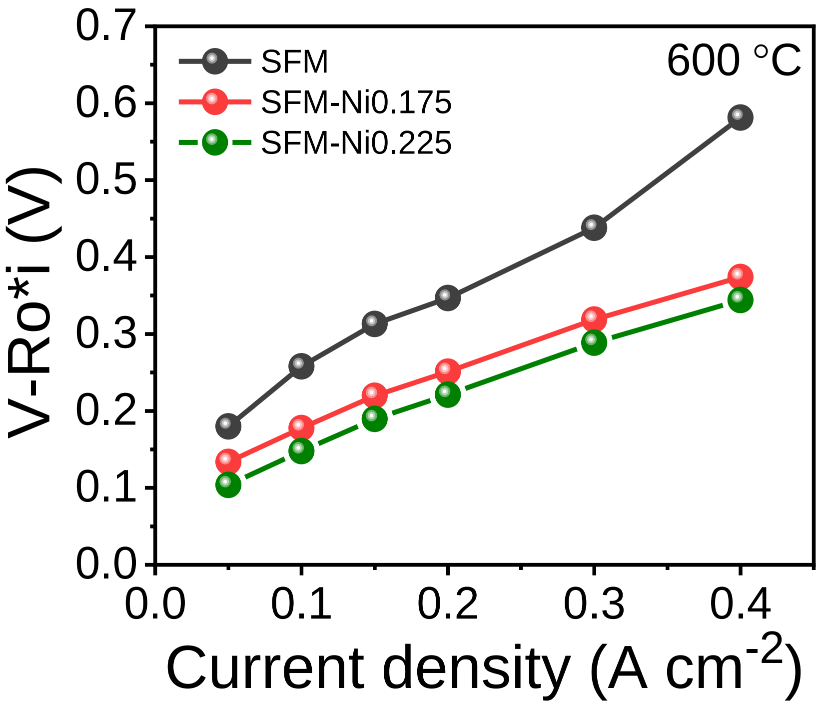


CO_2 methanation catalysts



CO_2 conversion in PCECs





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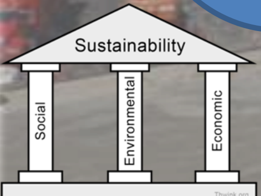
Energy Storage and Management

For more information, please contact Dr. Pejman Kazempoor (pkazempoor@ou.edu)



Developing solutions that are locally appropriate; socially beneficial; economically and technically feasible; and environmentally responsible.

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