Rational Design Platform for Transition Metal Catalyzed Electrochemical Synthesis

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Lawrence Livermore National Laboratory/Opus 12/TOTAL
5/1/2018-4/30/2020

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Overview

Project Title:
RATIONAL DESIGN PLATFORM FOR TRANSITION METAL CATALYZED ELECTROCHEMICAL SYNTHESIS

Timeline:

Project Start Date: 05/01/2018

Budget Period End Date: 04/30/2020

Project End Date: 04/30/2020

Project Budget and Costs:

Budget	DOE Share	Cost Share	Total	Cost Share %
Overall Budget	$1,000,000	$460,000	$1,460,000	31.5%
Approved Budget (BP-1&2)	$1,000,000	$460,000	$1,460,000	31.5%
Costs as of 5/16/19	$599,000	$116,000	$715,000	16%

Barriers and Challenges:

Electrochemical conversion of CO_2 to methane and other fuels requires the improvement of the energy efficiency and selectivity of electrochemical catalysts.

AMO MYPP Connection:
Reference AMO MYPP Area(s) being addressed

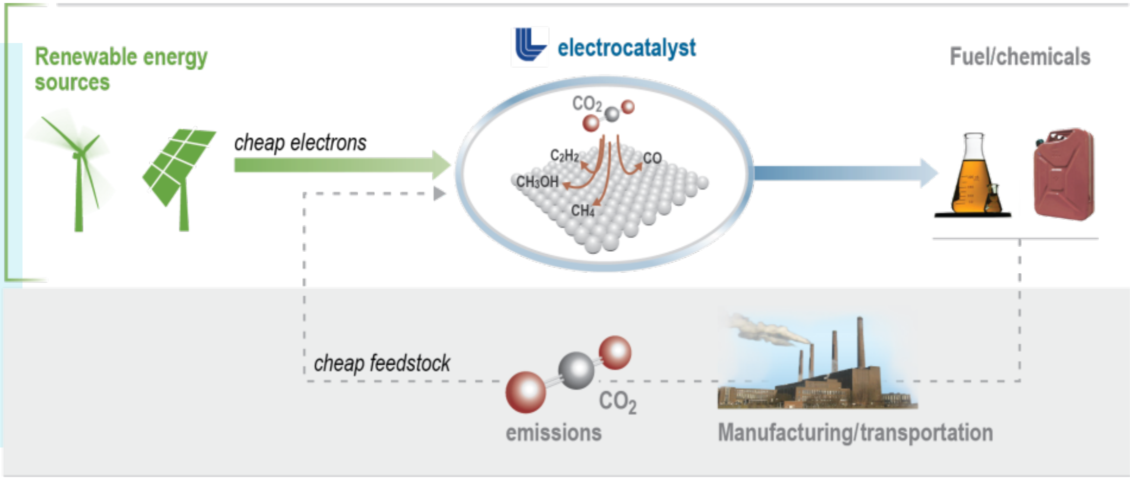
Project Team and Roles:

Opus 12:
Brings in expertise in CO2RR catalyst testing under industry relevant conditions and commercial scaling

TOTAL:
Provides industry insight regarding electrochemical CO_2 reduction to target molecules.

Project Objectives

Project Goal:
Optimize Catalyst Performance
Improve the energy efficiency and selectivity of Cu-based catalysts for electrochemical CO_2-to-fuel conversion by engineering the potential energy landscape



Objective 1: Morphology Control at the nano/meso/macro scale
- Increase number and accessibility of active sites.
- Control the local atomic configuration of the active sites

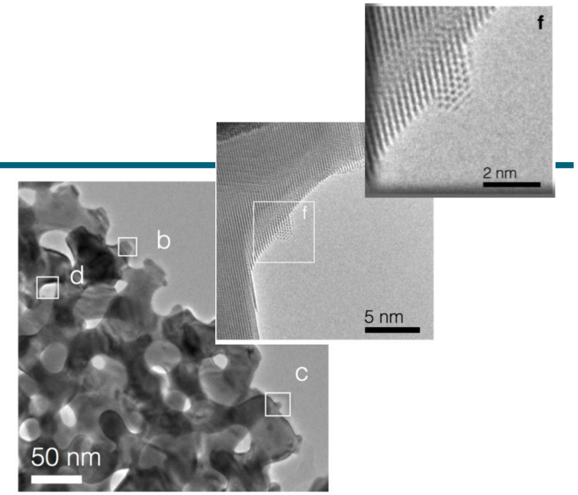
Objective 2: Catalyst Composition Tuning
- Localized and selective stabilization of critical intermediates to improve catalyst reactivity and selectivity

Objective 3: Interface Environment Control
- Control the local potential energy landscape by tuning solvent, local electric field and ionic strength

Advanced Manufacturing Office Mission Relevance of CO_2-to-fuel conversion :
Conversion of inexpensive, renewable electrical energy to chemical energy for grid energy storage, fuels, and chemicals is broadly attractive to utilities and chemical manufacturers, and contributes to reduce the life-cycle energy consumption of manufactured goods.

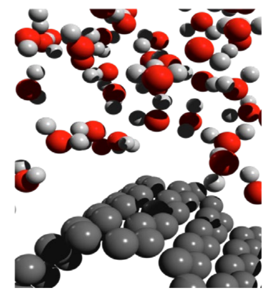
Technical Approach

Full integration of the team's unique expertise:



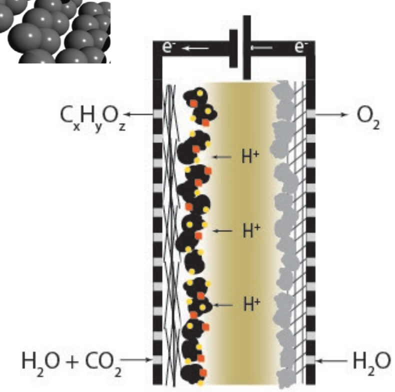
Novel nanoporous metals and dilute alloys (LLNL)

- Synthesis and characterization of nanoporous dilute alloy catalysts
- Additive manufacturing of engineered hierarchical nanoporous metals
- Leverages catalyst development expertise and investment gained through DOE's EFRC *IMASC*.



Large-scale quantum simulations of realistic electrochemical interfaces (LLNL)

- Takes advantage of LLNL's unique expertise in multiscale modeling of explicit electrochemical interfaces
- Leverages cutting-edge methods development under 2017 METI-DOE U.S.-Japan Agreement on Clean Energy Research
- LLNL's leadership-class computing platforms



CO2RR catalyst testing and commercial scaling (Opus 12)

- Provides access to Opus12's advanced Proton Exchange Membrane (PEM) electrolyzer technology

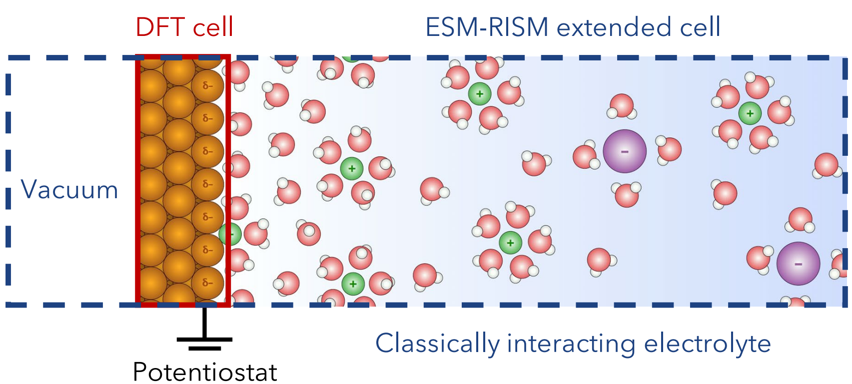
Industry insight (TOTAL)

- As one of the world's largest integrated international oil and gas companies with operations in more than 130 countries worldwide, and leading integrated producer of chemicals, TOTAL provides valuable industry insight regarding the electrochemical CO2 reduction to target molecule.

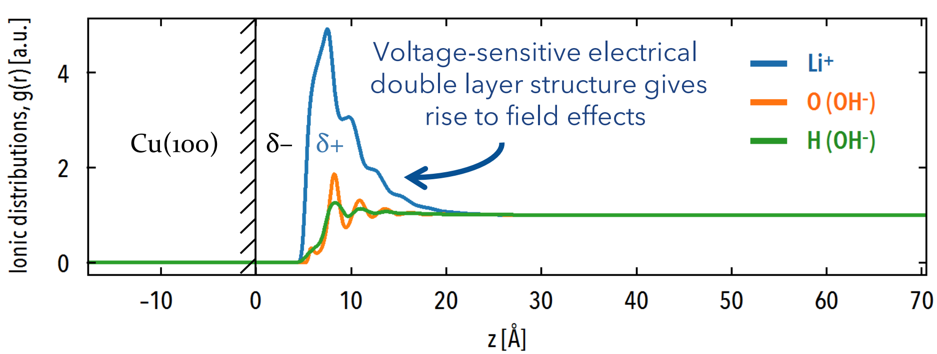
Technical Approach

Interfacial modeling via Density Functional Theory (DFT) and the Effective Screening Medium – Reference Interaction Site Method (ESM-RISM) provides a direct approach to theoretically assess both electrode and electrolyte composition effects on catalyst selectivity.

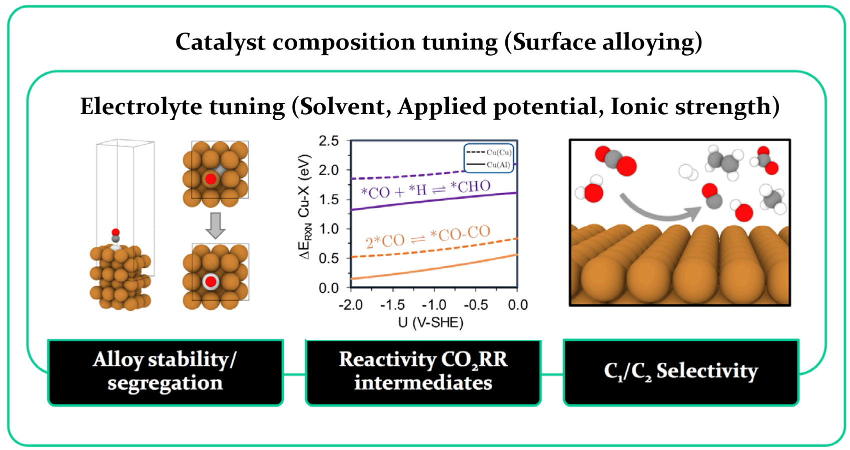
DFT–Continuum modeling via ESM-RISM



Example: Cu(100) + 0.1 M LiOH at negative bias



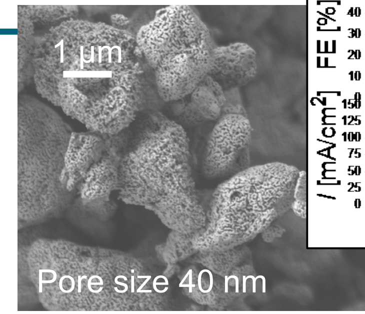
Environment and catalyst tuning to lower the activation barrier of key rate limiting steps

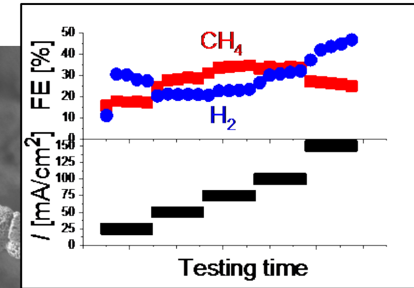


Overall Simulation Approach:
Improve the energy efficiency and selectivity of Cu-based catalysts by manipulating the potential energy landscape through tuning the composition, morphology, and environment of the catalyst.

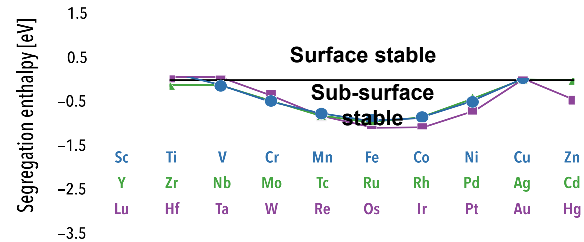
Results and Accomplishments

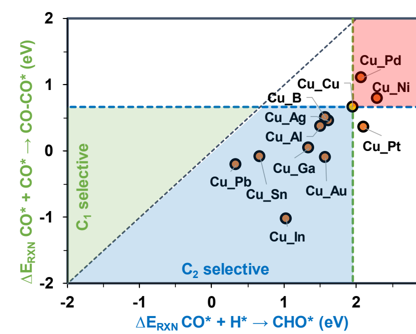
- Developed and tested nanostructured Cu catalysts (3 different catalyst morphologies/surface areas) for CO_2 reduction reactivity and selectivity.
- Tested catalyst integration in Opus-12 electrolyzer; down-selected np-Cu particle catalyst prepared by ball milling as the most promising system (FY-C_2H_4 30% vs 51% @250 mA/cm² for optimized Opus-12 catalyst)
- Developed capability to screen the effect of environment on the energy landscape
- Developed microkinetic model for CO_2 reduction on Cu
- Evaluated stability of surface composition of dilute Cu alloys under reaction conditions.
- Alloying induced thermodynamic C1/C2 selectivity changes up to 10 orders of magnitude relative to Cu
- Prepared and tested 4 (out of 7) dilute Cu-based alloy systems
- Next: Combine dilute alloy and nanoporous Cu particle technology; Optimize catalyst composition and integration; Test performance (Goal FY-C_2H_4 > 70% at >250 mA/cm²)

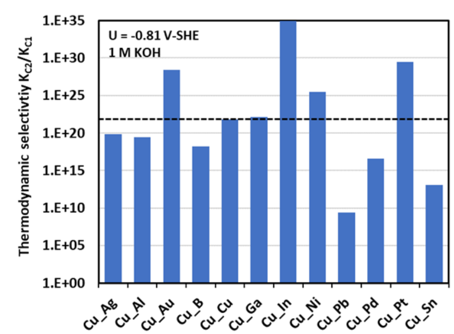




30 (70) % Faradaic efficiency for methane (hydrocarbon) formation at 100 mA/cm²







Transition (beyond DOE assistance)

- The main commercial partner is Opus 12, which is developing the first commercial PEM CO_2 electrolyzers. They have an existing 5 kW CO2 electrolyzer that they developed with a manufacturing partner in the US. The team has been involved with the project by testing the catalyst made a LLNL within their scalable, CO2 reactors.
- Opus 12's commercialization strategy will be to introduce one molecule at a time to the market, then scale that system to larger capacities before introducing the next molecule. Opus 12's addressable market will ultimately touch on a $300 billion cross-section of the petrochemicals sector, with the potential to offset over 1 billion tons of CO_2 emissions over time.