Electrochemical Conversion of Carbon Dioxide to Alcohols

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Department of Chemical and Biomolecular Engineering
University of Delaware, Newark, DE (USA)

DOE/NETL Project Kick-Off Meeting, Morgantown, WV
July 10, 2017
Electro-fuels

CO₂ or H₂O

My research

Representative publications:
Faculty at UD (CO₂ related)

**Feng Jiao**
Associate Professor  
Chemical & Biomolecular Engineering  
Expertise:  
Electrocatalysis, CO₂ Utilization, Nanomaterials, Batteries.

**Raul Lobo**
Claire D. LeClaire Professor  
Chemical & Biomolecular Engineering  
Expertise:  
Heterogeneous Catalysis, CO₂ Capture, Zeolites, Biomass.

**Yushan Yan**
Distinguished Engineering Professor  
Chemical & Biomolecular Engineering  
Expertise:  

**Dionisios G. Vlachos**
Allan and Myra Ferguson Professor  
Chemical & Biomolecular Engineering  
Expertise:  
Heterogeneous Catalysis, Computational Modeling, Biomass.

**Bingjun Xu**
Assistant Professor  
Chemical & Biomolecular Engineering  
Expertise:  
Electrocatalysis, Spectroscopies, Thermochemical Cycles.
Key capabilities: CO$_2$ capture and utilization
Introduction to CO₂ Electrolysis

Key half reactions:
Oxidation: \[ 2H₂O \rightarrow 4H^+ + 4e^- + O₂ \]
Reduction: \[ CO₂ + 2H^+ + 2e^- \rightarrow CO + H₂O \]
or \[ 2H^+ + 2e^- \rightarrow H₂ \]

Carbon monoxide:
- 2-electron process
  - low electricity consumption
- Gas at ambient conditions
  - easy to separate from liquid
- Important feedstock for existing chemical processes
- High selectivity (>90%, Ag) can be achieved.

Other products:
- Formate/formic acid (80%, Sn)
- Ethanol (15-20%, Cu)
- Propanol (15-20%, Cu)
- Acetaldehyde (20-30%, Cu)

Technical Challenges:
- **Selectivity**
  - multiple pathways with similar potentials
- **Overpotential** (energy penalty)
  - additional energy cost

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<table>
<thead>
<tr>
<th>Reaction</th>
<th>E (V) vs. SHE</th>
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<tbody>
<tr>
<td>CO₂ + 2H⁺ +2e⁻ → HCOOH</td>
<td>-0.61</td>
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<tr>
<td>CO₂ + 2H⁺ +2e⁻ → CO + H₂O</td>
<td>-0.53</td>
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<tr>
<td>CO₂ + 4H⁺ +4e⁻ → C + 2H₂O</td>
<td>-0.20</td>
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<tr>
<td>CO₂ + 4H⁺ +4e⁻ → HCHO + H₂O</td>
<td>-0.48</td>
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<tr>
<td>CO₂ + 6H⁺ +6e⁻ → CH₃OH + H₂O</td>
<td>-0.38</td>
</tr>
<tr>
<td>CO₂ + 8H⁺ +8e⁻ → CH₄ + 2H₂O</td>
<td>-0.24</td>
</tr>
</tbody>
</table>

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Electrocatalysts: CO$_2$ to Ethanol

\[ CO_2 + 2H^+ + 2e^- \rightarrow CO + H_2O \quad E^0 = -0.10 \, V \]
\[ CO_2 + 8H^+ + 8e^- \rightarrow CH_4 + 2H_2O \quad E^0 = 0.17 \, V \]
\[ 2CO_2 + 12H^+ + 12e^- \rightarrow C_2H_5OH + 3H_2O \quad E^0 = 0.09 \, V \]

Liquid products (alcohols) are ideal:
- High volumetric energy density, portability
- Valuable, easily incorporated in current infrastructure

No single electrocatalyst can convert CO$_2$ into ethanol with an appreciable performance.

Copper is the only monometal that catalyzes CO$_2$ reduction to hydrocarbons in aqueous.

**Proposed mechanism:**

- The presence of CO$_{ad}$ is crucial for C-C coupling to form C$_2$/C$_3$ compounds.
- Enhancing C$_2$/C$_3$ selectivity by tuning CO binding energy cannot be achieved using a single electrocatalyst.

Project Objectives and Approach

1) Development of critical components for an electrochemical system that is able to convert CO$_2$ into C$_2$/C$_3$ alcohols
2) Demonstration of key functions of an integrated electrochemical system for CO$_2$ conversion using flue gas from coal-fired power plants
3) Full analysis of economics and life-cycle of the CO$_2$ electrolysis technology for CO$_2$ emissions mitigation from coal-fired power plants

Our Approach:
Project Management

UD Research Office
Dawn Jory
Contract & Grant Specialist

NETL Program Manager

Project Management
Project Lead: Jiao

Task 1.0
Project Management and Planning

Task 2.0
CO$_2$ Electrolyzer Subsystem

Task 3.0
CO Electrolyzer Subsystem

Task 4.0
Integration of Prototype

Task 5.0
Economics & Lifecycle Analysis

Task 1.3
Safety Analysis
Proposed Two-stage Process and its Chemistry

**CO₂ → Electrochemical CO₂ Reduction → CO → Electrochemical CO Reduction → Alcohols**

Subsystem: CO₂ electrolyzer
- Cathode reaction: \(\text{CO}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{CO} + \text{H}_2\text{O}\)
- Anode reaction: \(2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + \text{O}_2 + 4\text{e}^-\)
- Overall reaction: \(2\text{CO}_2 \rightarrow 2\text{CO} + \text{O}_2\)

Subsystem: CO electrolyzer
- Cathode reaction: \(2\text{CO} + 8\text{H}^+ + 8\text{e}^- \rightarrow \text{C}_2\text{H}_6\text{O} + \text{H}_2\text{O}\)
- Anode reaction: \(2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + \text{O}_2 + 4\text{e}^-\)
- Overall reaction: \(2\text{CO} + 3\text{H}_2\text{O} \rightarrow \text{C}_2\text{H}_6\text{O} + 2\text{O}_2\)
Proposed Two-stage Process and its Chemistry

**Subsystem: CO₂ electrolyzer**
Cathode reaction: \( \text{CO}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{CO} + \text{H}_2\text{O} \)
Anode reaction: \( 2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + \text{O}_2 + 4\text{e}^- \)
Overall reaction: \( 2\text{CO}_2 \rightarrow 2\text{CO} + \text{O}_2 \)

**Subsystem: CO electrolyzer**
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Overall reaction: \( 2\text{CO} + 3\text{H}_2\text{O} \rightarrow \text{C}_2\text{H}_6\text{O} + 2\text{O}_2 \)
Silver electrocatalyst for CO$_2$ reduction

• Highly selective for CO$_2$ to CO conversion
• Ag ($15/oz$.) is cheaper than Au ($1162/oz$.)
• Stable in aqueous electrolytes

A step surface of Ag is more favorable for electrochemical CO$_2$ reduction.

- High surface area: 150 times larger than thin film
- Rich step sites on the highly curved surface
- Synthetic method can be extended to other metals
- Self-supporting catalyst: no conductive substrate
CO$_2$ electrolyzer prototype

- Funded by NASA
- Built in June 2016
- 36 electrochemical cells arranged in 6 stacks
- 2 membrane separator
- Engineering safety controls
- Customized software for data acquisition
CO₂ Electrolyzer Prototype

- CO₂ Process Rate: 450 grams/day
- Power Consumption: 50 watts (per stack)
Task 2.0: CO₂ Electrolyzer Subsystem Development

Subtask 2.1: Conceptual Design of CO₂ Electrolyzer Subsystem
- Process control & optimization

Subtask 2.2: Development of Nanostructured Ag Cathode
- High current density (production rate) & low overpotential (energy penalty)
- High selectivity towards CO
- Robust & stable

Subtask 2.3: Development of Non-Precious Metal-based Anode
- High current density & low overpotential
- Robust & stable

Subtask 2.4: Development of Gas/Liquid Contactor and Gas/Liquid Separator
- CO₂ delivery to catalyst (active site)
- Product separation

Subtask 2.5: Fabrication of CO₂ Electrolyzer Subsystem
- Scale up
- Integration

Subtask 2.6: Evaluation of CO₂ Electrolyzer Subsystem Performance

Subtask 2.7: Alternative CO₂ Electrolyzer Design for Performance Enhancement
- Boost performance using alternative designs

The Key Performance Parameters for the proposed CO₂ electrolyzer subsystem are:
- CO Faradaic efficiency higher than 70% at the subsystem level
- Cell voltage less than 3.0 V with a total current of 5 A
- Continuous operation for more than 3 hours with less than 20% performance loss
Proposed Two-stage Process and its Chemistry

\[ \text{CO}_2 \rightarrow \text{Electrochemical CO}_2 \text{ Reduction} \rightarrow \text{CO} \rightarrow \text{Electrochemical CO Reduction} \rightarrow \text{Alcohols} \]

Subsystem: CO$_2$ electrolyzer
Cathode reaction: \( \text{CO}_2 + 2\text{H}^+ + 2\text{e}^- \rightarrow \text{CO} + \text{H}_2\text{O} \)
Anode reaction: \( 2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + \text{O}_2 + 4\text{e}^- \)
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Overall reaction: \( 2\text{CO} + 3\text{H}_2\text{O} \rightarrow \text{C}_2\text{H}_6\text{O} + 2\text{O}_2 \)
Annealed Cu Particles for CO Reduction

Cu particles (≈ 1 μm) were annealed at 500 °C for 6 hrs and deposited on carbon paper GDL (1 mg/cm²).

- Selective towards alcohols at moderate overpotentials
- Max. current density: 0.5 mA/cm² with n-PrOH selectivity of 10%
- Low current density is due to the low solubility of CO in the aqueous electrolyte
Flow cell design for CO to alcohols

The low solubility of CO in aqueous electrolyte motivates a direct gas feed.

Flow cell schematic:

A gas diffusion layer allows CO to be fed directly to the catalyst/electrolyte interface.
Task 3.0: Development of CO Electrolyzer Subsystem

Subtask 3.1: Conceptual Design of CO Electrolyzer Subsystem
• Process control & optimization

Subtask 3.2: Development of Nanostructured Cu Cathode
• High current density (production rate)
• High selectivity towards alcohols
• Robust & stable

Subtask 3.3: Development of CO Electrolysis Flow Cell and Multi-cell Stack
• Electrode/electrolyte interface

Subtask 3.4: Fabrication and Evaluation of CO Electrolyzer Subsystem
• Scale up
• Integration

The Key Performance Parameters for the proposed CO electrolyzer subsystem are:
• Alcohol Faradaic efficiency higher than 40% at the subsystem level
• Cell voltage less than 3.0 V with a total current of 10 A
• Continuous operation for more than 3 hours with less than 20% performance loss
Subsystem integration efforts:
- CO/CO₂ separation strategy
- Pressures and flow rates between subsystems
- Production rates of subsystems
- Process control & safety
- System compatibility with flue gases
Task 4.0: Integration and Evaluation of the Complete Electrolyzer System

Subtask 4.1: Conceptual Design of Integrated Electrolyzer System for $C_2/C_3$ Alcohol Production
  • Process control & optimization
Subtask 4.2: Fabrication and Integration of CO$_2$ Electrolyzer and CO Electrolyzer Subsystems
  • Scale up (matches of production rates)
  • Integration (balance of pressure, flow rate, and heat)
Subtask 4.3: Evaluation of the Performance of the Complete Electrolyzer System
Subtask 4.4: Optimize the Performance of the Complete Electrolyzer System
Subtask 4.5: Investigation of Flue Gas Compatibility
  • SO$_x$, NO$_x$, N$_2$, O$_2$, H$_2$ contamination effects

The Key Performance Parameters for the integrated electrolyzer system are as follows:
  • Overall energy efficiency higher than 28% at the system level
  • Continuous operation for more than 3 hours with less than 30% performance loss
Task 5.0: Economics and Life-cycle Analysis

Subtask 5.1: Technical Scalability and Economic Feasibility Study
Subtask 5.2: Life-cycle Analysis
Subtask 5.3: Technology Gap Analysis

The team will conduct a process scalability assessment, an economic feasibility study, lifecycle analysis and technology gap analysis based on the data collected from the R&D project. The compiled results from these studies will be delivered at the conclusion of the task.
## Project Schedule and Milestones

<table>
<thead>
<tr>
<th>Task 1.0 - Project Management and Planning</th>
<th>Start Date</th>
<th>End Date</th>
<th>Cost</th>
<th>Budget Period 1</th>
<th>Budget Period 2</th>
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### Project Schedule and Milestones

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<td>Subtask 3.3 - Development of CO Electrolysis Flow Cell and Multi-cell Stack</td>
<td>3/1/2018</td>
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<td>Subtask 3.4 - Fabrication and Evaluation of CO Electrolyzer Subsystem</td>
<td>6/1/2018</td>
<td>11/30/2018</td>
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**Milestones**

- Milestone 3.a - Complete the Conceptual Design of CO Electrolyzer: X
- Milestone 3.b - Complete the Fabrication of CO Electrolyzer Subsystem: X
- Milestone 3.c - Complete the Evaluation of CO Electrolyzer Subsystem: X

<table>
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<tr>
<th>Task 4.0 - Integration and Evaluation of the Complete Electrolyzer System</th>
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**Milestones**

- Milestone 4.b - Complete the Fabrication of the Integrated Electrolyzer System: X
- Milestone 4.e - Complete the Flue Gas Compatibility Investigations: X

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<tr>
<th>Task 5.0 - Economics and Life-cycle Analysis</th>
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**Milestones**

- Milestone 5.a - Complete the Cost Analysis: X
- Milestone 5.b - Updated Performance Metrics: X
- Milestone 5.c - Complete the Life-cycle Analysis: X
## Key Decision Points

<table>
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<th>Decision Point</th>
<th>Date</th>
<th>Success Criteria</th>
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<tbody>
<tr>
<td>Go/no-go Decision Point #1</td>
<td>11/30/2018</td>
<td>[1] Completion of subsystem fabrication and evaluation. [2] CO$_2$ electrolyzer subsystem meets all the Key Performance Parameters: CO Faradaic efficiency higher than 70% at the subsystem level, cell voltage less than 3.0 V with a total current of 5 A, continuous operation for more than 3 hours with less than 20% performance loss. [3] CO electrolyzer subsystem meets all the Key Performance Parameters: alcohol Faradaic efficiency higher than 40% at the subsystem level, cell voltage less than 3.0 V with a total current of 10 A, continuous operation for more than 3 hours with less than 20% performance loss.</td>
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<tr>
<td>Go/no-go Decision Point #2</td>
<td>5/31/2020</td>
<td>[1] Completion of system fabrication, integration, and evaluation. [2] Demonstration of the performance of integrated electrolyzer system meeting all the Key Performance Parameters: overall energy efficiency higher than 20% at the system level, continuous operation for more than 3 hours with less than 30% performance loss.</td>
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## Project Funding Profile

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<th><strong>Budget Period 1</strong> 06/01/2017 - 11/30/2018</th>
<th><strong>Budget Period 2</strong> 12/01/2018 - 05/31/2020</th>
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<td>Cost Share</td>
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Anticipated Outcomes

• The feasibility of the proposed approach for reduction of CO₂ emission will be demonstrated.
• Two subsystems, i.e., CO₂ electrolyzer and CO electrolyzer, will be designed, fabricated, and evaluated. The key functions will be demonstrated for the integrated system in the laboratory environment.
• Economic and life-cycle models will be established and assessed using experimental data.
• A Technology Readiness Level 4 (TRL 4) will be reached at the end of project.
Acknowledgements

Collaborators:
Dion Vlachos, Yushan Yan, Bingjun Xu, John Xiao, Robert Birkmire, University of Delaware
Jingguang Chen, Columbia University
Ken Burke, NASA Glenn Research Center
Thank you