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Integrated Capture and Electrocatalytic Conversion of Carbon Dioxide to Alcohols

November 2019

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Prepared for the U.S. Department of Energy under Contract DE-AC05-76RL01830

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Abstract

The goal of this SEED LDRD project was to perform proof of concept studies that would provide proof of concept data that would demonstrate viability of an R&D approach to develop an innovative, scalable electrochemical technology for synthesis of liquid fuels and chemicals (e.g., ethanol) through coupling of H₂ produced from water splitting and CO₂ captured in solvents from industrial waste streams.

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1.0 Research

The project team attempted electrochemical conversion of CO₂ in a capture solvent using a typical sandwich electrolysis cell. The cell uses an iridium oxide coated porous carbon electrode, anion exchange membrane (Fumatech FAA-3) and Ag/C catalyst as anode, electrolyte and cathode, respectively. In our electrolysis cell, the capture solvent with CO₂ loading and 0.2 M NaOH water solution are used as catholyte and anolyte, respectively. A peristaltic pump circulated the catholyte and anolyte to accomplish the electrode reaction at cathode and anode, respectively.

Anode:
$$40H^- - 1e^- \rightarrow 0_2 + 2H_2O \quad E^0 = 0.48V$$
 (1)
Cathode: $kCO_2 + aH_2O + be^- \rightarrow P + cOH^-$ (2)

Product name (P)	k	a	b	с	Eo
CO	1	1	2	2	-0.1 V
H ₂	0	1	1	1	0 V
CH ₄	1	6	8	8	-0.34 V

Table 1. Main products of the CO2 electrolysis process

Initially, the feed gas of humidified CO₂ was flowed into cathode side, the voltage of 3V was maintained when the cell was held at a constant current density of 30 mA/cm2, as shown in Fig.1a. The main reduction products of CO₂ electrolysis are hydrogen and CO (Table 2), indicating that Ag/C is a good catalyst for CO₂ reduction. When the feed changes to capture solvent with 2.5 wt% CO₂ loading, the results are shown in Fig. 1b, and the current density for our cell is approximately 75 mA/cm2 when the cell was charged at 3V. However, only small amount of CH₄ could be observed through GC analysis, and the main reduction product is hydrogen. The voltage was then jumped up to 3.5 V as increasing the current density to 110 mA/cm2. But again, the main reduction product is still hydrogen, and only small amount of CO and CH₄ could be detected through GC analysis on gas samples.

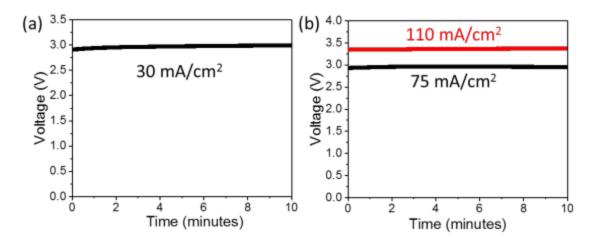


Figure 1. The cell's electrochemical testing with the feed gas of humidified CO_2 (a) and capture solvent with 2.5 wt% CO_2 loading (b) at various current densities at room temperature. Electrolyte flow rate: 20mL/min.

	Applied voltage (V)	Current (mA/cm ²)	Product
Humidified CO ₂ (CO ₂ flow through water at RT) Flow rate (20 ml/min)	3	30	26.8% H ₂ , 5.1% CO and 66% CO ₂
Liquid (2.5 CO ₂ wt%) Flow rate (20 ml/min)	3	75	99.2% $\rm H_2,$ no detectable amount of CO, 0.1% $\rm CH_4$ and 0.7% $\rm CO_2$
Liquid (2.5 CO ₂ wt%) Flow rate (20 ml/min)	3.5	110	99.5% $\rm H_{2},$ <0.1% CO, <0.1% $\rm CH_{4}$ and 0.4% $\rm CO_{2}.$

2.0 Summary

In summary, the CO₂ electrolysis test using capture solvent was performed and CH₄ is being produced, albeit at a very slow rate. This SEED project indicates that CO₂ could be electrochemically reduced in a carbon capture solvent, however, the electro-reduction rate of CO₂ is still quite low and the test conditions (CO₂ loading, operation temperature, and various catalyst, et. al) would need to be optimized to improve the electrochemical performance of CO₂ electrolysis in a working process. The project team will use these results in a proposal for CO₂ utilization to The Fossil Energy Office at DOE in FY20.

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