Electrocatalytic CO_2 Reduction in Capture Solvents

Sophie B. Rubashkin, Eric S. Wiedner, Aaron M. Appel
sophie.rubashkin@pnnl.gov, Pacific Northwest National Lab

Motivation

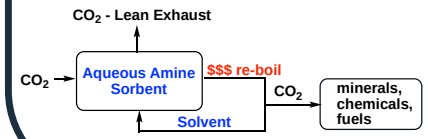
CO_2 capture and utilization (CCU) is a vital tool in the fight against climate change

Mature technologies utilize aqueous scrubbers that are costly to recycle

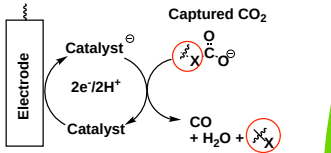
Organic liquids with high CO_2 binding affinity have lower energy cost

In situ CO_2 reduction to CO in capture solvent generates a valuable platform chemical via a low emission pathway

Aqueous Amine Scrubbing



Electrocatalytic CCU



Research Highlights and Impact

Carbon Capture and Utilization (CCU) is a strategy to help mitigate CO_2 emissions from industrial post-combustion flue gas

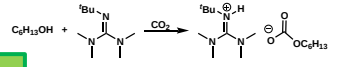
CO_2 → **Cleaner emissions**

→ **CO**

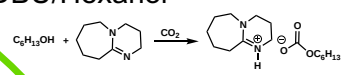
Electrochemical CO_2 reduction to CO via CCU is a green approach to the synthesis of platform chemicals used in detergents, fragrances, pharmaceuticals and more

Results

*t*BuTMG/Hexanol

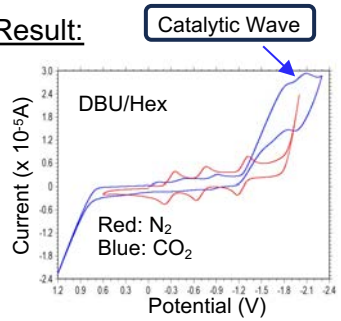


DBU/Hexanol



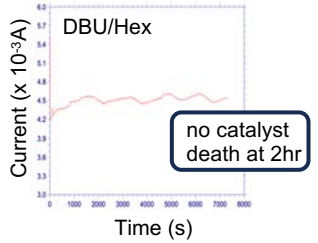
Base/Alcohol mixtures uptake 130 wt % CO_2 and release upon bubbling with N_2

CV Result:



CPE Results*:

CO_2 Reduction at -1.9V vs $Ag^{0/+}$



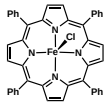
*t*BuTMG/Hex: 7 TON CO

DBU/Hex: 8 TON CO

*2 hr rxn time

Materials And Methods

Electrocatalysis uses an applied potential to generate the energetic driving force for the reaction

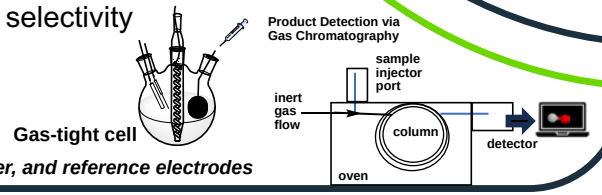


Catalysts, like this iron porphyrin, enable CO_2 reduction to CO over and over without getting used up during the reaction

Cyclic Voltammetry (CV)– measures electron transfer events across a range of potentials on a fast time scale

Controlled Potential Electrolysis (CPE)– batch reaction at single potential for determination of products and selectivity

Activity reported as catalyst turnover numbers (TON)



equipped with working, counter, and reference electrodes

Conclusion & References

Organic solvent mixtures containing both basic and alcohol constituent groups have **a high affinity for CO_2 uptake** while maintaining **low-energy recyclability pathways**

Molecular iron catalysts can **electrochemically transform captured CO_2 to CO directly in the capture solvent**

While proof of concept has been demonstrated, **further studies are needed to optimize the catalytic activity and maximize CO_2 conversion under capture conditions**

(1) Heldebrant, D. J.; Yonker, C. R.; Jessop, P. G.; Phan, L *Energy & Environmental Science* **2008**. (2) Bhugun, I.; Lexa, D.; Savéant, J.-M. *J. Am. Chem. Soc.* **1996**, *118*, 1769-1776. (3) Siegel, R. E.; Pattanayak, S.; Berben, L. A. *ACS Catalysis* **2022**, 766-784.

U.S. DEPARTMENT OF ENERGY