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Electrochemically Regenerated Solvent for Direct Air Capture with Cogeneration of Hydrogen at Bench-Scale

DE-FE0032125

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U.S. Department of Energy

National Energy Technology Laboratory

Carbon Management Project Review Meeting

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Project Overview

Funding: DOE $1,313,887 and Cost Share $329,084

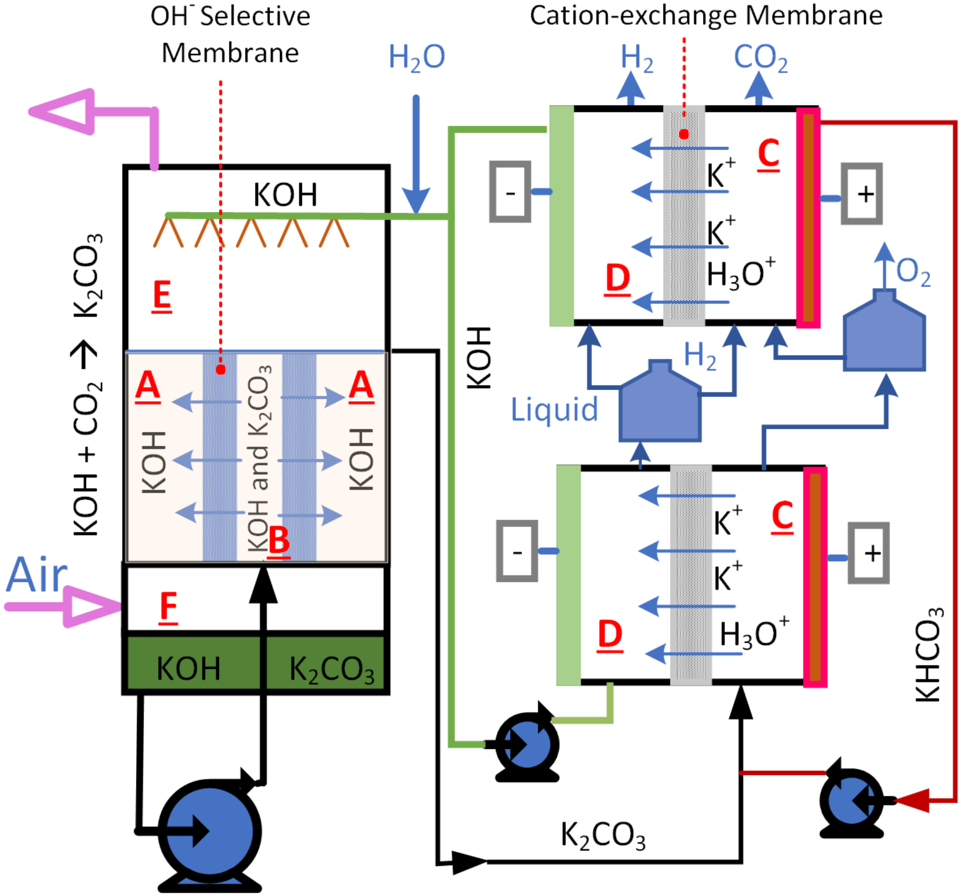
Overall Project Performance Dates: 10/1/2021 – 9/31/2023

Project Participants: University of Kentucky, Vanderbilt University, and EPRI

Project Objectives

- Developing a capture technology to extract CO_2 from the atmosphere that reduces the cost of capture through:
- Developing a 2-unit process operation with $\geq$90% CO_2 capture at gaseous pressure drop $\leq$0.2 psi in the CO_2 absorber while regenerating CO_2 at less than 3.5 V in the ER by using a catalytic electrode
- Achieving a H^+/K^+ crossover (through a cation-exchange membrane) ratio of $\leq$15% in the ER by optimizing a flow channel design, and spacer thickness;
- Demonstrating a continuous and reliable DAC process at the air flow rate of 10 cfm with $\geq$90% CO_2 capture efficiency for $\geq$100 hours, thereby establishing the data for the next-scale development.

Technology Background



A: Permeate chamber for CO_2 absorption
B: Feed chamber for mixture of KOH and K_2CO_3
C: Positive chamber D: Negative chamber
E: Open-tower for CO_2 absorption
F: Air entrance and liquid sump

Key Electrochemical Reactions

(0V, $2H_2O$ + electrons = $H_2 + 2OH^-$)
(-1.2V, $2H_2O = O_2 + 4H^+$ + electrons)

CO_2 Capture

$$CO_2 + 2OH^- = CO_3^{2-} + H_2O$$

(Fast)

CO_2 Release

$$H^+ + CO_3^{2-} \rightarrow HCO_3^-$$

$$H^+ + HCO_3^- \rightarrow CO_2 + H_2O$$

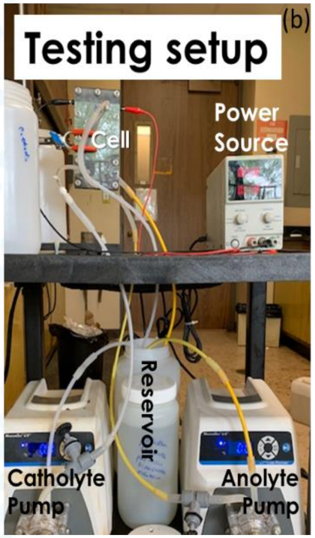
or

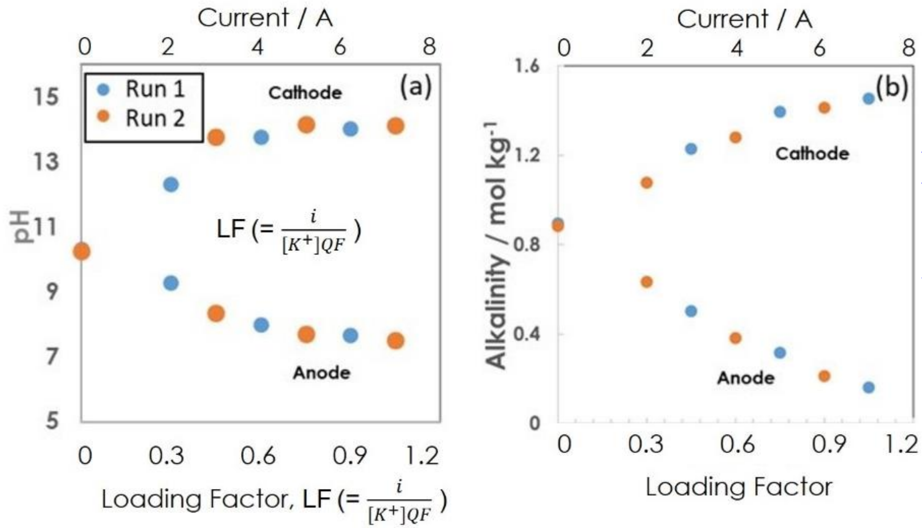
$$2HCO_3^- \rightarrow CO_2 + CO_3^{2-} + H_2O$$

via flashing

- Fast Kinetics Solvent
- Simplified Process and Operation
- Byproducts of H_2 and O_2 for sale

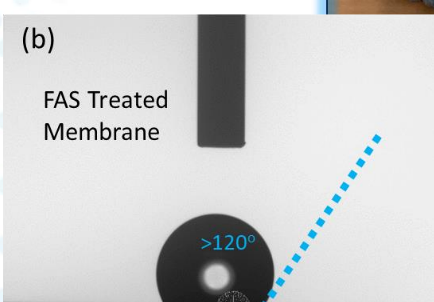
Technology Background- UKy Previous Results

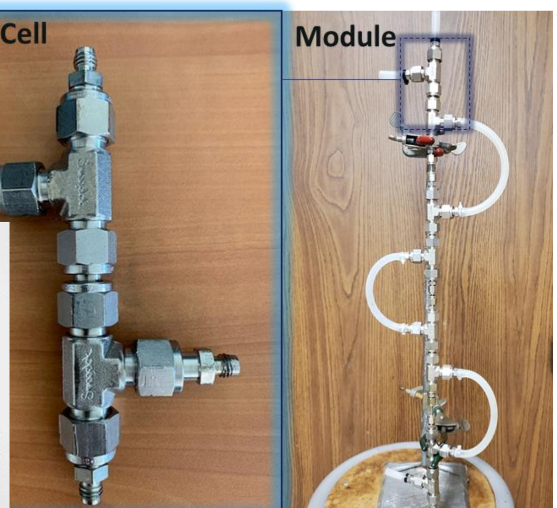


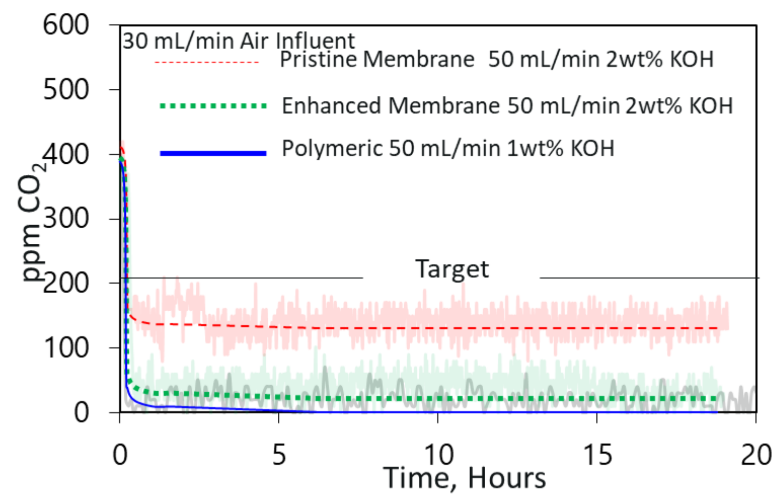


Developed electrochemical reactor for pH swing

Developed and utilized membrane contactor for hydroxide-based DAC







Project Scope

Work plan

- Design, fabrication, and research on hybrid absorber (HA), electrochemical regenerator (ER), and integrated process (HA + ER) (Task 2-4, BP1)
- Evaluation of integrated HA and ER including parametric and long-term evaluation and process modeling (Task 5, BP2)
- Process analysis including TEA and LCA and develop TMP (Task 1, 6 & 7)

Project Success Criteria		
Decision Point	Date	Success Criteria
Completion of BP1	09/30/22	• For MA, ≥60% of OH^- separation from CO_3^{2-} at liquid delivery rate > 2 L/hour • For ER, ≤3.7 of operating voltage; ≤20% of H^+/K^+ crossover ratio through membrane
Project Completion	09/30/23	10 CFM air DAC process with: • <0.2 psi gas-side pressure drop • ≥90% CO_2 capture for ≥100 hours • ≤2.7 V of operating voltage, and • ≤15% of H^+/K^+ crossover ratio through membrane

Membrane Evaluation for OH^- Separation

NFX and NF270 Selected as membranes for Hybrid Absorber

Designation	NFX	NF270
Brand	Synder	Filmtec
Material	Polyamide	Polypiperazine
Water Permeance (LMH/bar)	~4-7.5	~17
$MgSO_4$ Rejection	99 %	99%
NaCl Rejection	40 %	50%

Milestone: ≥60% OH^- permeance from influent stream with CO_3^{2-} in concentrate stream

Achieved 100% OH^- permeance from influent stream with a nominal selectivity ratio > 4

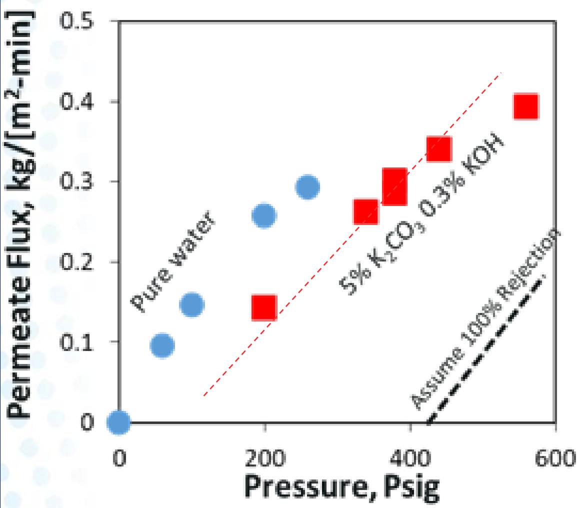
Rejection (1 – [$C_{X,permeate}$/$C_{X,feed}$])

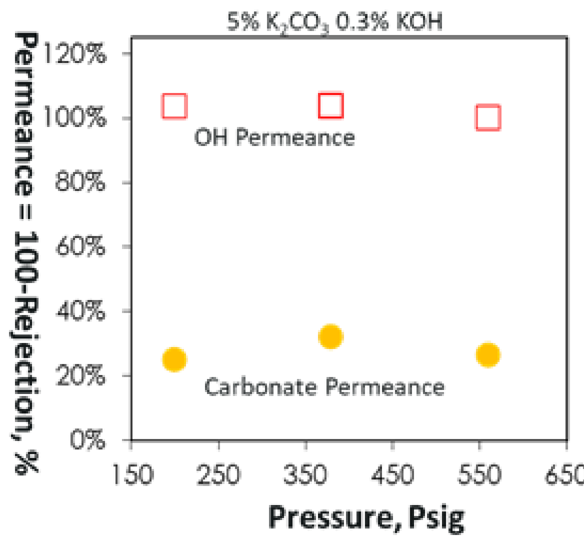
NF270 Membrane			30 LMH Constant Flux		
Start CO_3 Conc.			CO_3 Rej	OH Rej	Selectivity
(%)	Feed / Permeate	pH	(%)	(%)	#
0.1	Feed	11.83	83.90	39.74	3.74
0.1	Permeate	11.61			
0.1	Feed	12.96	64.45	14.89	2.39
0.1	Permeate	12.89			
1	Feed	11.84	84.54	0*	6.47
1	Permeate	11.92			
1	Feed	12.97	77.28	0*	4.40
1	Permeate	12.99			
3	Feed	11.83	78.53	0*	4.66
3	Permeate	12.29			
3	Feed	13.05	76.13	0*	4.19
3	Permeate	13.22			

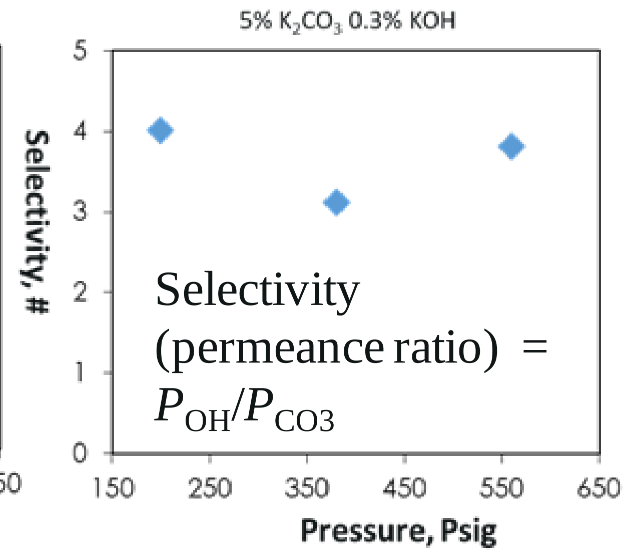
Membrane Evaluation for OH^- Separation

Solvent Circulation: 100 mL/min
Temperature: Room Conditions
Membrane: Synder NFX
Membrane Area: 140 cm^2

Solvent	Osmotic Pressure at 100% Rejection
5% K_2CO_3 0.3% KOH	~421 psig

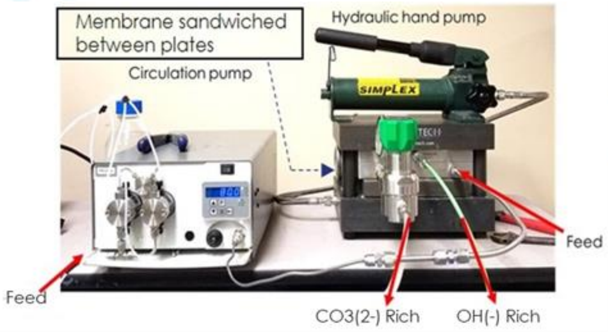






Achieved ~100% OH^- permeance from influent stream with a selectivity ratio of 4

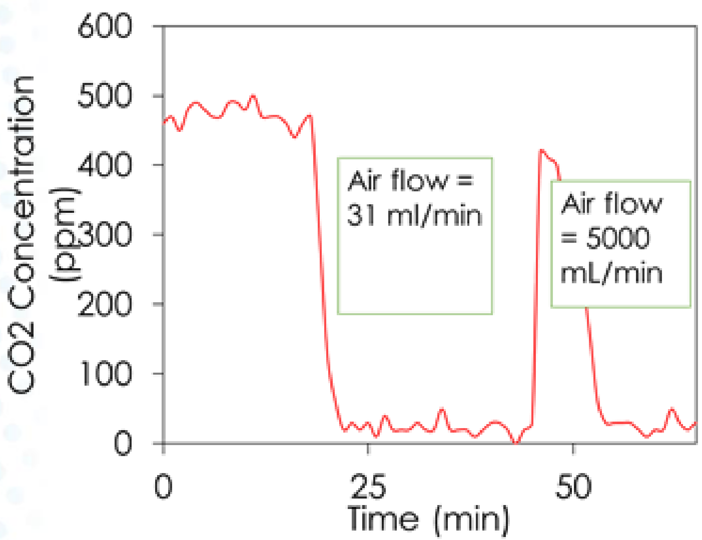
Capture Performance of Nanofiltration

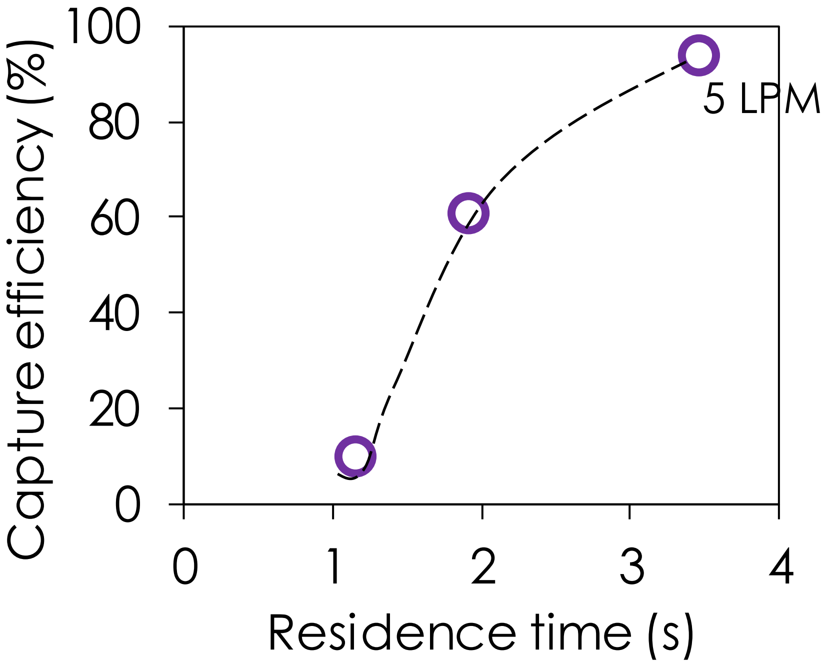


$140\ cm^2$ Flat-sheet Membrane

$7.6\ m^2$ Spiral-wound Membrane

5 wt% K_2CO_3 Feed, NF@ 30 psig back-pressure, CO_2 Fed to Permeate side





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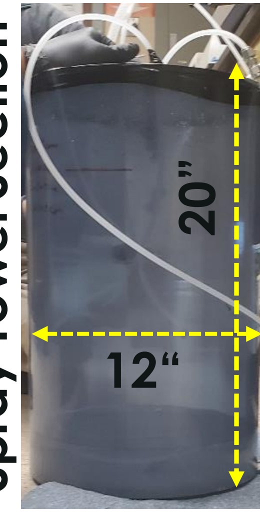
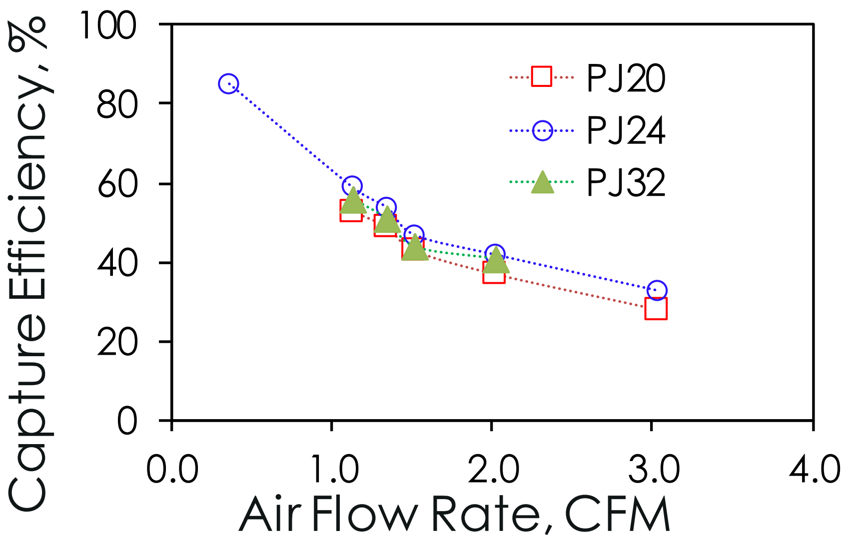
Testing of Capture in Spray Column Section

	PJ20	PJ24	PJ32
2wt% KOH spray rate (L/min)	0.44	0.5	0.65
Nozzle pressure (psig)	120	90	40
Approximate coverage (inches)	12	16	22
Approximate spray height (inches)	6	8	11
Droplet size (µm)	41	50	62
Solution height (inches)	1.5	1.5	1.5
Air inlet above solution (inches)	3	3	3
Nozzle to solution (inches)	15.5	15.5	15.5

Spray Tower Section

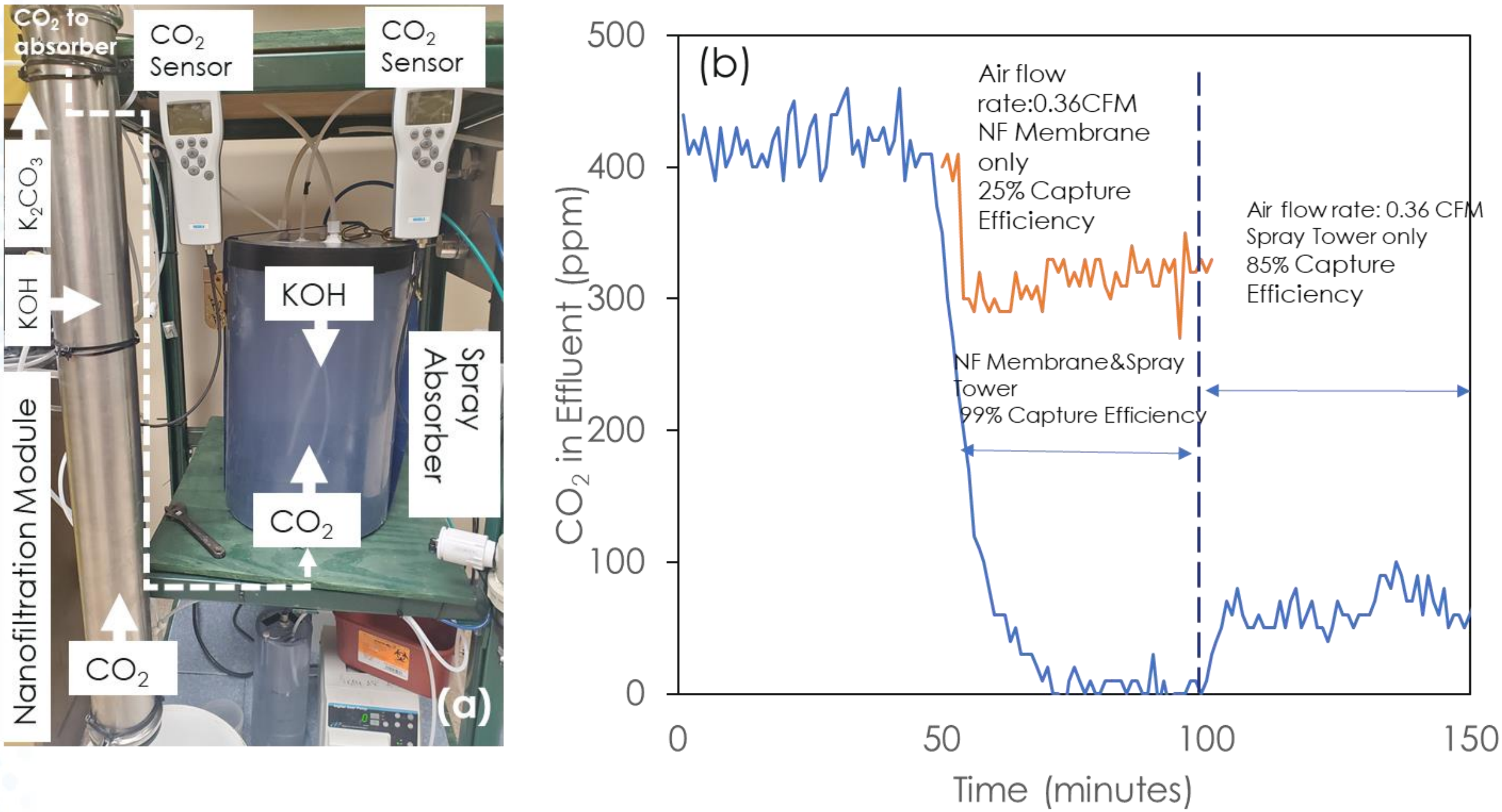




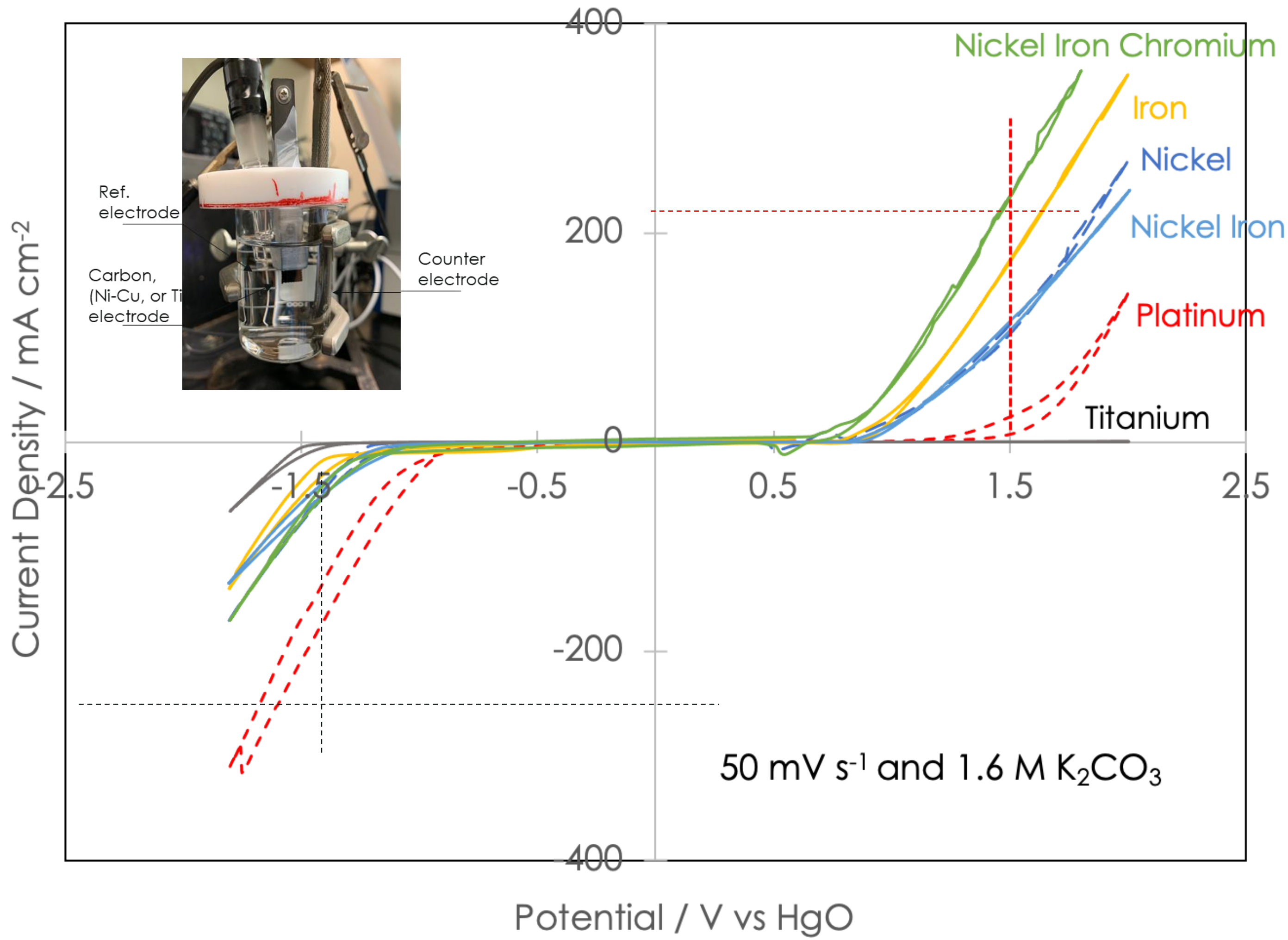
Capture enhanced at low flow (increased residence time) with ~90% achieved at 0.4 CFM Air

Performance of Hybrid membrane System

A. 5 wt% K_2CO_3 Feed, NF@ ~30 psig back-pressure, 0.36 CFM CO_2 Fed to Permeate side
B. CO_2 from NF Permeate side continues to Spray Tower
C. 2 wt% KOH Feed to spray section

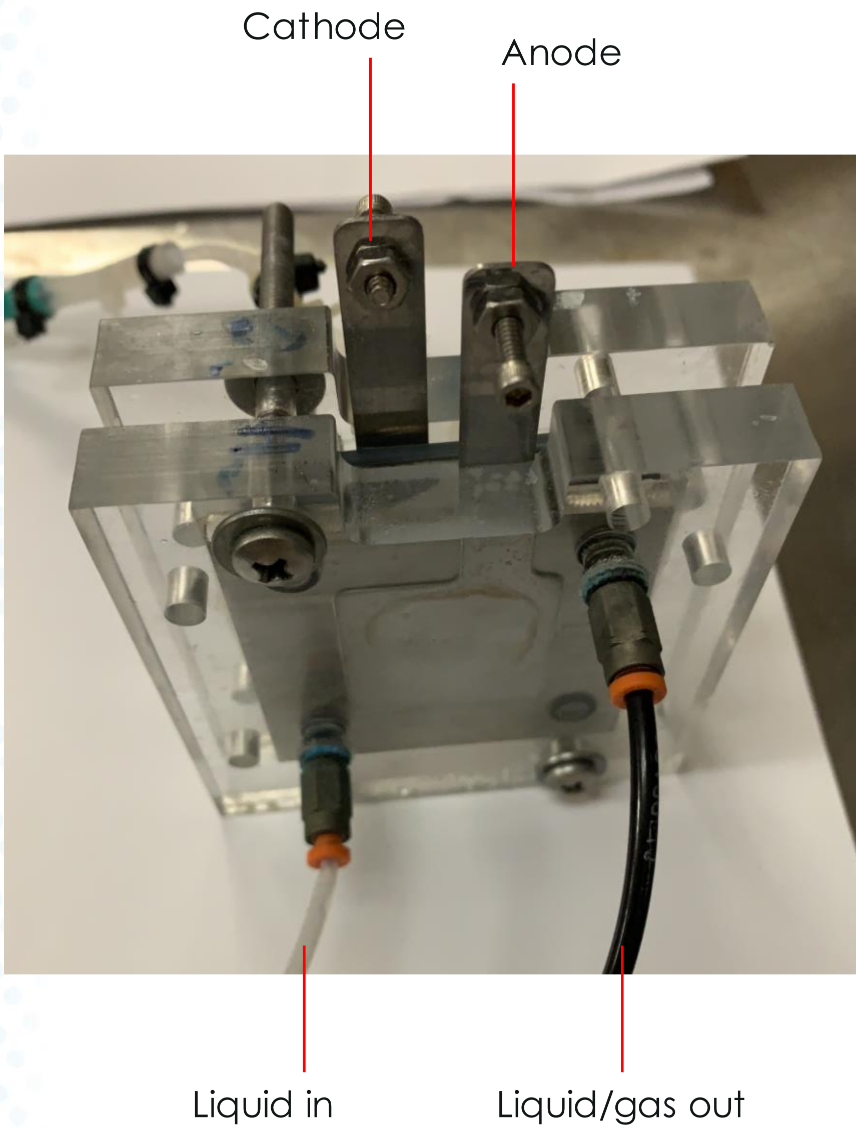


Catalytic Electrode Selection for ER



1. High current density under same potential means less energy to produce same H_2
2. Pt/C Cathode and Inconel (Ni-Fe-Cr) Anode appear as the best combinations

Full Cell ER Testing



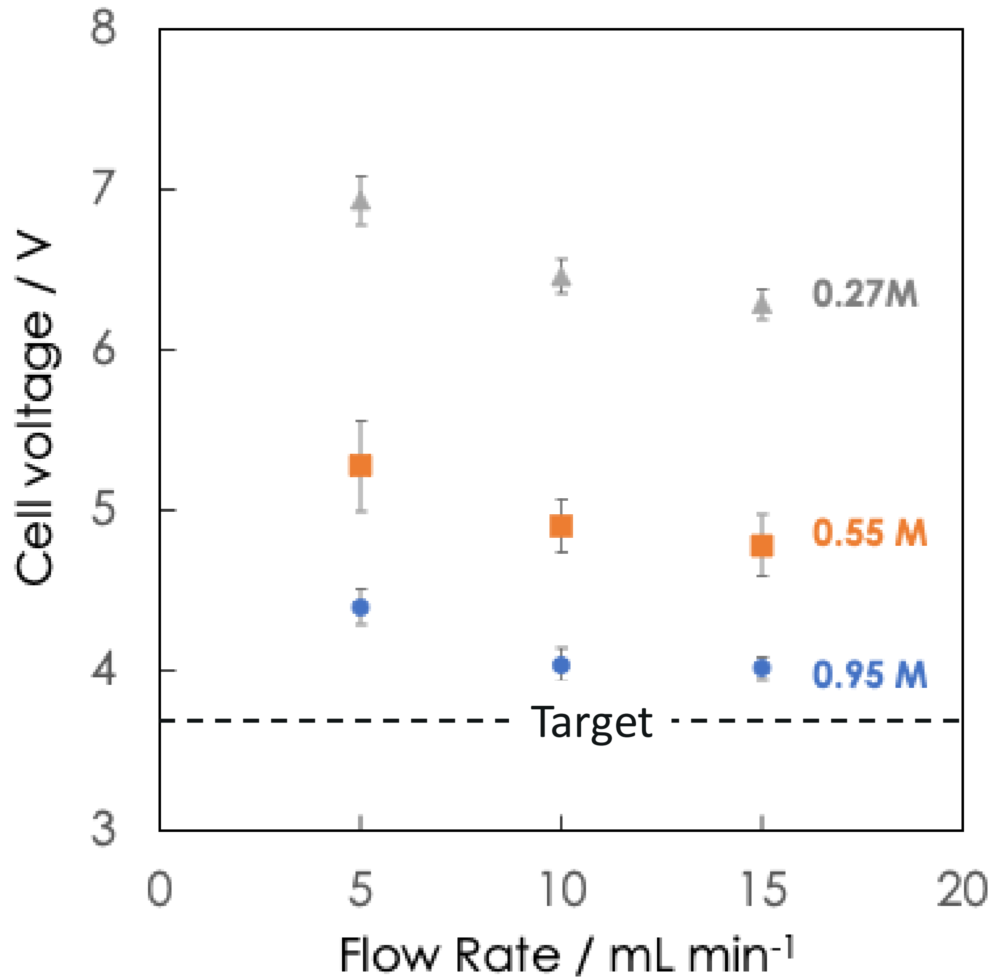
Testing Conditions:

- Inconel anode and Pt-C cathode
- K_2CO_3 circulated at room temperature
- 16 cm^2 active area
- Constant current charging technique was applied, and cell voltage was measured
- Zero electrode space gap to 3 mm gap configuration.

Full ER Cell – Suppressing Voltage loss

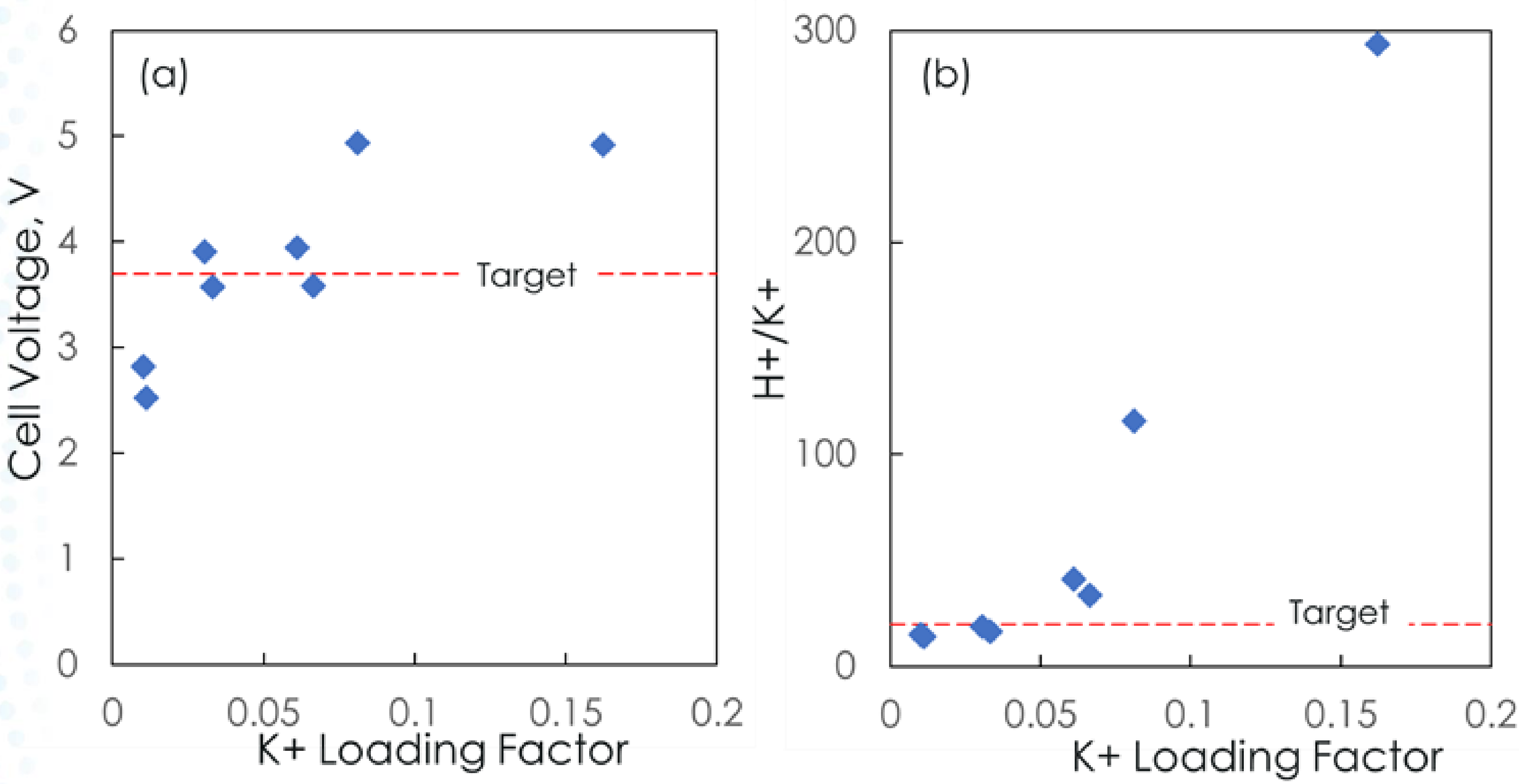
Testing Conditions:

- 0.27 to 0.95M K_2CO_3 was circulated at 5-15 mL min-1 at room temperature;
- Constant current (3A) charging technique was applied, and cell voltage was measured;
- Electrode space gap was 1.5 mm each channel.
- The solvent regenerator was equipped with the Inconel anode and Pt-C cathode.



Concentration has a stronger effect on voltage

Full ER Cell – Suppressing Voltage loss and Mitigating Proton Crossover



Cell voltage (a) and H^+/K^+ (b) as functions of K^+ loading.

H/K increases with increasing loading factor (increasing current, decreasing concentration, decreasing flow rate)

Plans for Testing and Future Development

- Boost hydroxide concentration and gas-liquid contact zones and complete scale up of hybridized membrane absorber for 90% capture from 5-15 CFM feed

- Explore internal integration of MA (Spray Tower with Nanofiltration) via tubular and flat-sheet membranes

- Parametric and long-term continuous testing of integrated absorber and ER at 5-15 CFM Air

- Scale-up of ER system to 5 kg CO_2/hour (4000 CFM Air) and demonstrate negative emissions via solar integration

Summary

(1) NFX and NF270 Nanofiltration membranes show capability to concentrate carbon loading (e.g., CO_3^{2-}) by removing OH- prior to sending ER for regeneration toward reducing energy consumption

(2) Interim results show that the hybrid absorber including spray absorber and selective membrane is an effective absorber for DAC with capture facilitated by KOH solvent solvent

(3) Electrochemical regeneration performance and energy requirement of the ER were found to strongly depend on inter-electrode spacing and loading factor - solvent concentration, flow rate and current

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Acknowledgements

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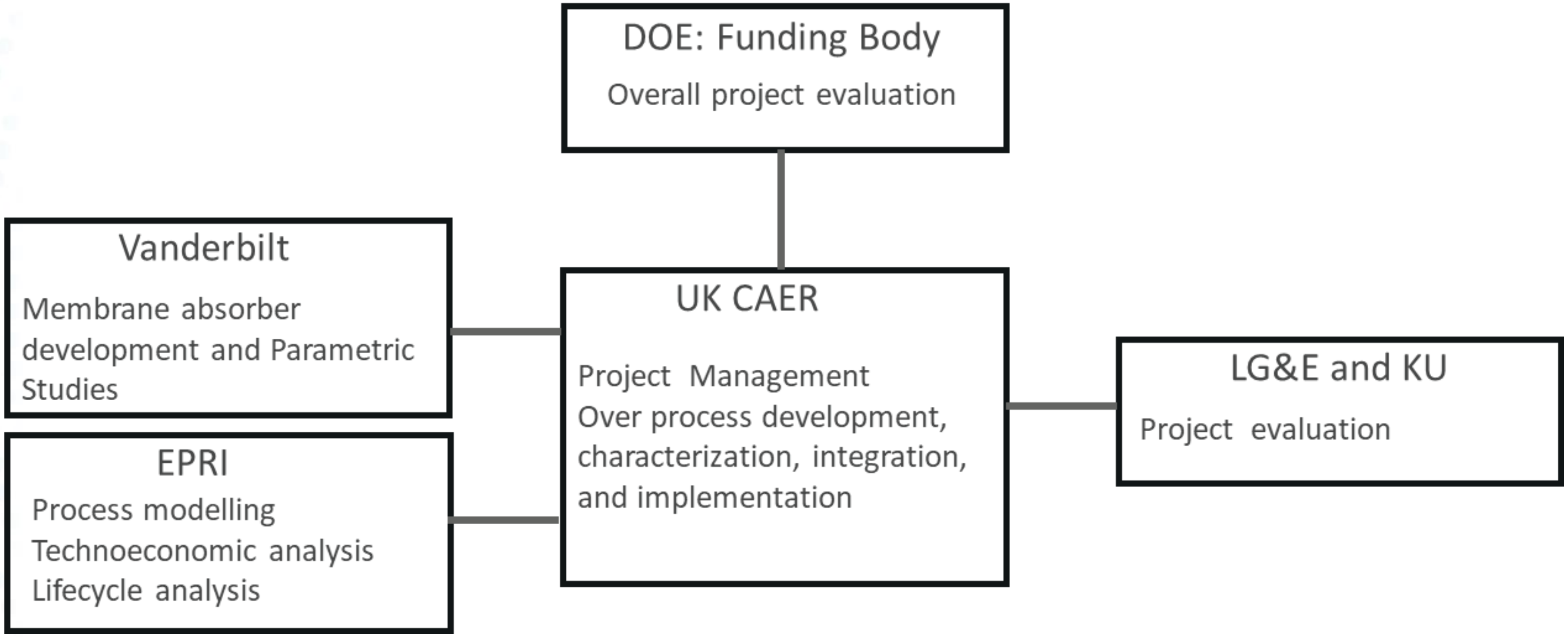
- **Vanderbilt**: Shihong Lin

Appendix - Organization Chart



Appendix - Gantt Chart

Completion

#	TASK DESCRIPTION	Start Date	End Date
1	**Project Management and Planning**	10/1/2021	9/30/2023
BP1	**Budget Period 1**	10/1/2021	9/30/2022
1.1	1A. Update Project Management Plan	10/1/2021	10/31/2021
1.1	1B. Kickoff Meeting	10/1/2021	12/30/2021
1.2	2A. Initial Technology Maturation Plan	10/1/2021	12/30/2021
1.2	2B. Final Technology Maturation Plan	8/1/2023	9/30/2023
2	**Membrane Contactor Design and Fabrication**	10/1/2021	5/31/2022
2.1	Design Membrane Absorber	10/1/2021	11/30/2021
2.2	Membrane Fabrication	12/1/2021	2/28/2022
2.3	Modulation	3/1/2022	5/31/2022
3	**Electrochemical Regenerator Construction**	10/1/2021	6/1/2022
3.1	Catalytic Electrode Design and Fabrication	10/1/2021	1/31/2022
3.2	Flow Channel Design	2/1/2022	6/1/2022
4	**System Integration**	4/1/2022	9/30/2022
4.1	Process Control and Monitoring and P&ID Development	4/1/2022	6/30/2022
4.2	Procurement and Balance of Plant	6/1/2022	7/31/2022
4.2	Startup and Commisioning	7/15/2022	9/30/2022
BP2	**Budget Period 2**	10/1/2022	9/30/2023
5	**Parametric Study Through Longterm Operation**	10/1/2022	7/31/2023
5.1	Electrochemical Regenerator Cell Voltage Reduction	10/1/2022	3/1/2023
5.2	Proton Transport Mitigation Through Membrane	10/1/2022	3/1/2023
5.3	Effectiveness of CO_2 Capture	10/1/2022	3/1/2023
5.4	Longterm Operation to Characterize Material Degradation	1/1/2023	7/31/2023
6	**Technoeconomic Analysis**	4/1/2023	9/30/2023
6.1	Process Modeling	4/1/2023	5/1/2023
6.2	Technoeconomic Analysis	5/1/2023	9/30/2023
7	**Life Cycle Analysis**	4/1/2023	9/30/2023
7.1	Life Cycle Analysis	4/1/2023	9/30/2023