Energy-Efficient Direct Air Capture System for High-Purity CO_2 Separation

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Project Overview

1) Funding: DOE share $1,499,999 and cost share $393,650

2) Overall Project Performance Dates: 10/1/2021 – 12/31/2024

3) Project Participants: University of Cincinnati (UC), BASF, Trimeric

4) Overall Project Objectives: demonstrate Recipient's DAC sorbent technology to capture CO_2 from ambient air and separate it at high purity.

Technology Background

- Modified amine-based sorbent technology with low desorption energy requirement and resistance to thermal and oxidative degradations
- Sorbents with high capacities, fast kinetics, small mass-transfer resistances, and low desorption energy requirement
- State-of-the-art coating technology for scale-up from powdered form to monolithic form
- High throughput of air flow with minimum pressure drop through sorbent-coated monolith in passive air contactor
- Passive air contactor without energy requirement during adsorption

Technical Approach/Project Scope

- Task 3: Manufacture CO_2 sorbent (UC)
  - ✓ Manufacture CO_2 sorbent
  - ✓ Evaluate long-term lab-scale performance

- Task 4: Manufacture sorbent-washcoated monolith structure (UC + BASF)
  - ✓ Develop sorbent-washcoated monolith
  - ✓ Determine cell size and length

- Task 6: Evaluate performances of sorbent-washcoated monolith in air contactor system (UC)
  - ✓ Measure temperature, humidity, velocity, and CO_2 concentration
  - ✓ Evaluate performances

- Task 7: TEA and LCA (UC + Trimeric)
  - ✓ Development of TCM and LCI
  - ✓ Assessments of DAC technology

Technical Approach/Project Scope

Major milestones

Task	Milestone Title & Description	Planned Completion Date
3	Manufacture of 10 kg of CO_2 sorbent	PSD+23 months
4	Manufacture of two sorbent-washcoated monolith prototypes	PSD+24 months
6	CO_2 capture efficiency, energy requirements, and overall volumetric CO_2 productivity	PSD+39 months

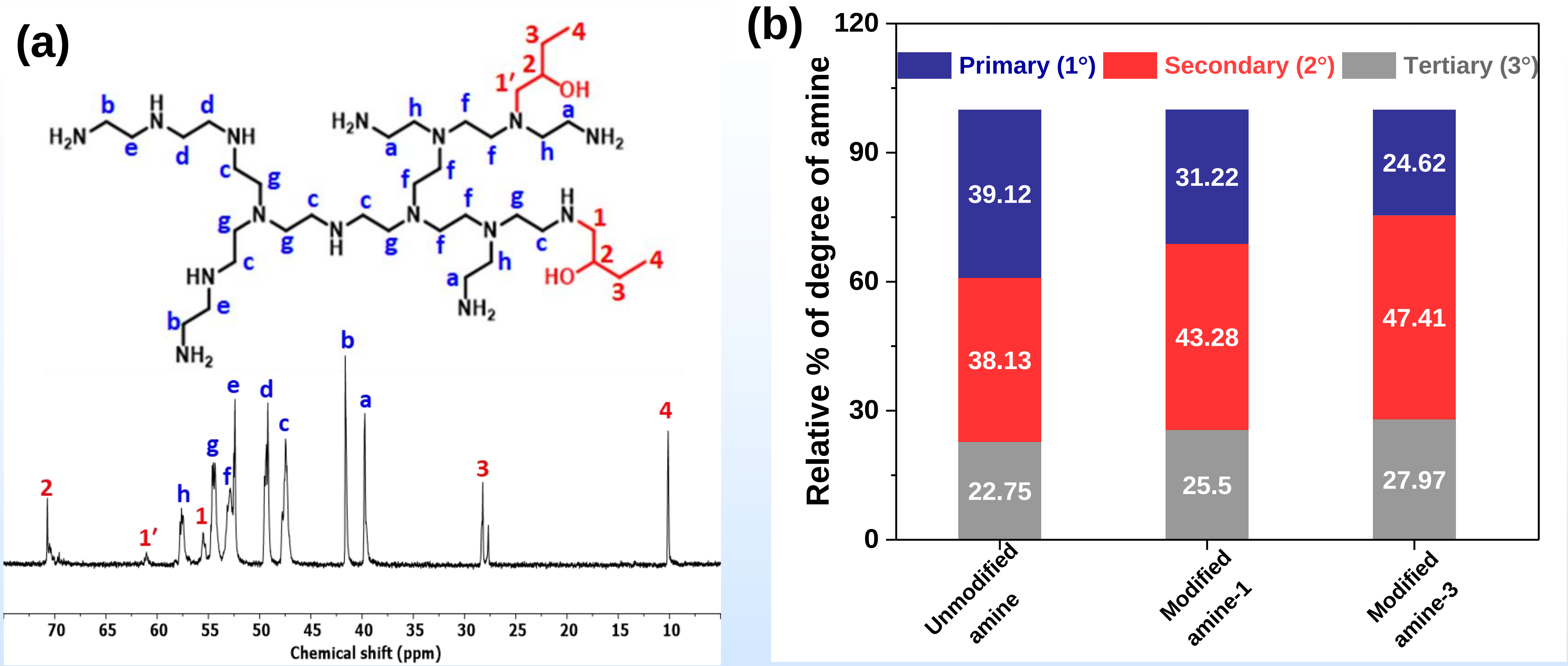
Major Success criteria

- 70% average CO_2 capture efficiency in passive air contactor with monolith with pressure drop of <100 Pa and <75 kJ/mol CO_2
- Overall volumetric productivity of ~2 (gmol CO_2/(hr x V(l))

Project Risks and Mitigation Strategies

- Low performance of sorbent-coated monolith: BASF will attempt many different coating formulations
- Low DAC system performance: parametric testing will be carried out to investigate the effects of parameters (i.e., operating conditions, materials, etc.)

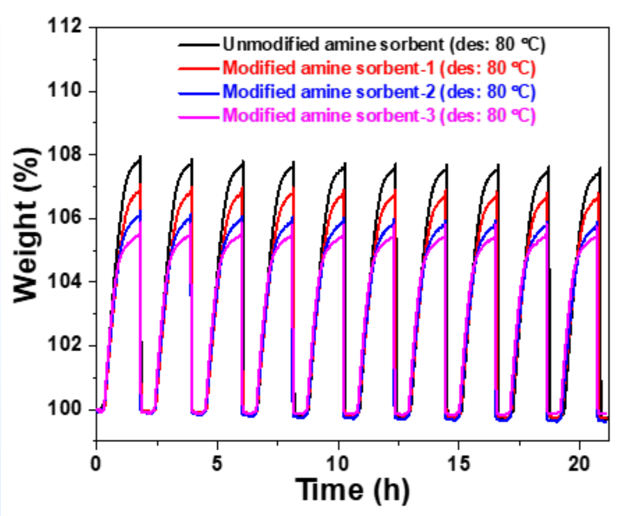
Modified PEI Sorbent



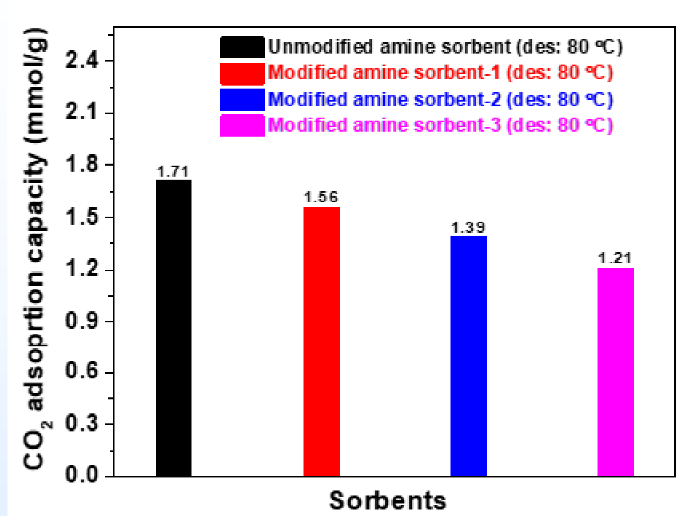
^{13}C NMR spectrum for (a) modified amine and (b) distribution of amine states

1) Increased resistance to thermal and oxidative degradations by lowering the basicity and increasing steric hindrance.
2) Reduced bonding strengths leading to low heat of desorption with decreased capacity
3) UC filed U.S. patent/PCT for 10 modifications.

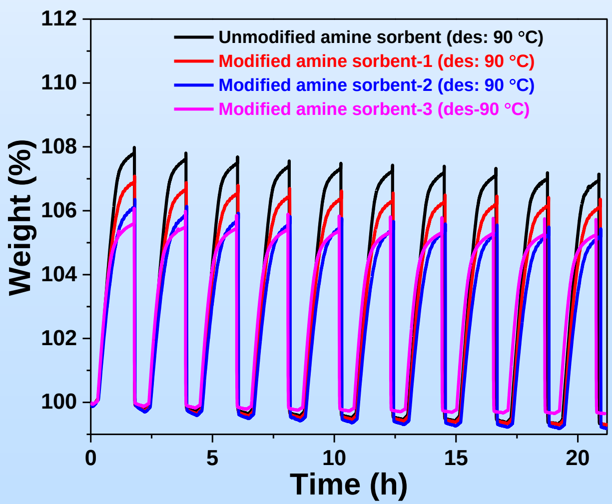
Performance Evaluations in TGA: 10 cycles



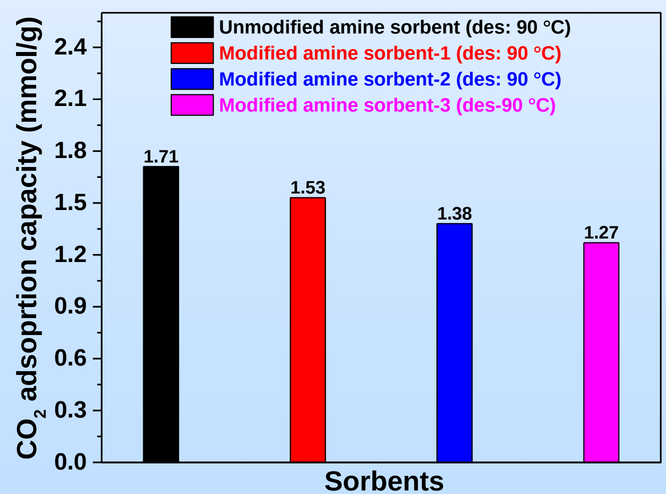
N_2 desorption at **80 °C**



Capacity decreases with an increase in level of modification.

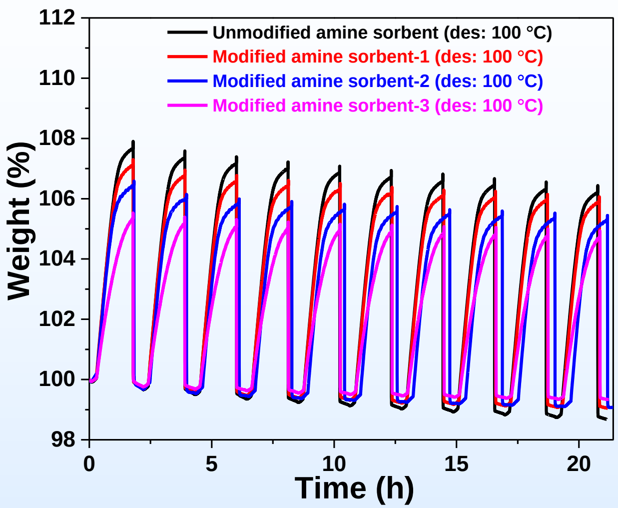


N_2 desorption at **90 °C**

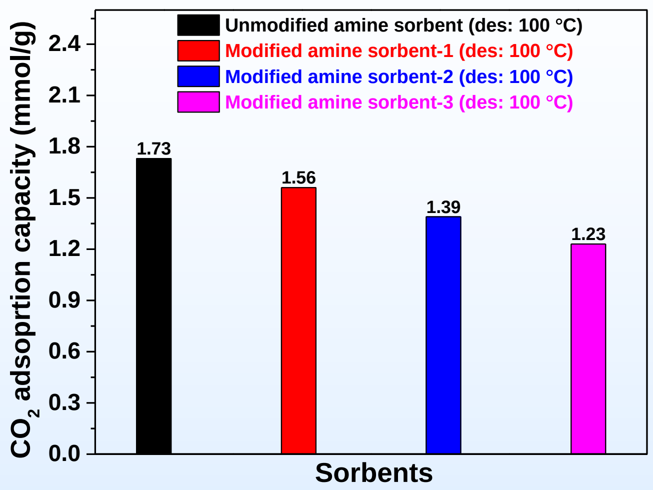


Adsorption: 400 ppm CO_2 in dry air at 30 °C and **Desorption: N_2 at (a) 80 and (b) 90 °C**

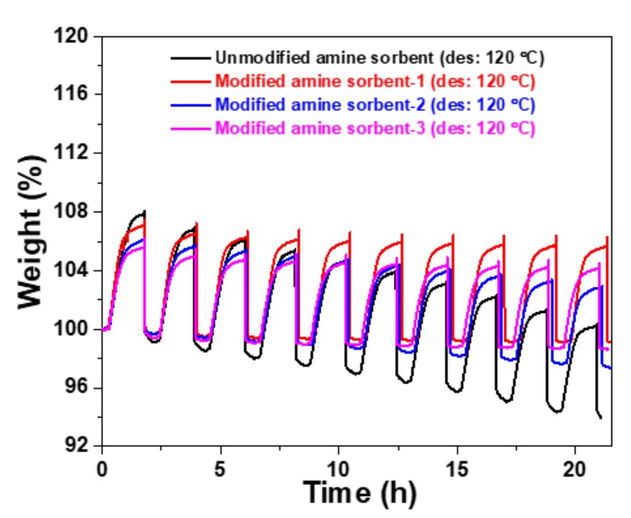
Performance Evaluations in TGA: 10 cycles



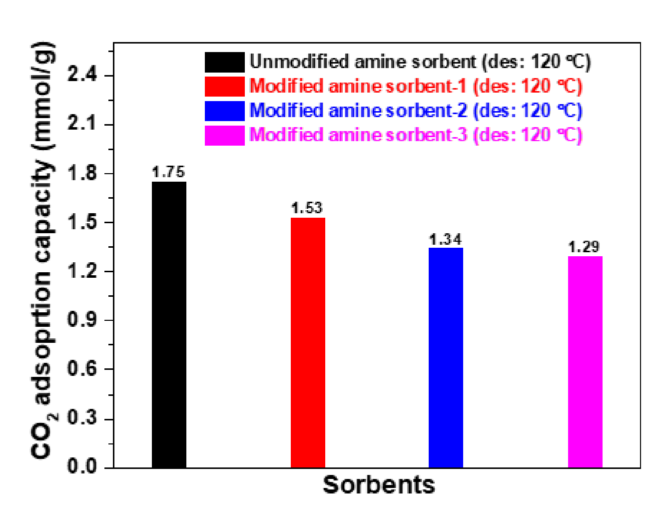
N_2 desorption at **100 °C**



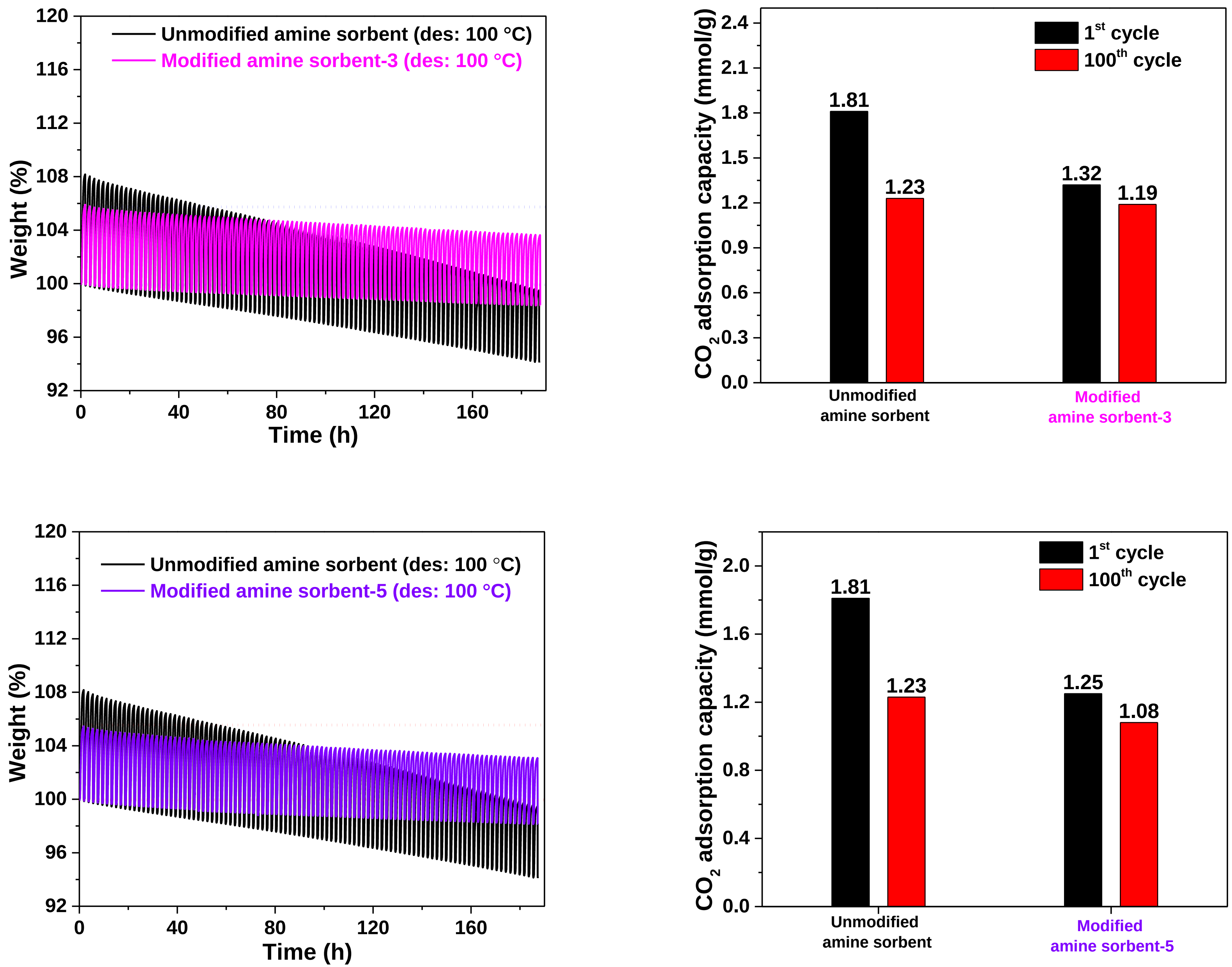
Capacity decreases with an increase in level of modification.



N_2 desorption at **120 °C**

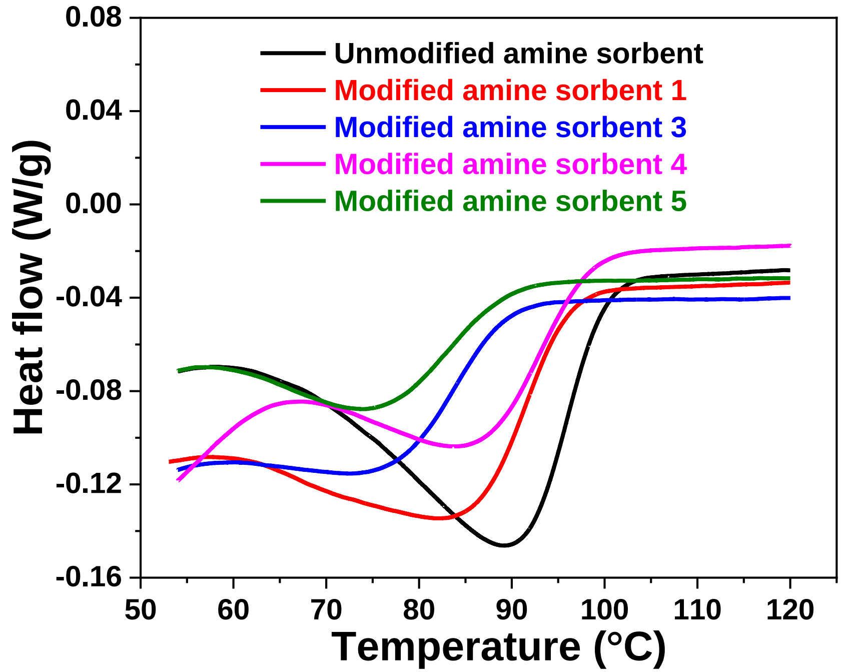


Adsorption: 400 ppm CO_2 in dry air at 30 °C and **Desorption: N_2 at (a) 100 and (b) 120 °C**



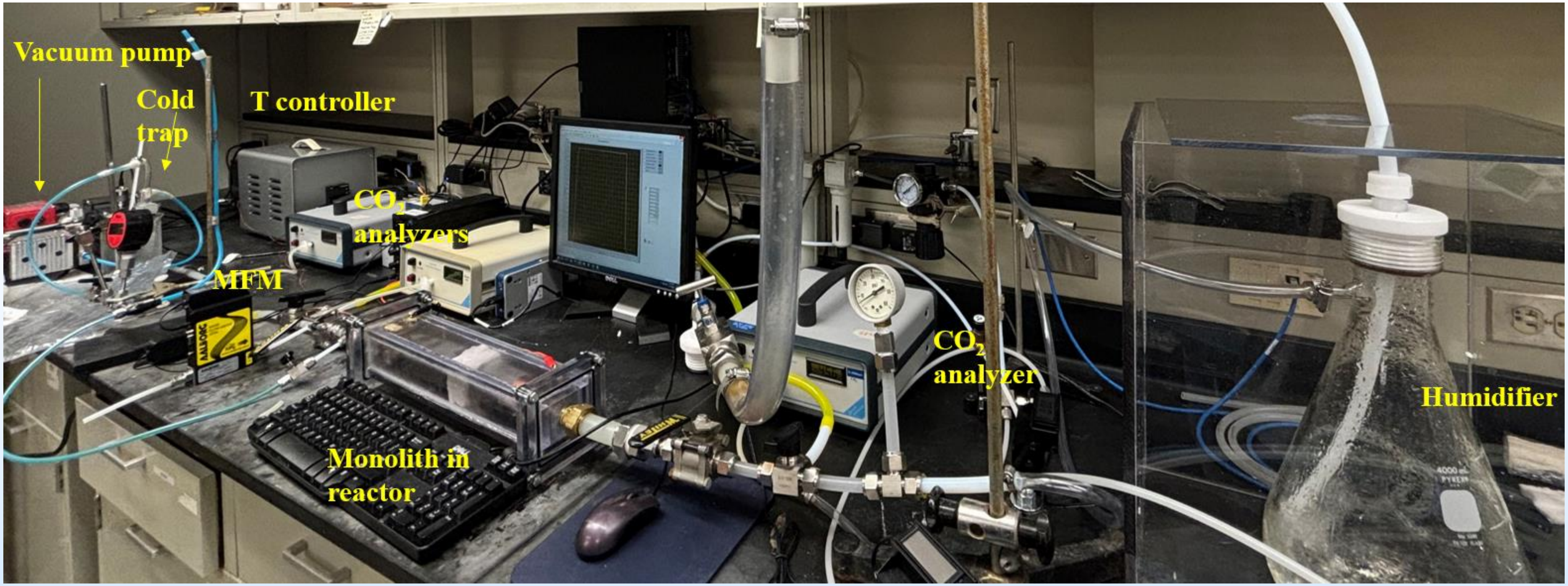
Adsorption: 400 ppm CO_2 in Dry Air at 30 °C and Desorption: N_2 at 100 °C

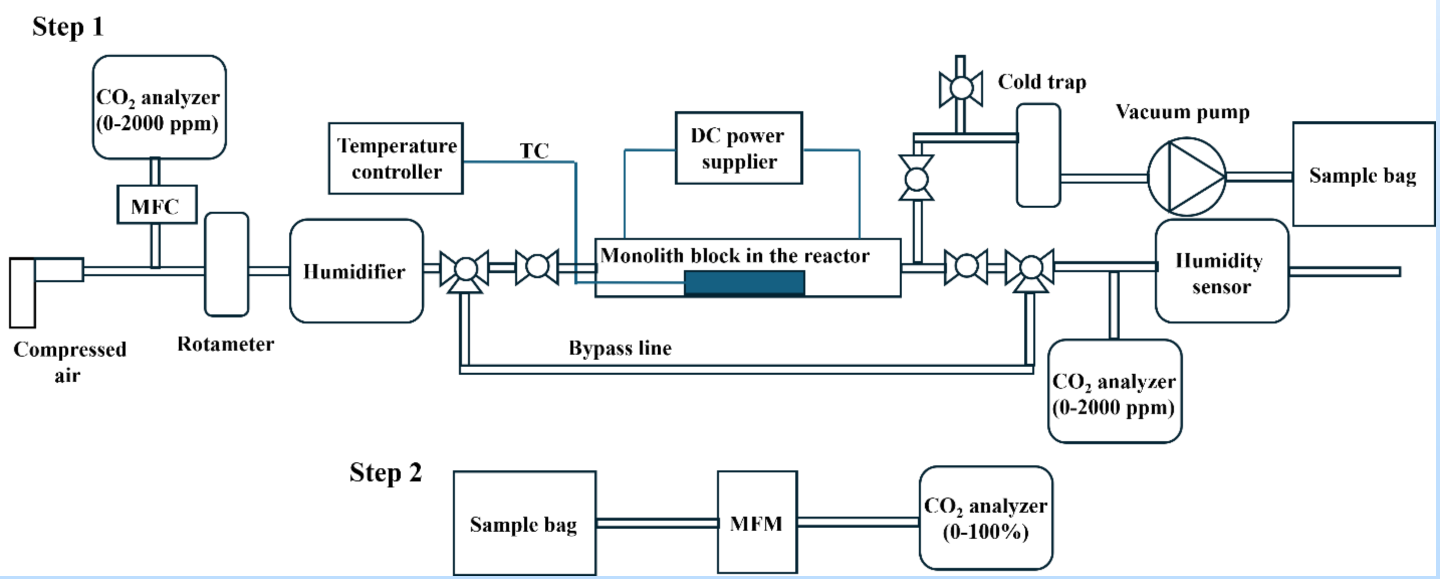
Heat of Desorption for Powdered Sorbents



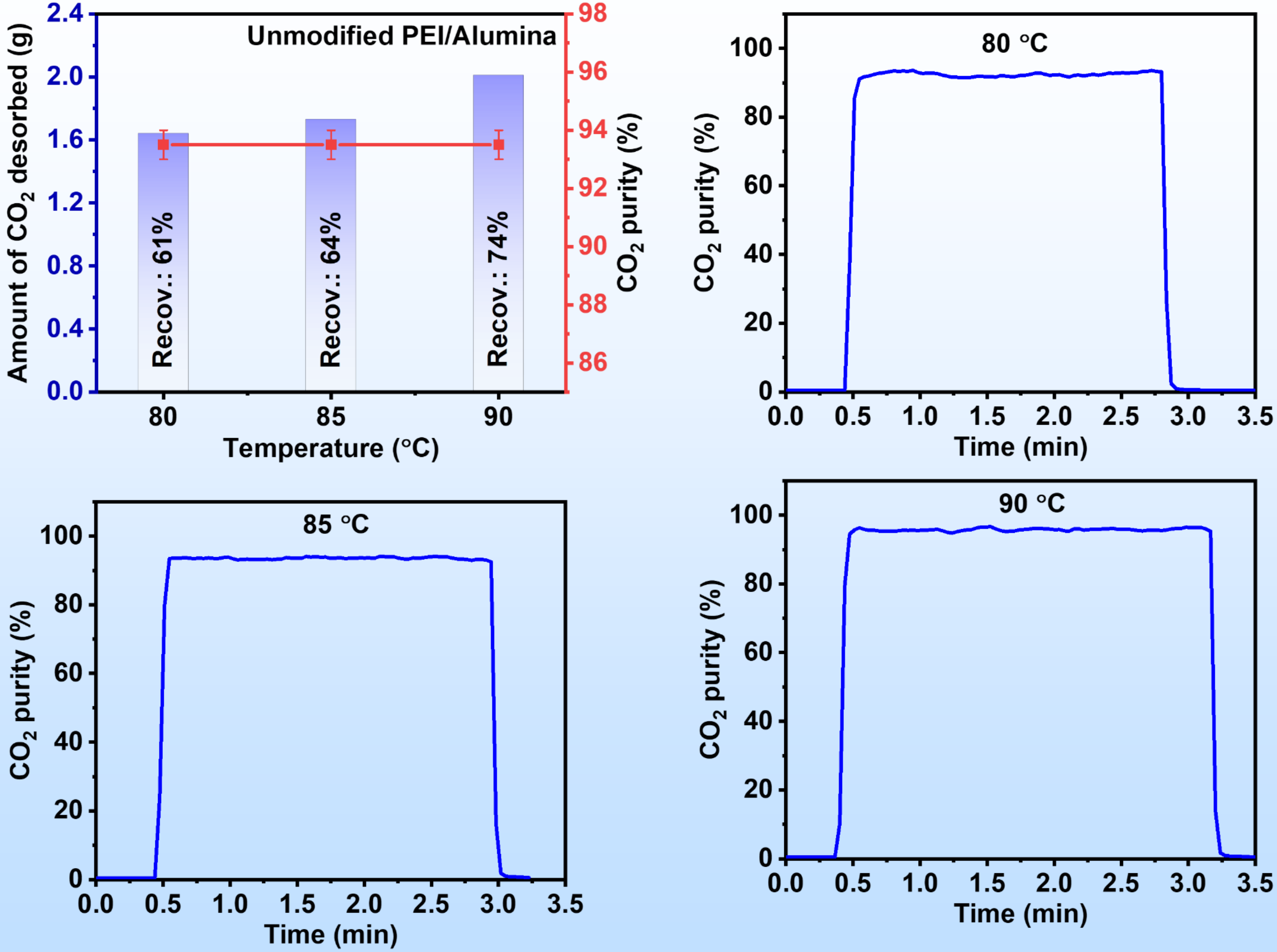
Sorbents	Heat of desorption (kJ/gmol CO_2)	Energy savings relative to unmodified amine sorbent (%)*
Unmodified amine sorbent	**73.74**	**0**
Modified amine sorbent 1	56.87	-22.9
Modified amine sorbent 3	**34.77**	**-52.8**
Modified amine sorbent 4	**35.83**	**-51.4**
Modified amine sorbent 5	**31.82**	**-56.8**

Performance Evaluations of Small Block (3 cm×3 cm×15 cm)



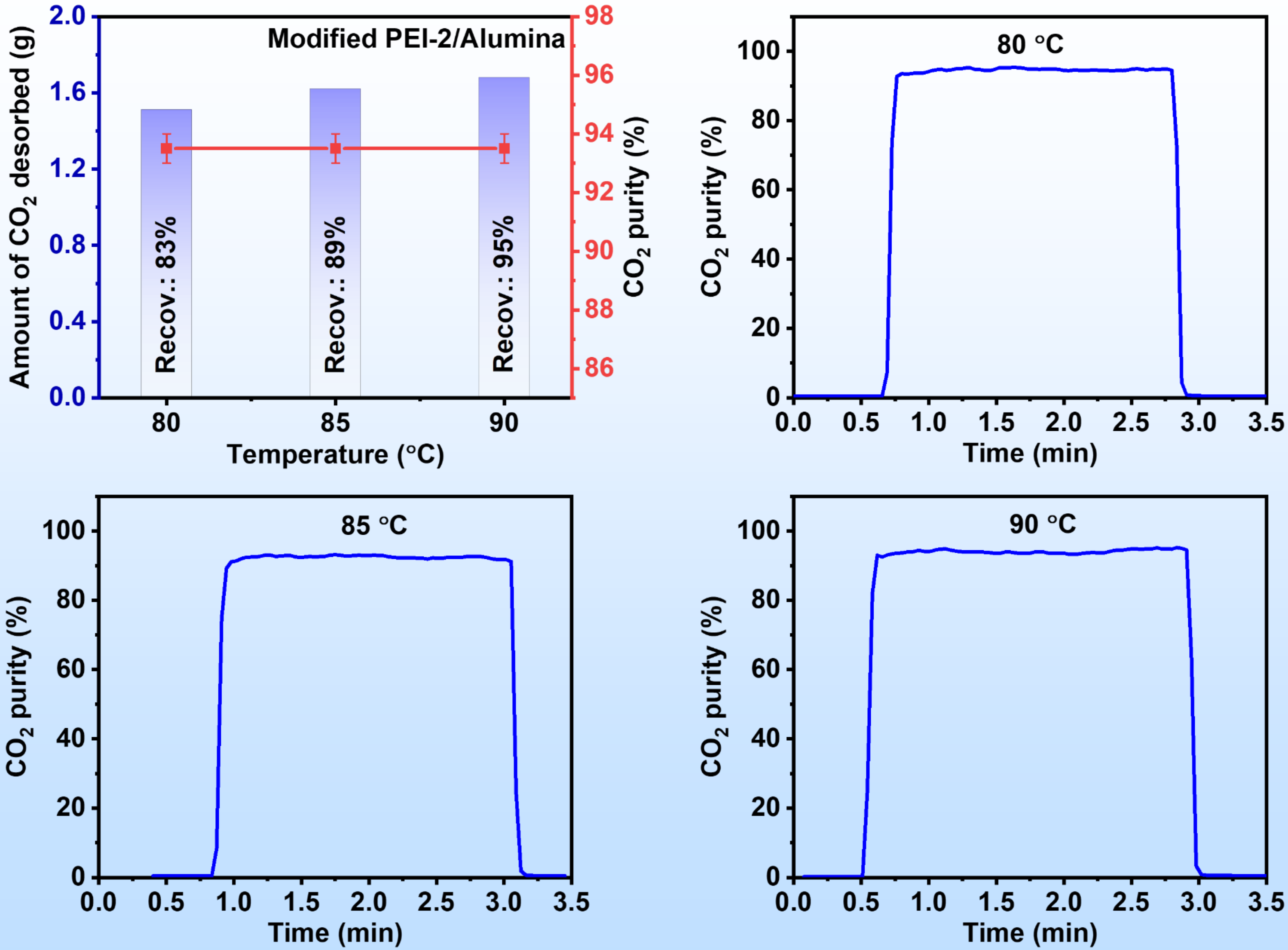


CO_2 Desorption by JH between 80-90 °C from Unmodified PEI/Alumina Block
(Block was saturated under 175 LPM air, SV: 77,778 hr^{-1}, 50% RH, 20 °C)



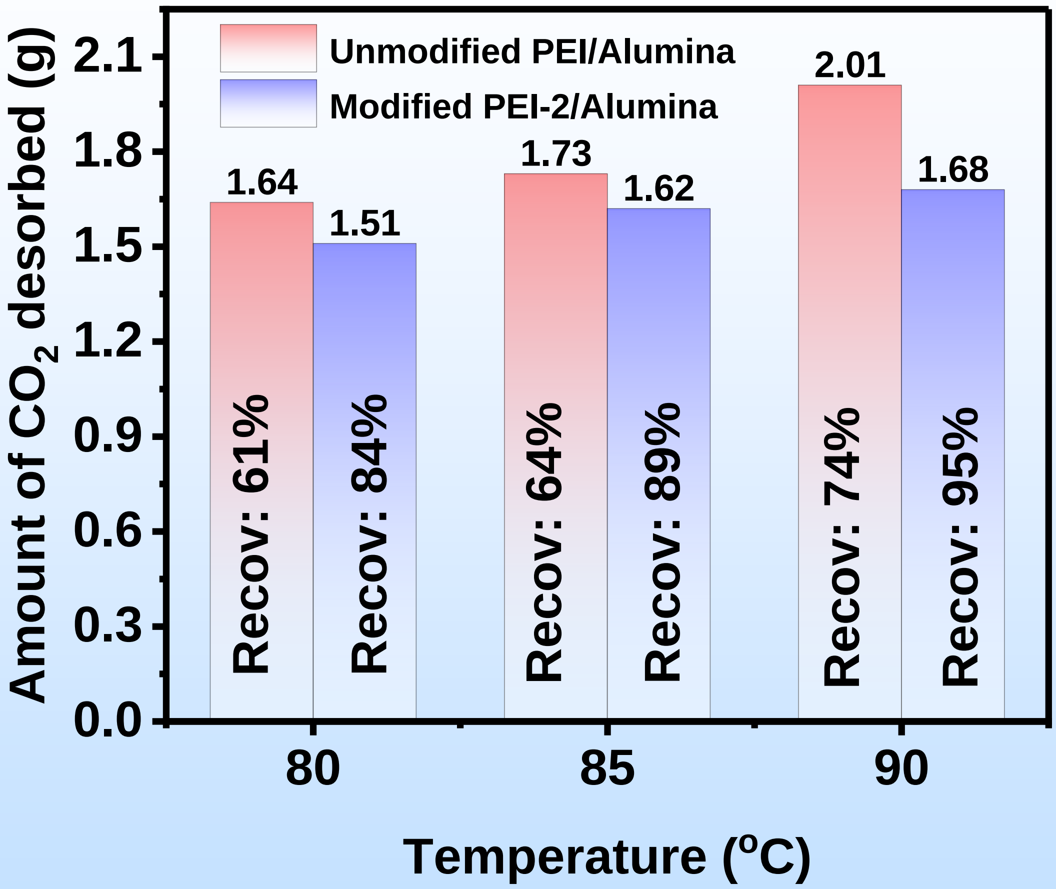
- Vacuum pressure of -28” Hg (full vacuum: -29.9” Hg)
- Initial air present inside the reactor: ~6.3%

CO_2 desorption by JH between 80-90 °C form Modified PEI-2/Alumina Block (Block was saturated under 175 LPM air, SV: 77,778 hr^{-1}, 50% RH, 20 °C)



- Vacuum pressure of -28” Hg (full vacuum: -29.9” Hg)
- Initial air present inside the reactor: ~6.3%

Working Capacity and CO_2 Recovery by Joule Heating between 80-90 °C



- Vacuum pressure of -28” Hg (full vacuum: -29.9” Hg)
- Initial air present inside the reactor: ~6.3%
- RH: ~50%; CO_2 purity: ~93-94%; SV: 77,778 hr^{-1}

CO_2 desorption between 80-90 °C

Modified PEI-2/Alumina block

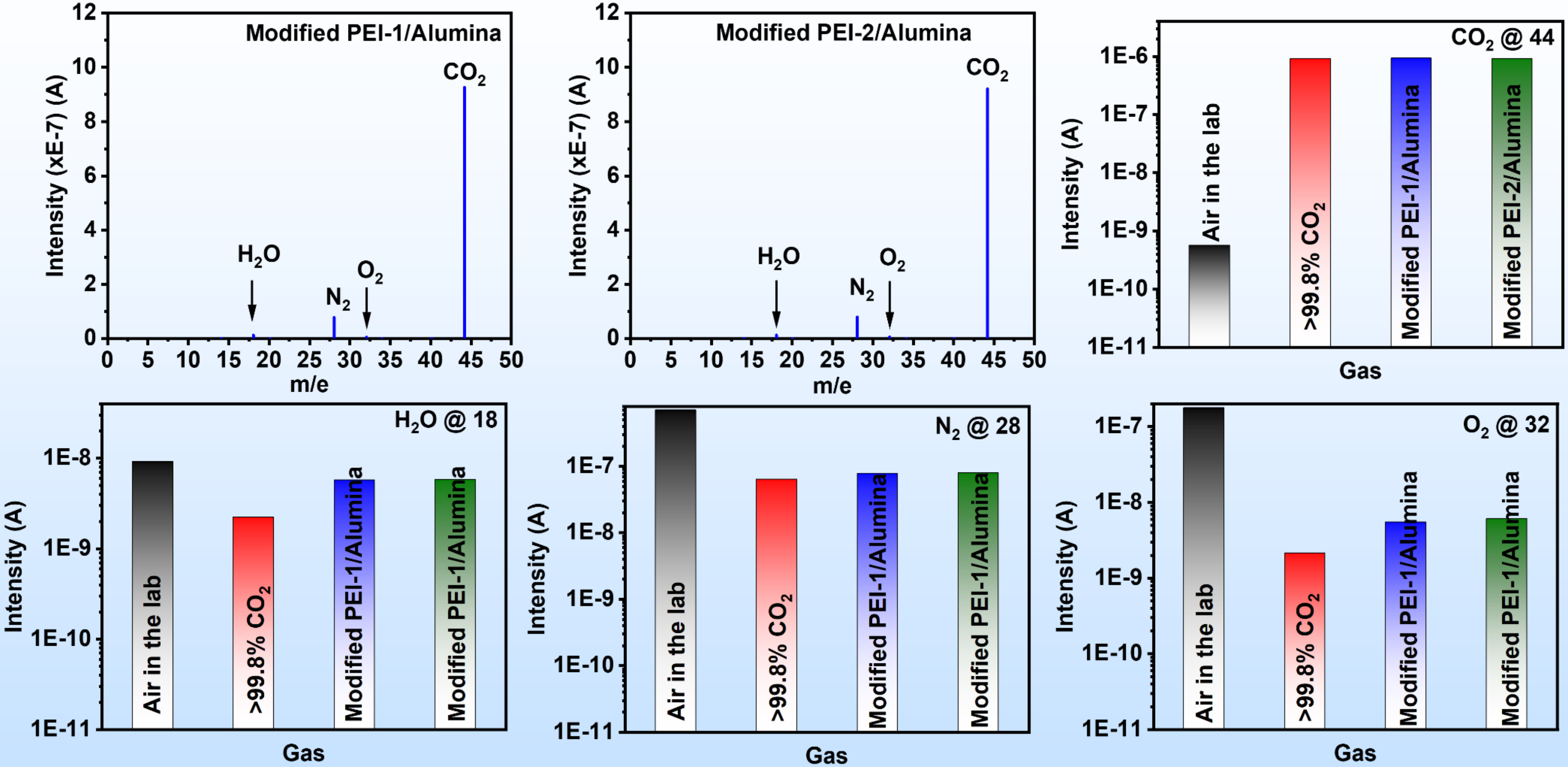
Des T (°C)	Adsorption capacity		Working capacity by JH (mmol)	Working capacity (mmol CO_2/ g sorbent)	Residual CO_2 desorbed by N_2 (mmol)	Total CO_2 desorbed (mmol)	CO_2 recovery from JH (%)	CO_2 purity (%)
	mmol CO_2/ g sorbent	mmol CO_2/ monolith						
80	1.19	31.96	**34.42** (1.51 g)	1.28	**6.76**	**41.18**	**84**	**94**
85	1.21	32.45	**36.76** (1.62 g)	1.37	**4.52**	**41.28**	**89**	**93**
90	1.17	31.35	**39.11** (1.68 g)	1.42	**1.95**	**41.06**	**95**	**94**

PEI/Alumina block

Des T (°C)	Adsorption capacity		Working capacity by JH (mmol)	Working capacity (mmol CO_2/ g sorbent)	Residual CO_2 desorbed by N_2 (mmol)	Total CO_2 desorbed (mmol)	CO_2 recovery from JH (%)	CO_2 purity (%)
	mmol CO_2/ g sorbent	mmol CO_2/ monolith						
80	1.68	46.66	**37.21** (1.64 g)	1.34	**23.76**	**60.97**	**61**	**93**
85	1.67	46.31	**39.24** (1.73 g)	1.41	**21.78**	**61.02**	**64**	**93**
90	1.66	46.07	**45.26** (2.01 g)	1.69	**15.09**	**60.85**	**74**	**94**

- **PEI block has ~30% higher saturation capacity than modified PEI-2 block.**
- **Adsorption T:** 20 °C
- **RH:** 50%
- **SV:** 77,778 hr^{-1}

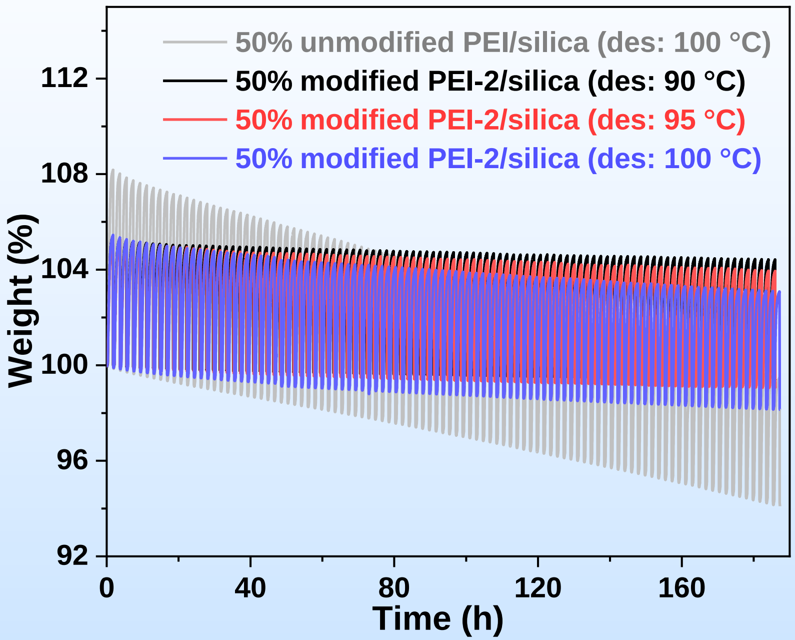
MS Analysis of Gas Desorbed by Joule Heating at 90 °C

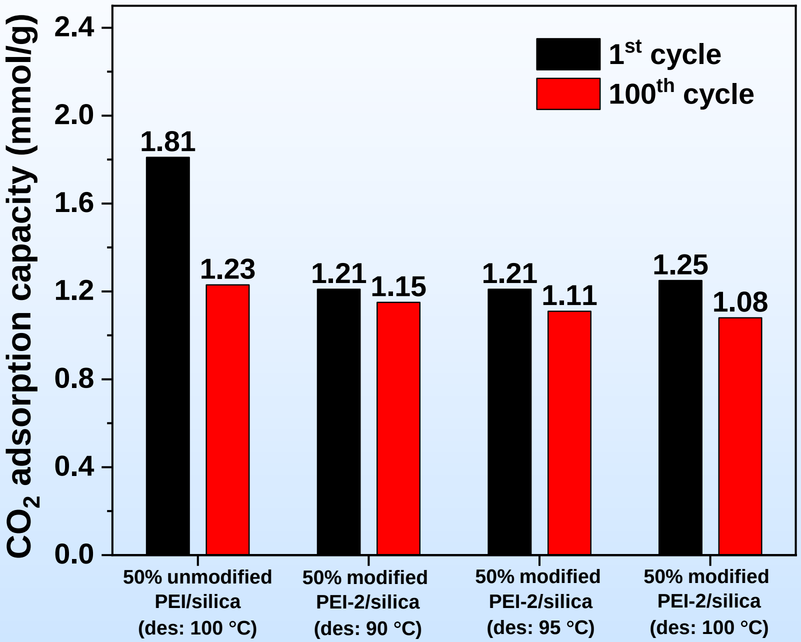


Tech-grade CO_2 gas compositions:

- CO_2: >99.8%
- O_2: < 20 ppm
- Moisture: < 20 ppm

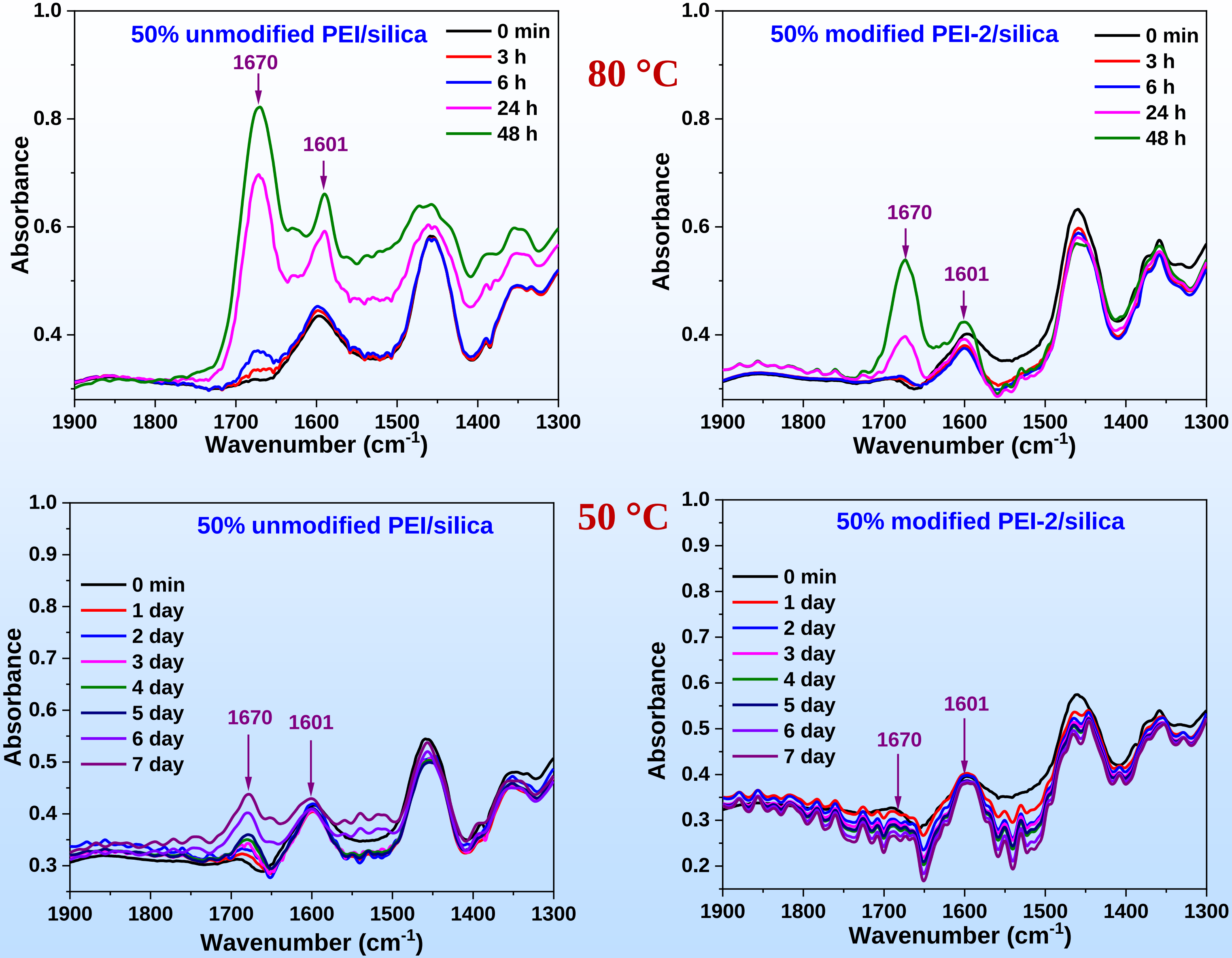
Thermal Stability of Modified PEI-2 Sorbent between 90-100 °C over 100 Cycles





- Thermal degradation at ≤90 °C seems to be negligible for modified PEI-2 sorbent.
- Adsorption under 400 ppm CO_2 in dry air at 30 °C
- Desorption under N_2

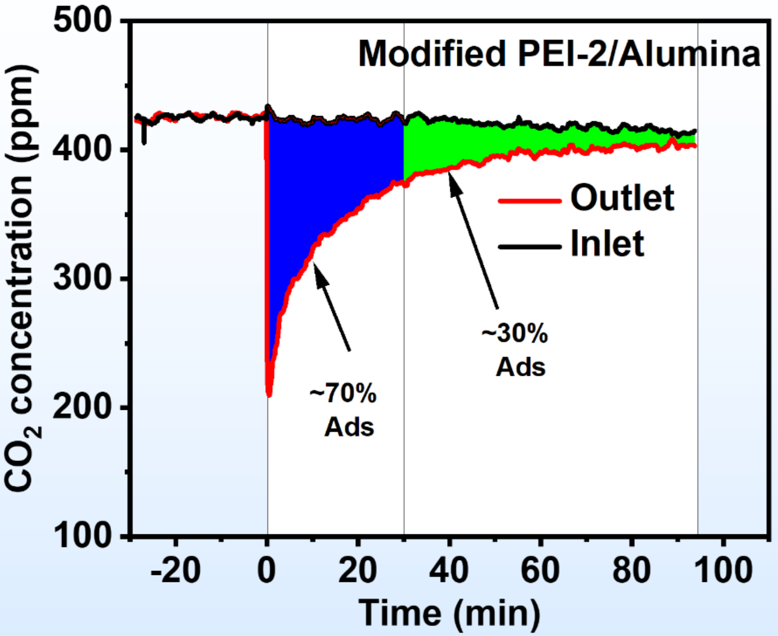
Thermal and Oxidative Degradations in Air



If sorbent is exposed to air at 50 °C during switch to adsorption for 5 min/cycle, no significant degradations are expected over 2,016 cycles.

- 1,670 cm^{-1}: either imine (C=N) or carbonyl (C=O) group.
- 1,601 cm^{-1}: NH_2 deformation of primary amines.

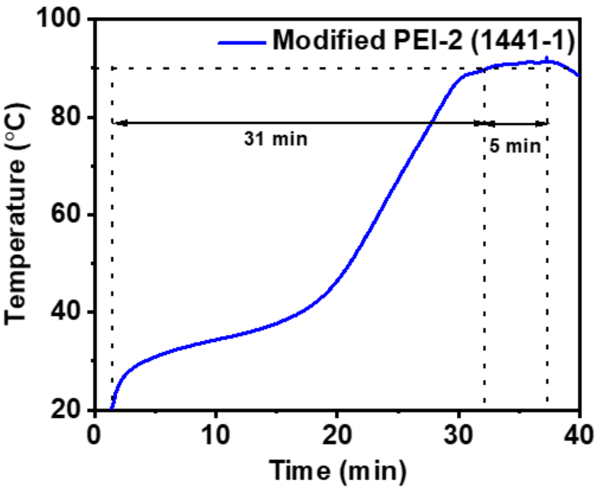
Adsorption and Desorption Profiles of Small Block Testing



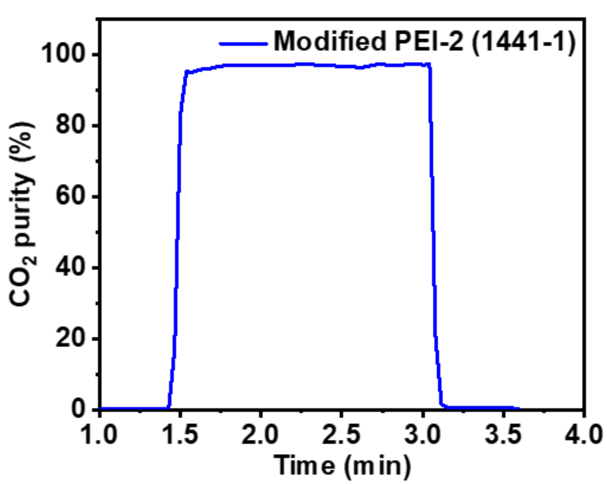
Adsorption conditions:
- ✓ **Flowrate:** 175 LPM
- ✓ **Relative humidity (RH):** ~50%
- ✓ **Temperature:** 20 °C

Desorption conditions:
- ✓ **Vacuum pressure: -28" Hg**
- ✓ **Initial air present inside reactor after pulling a vacuum: ~6%**
- ✓ **Temperature: 90 °C**
- ✓ **CO_2 recovery and purity: ~95%**

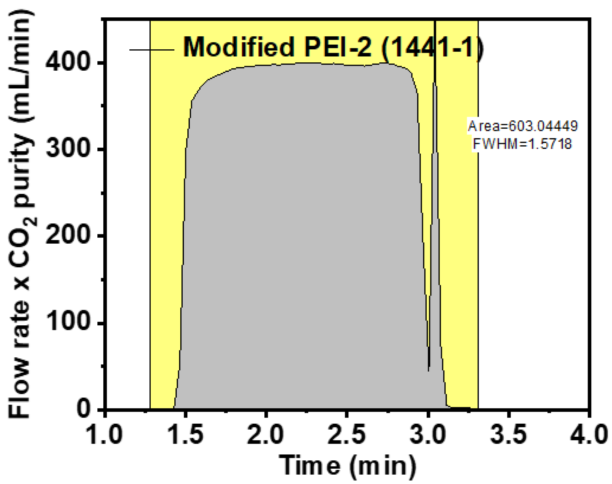
Temperature profile



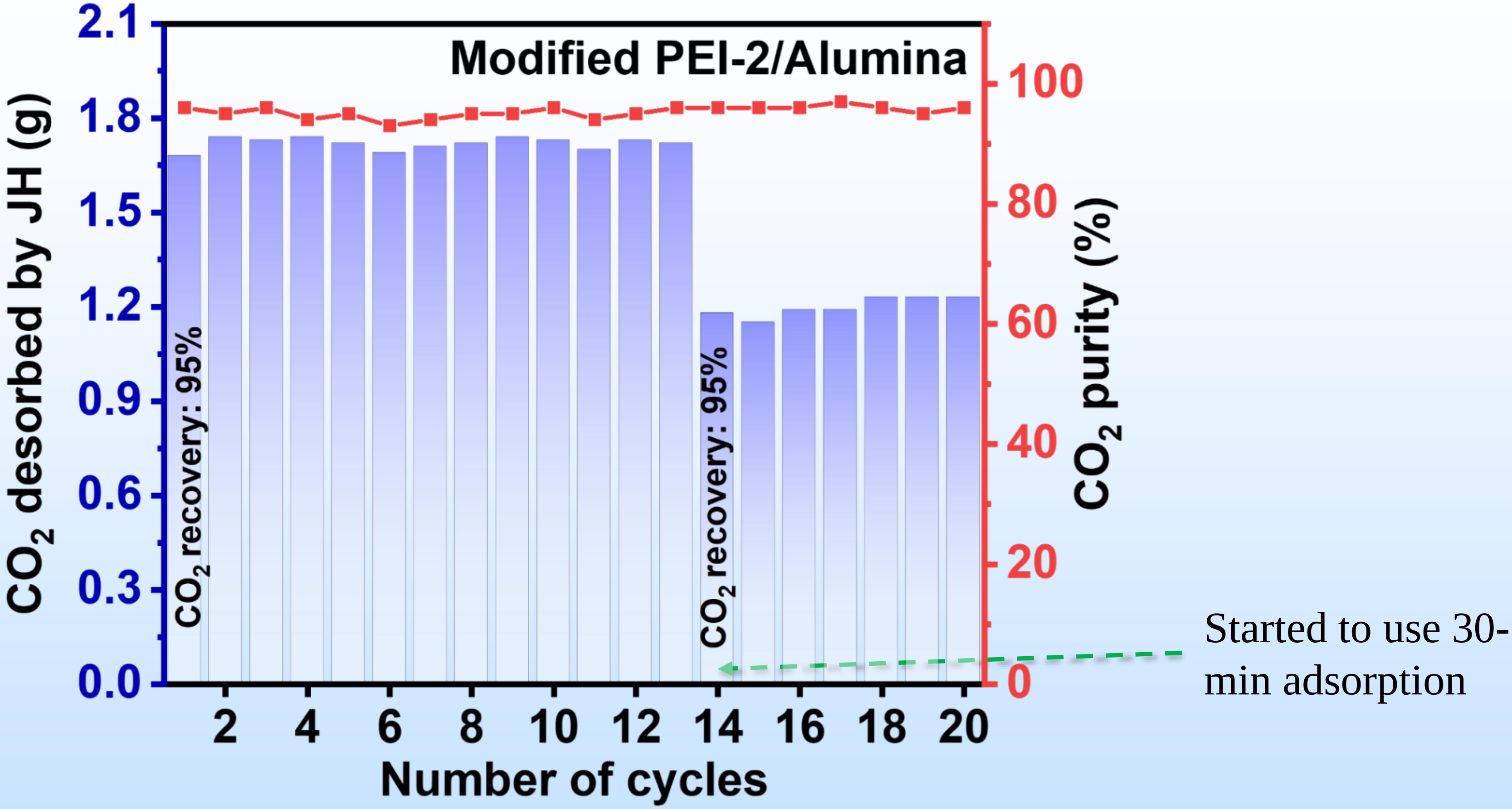
CO_2 purity profile



CO_2 volume profile

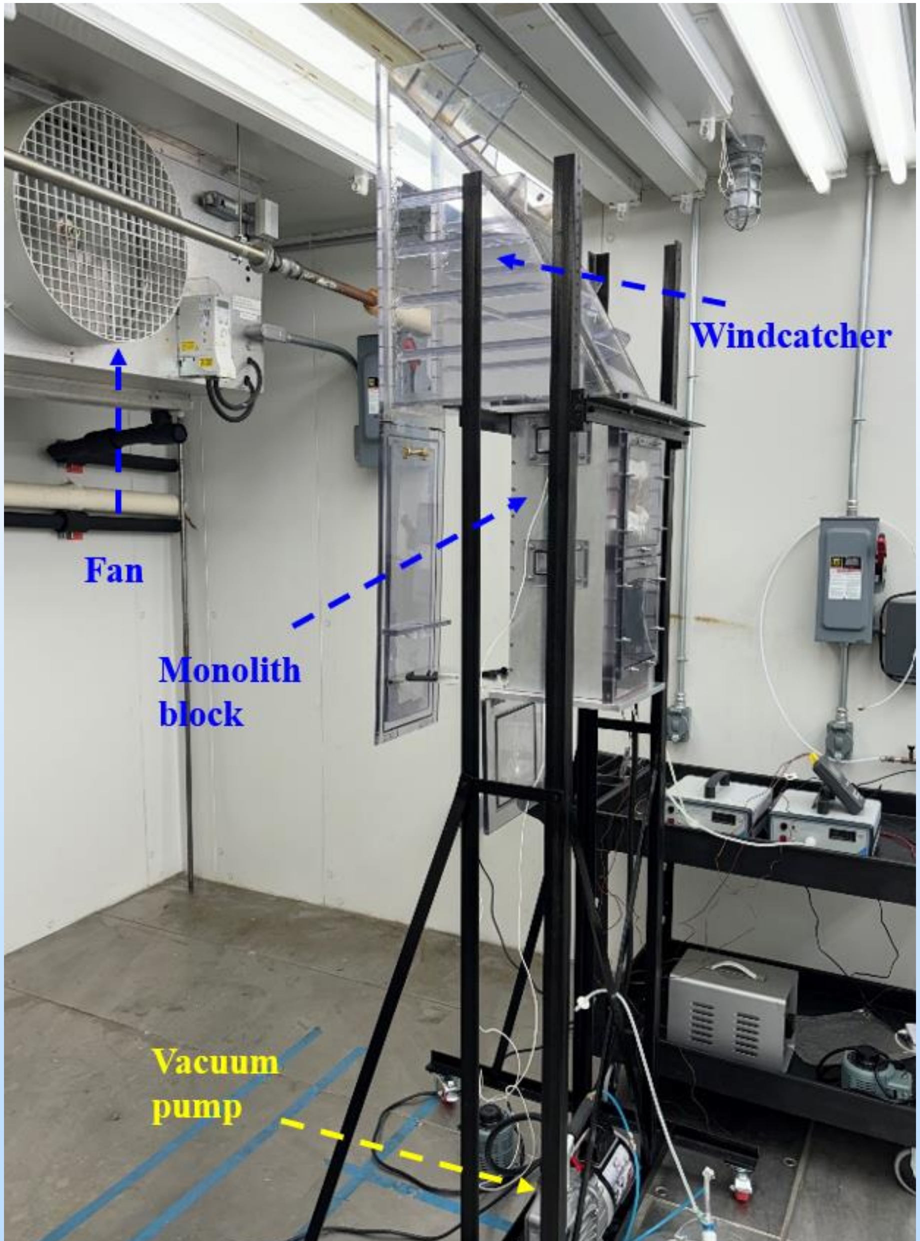


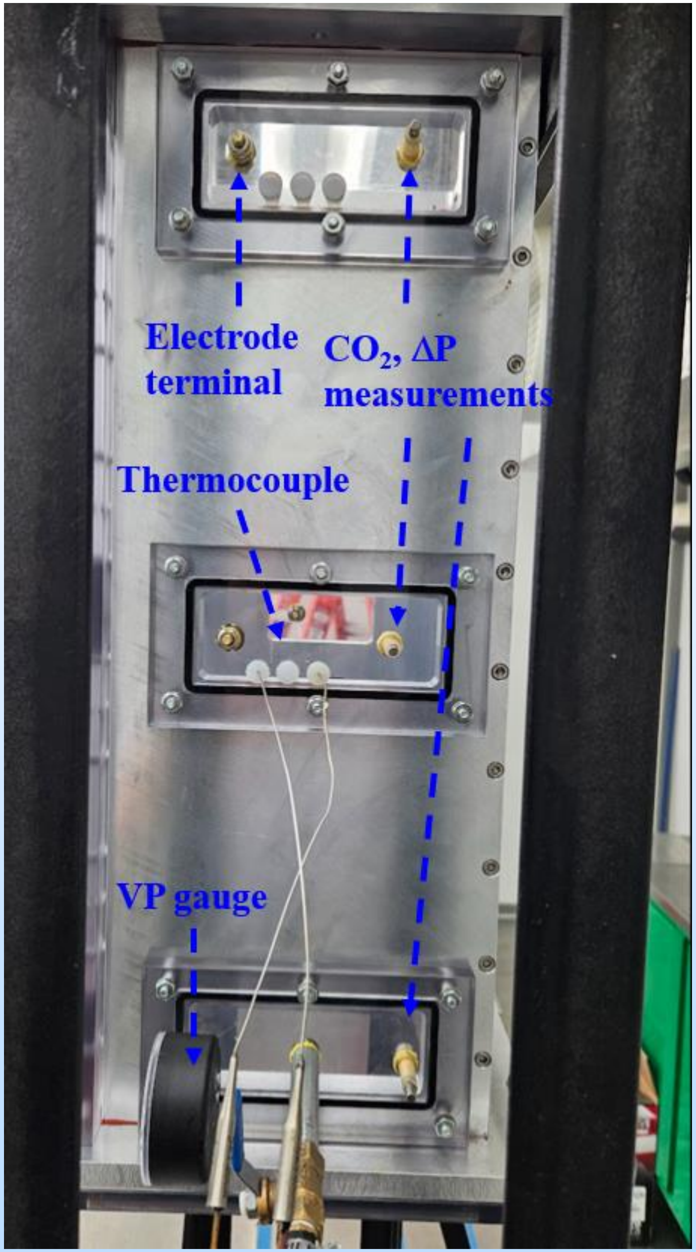
Working Capacity of Modified PEI-2 on Small Monolith



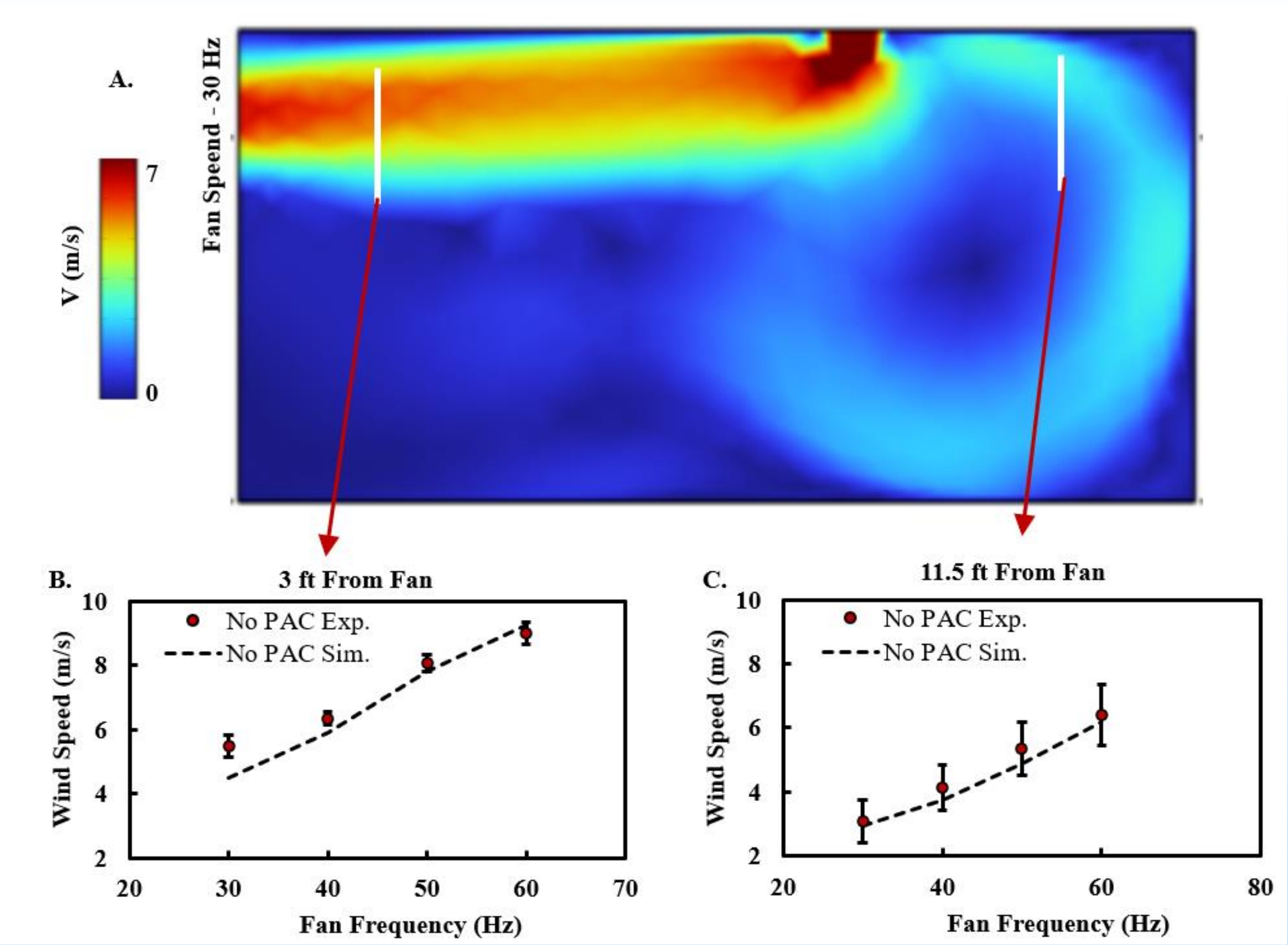
- ✓ **Goal: 100 cycles** for PEI and Modified PEI-2 blocks
- ✓ **Flowrate:**175 LPM air; **SV**: 77,778 hr^{-1}; **RH:** ~50%; **Adsorp. T:** 20 ℃; **Desorp. T:** 90 ℃; **Switch** from desorption to adsorption at 50 ℃
- ✓ **Adsorption time for Cycle 1-13:** 90 min for >~95% adsorption capacity
- ✓ **Adsorption time for Cycle 14-20:** 30 min for ~70% adsorption capacity

Evaluation of Large Monolith Block in PAC

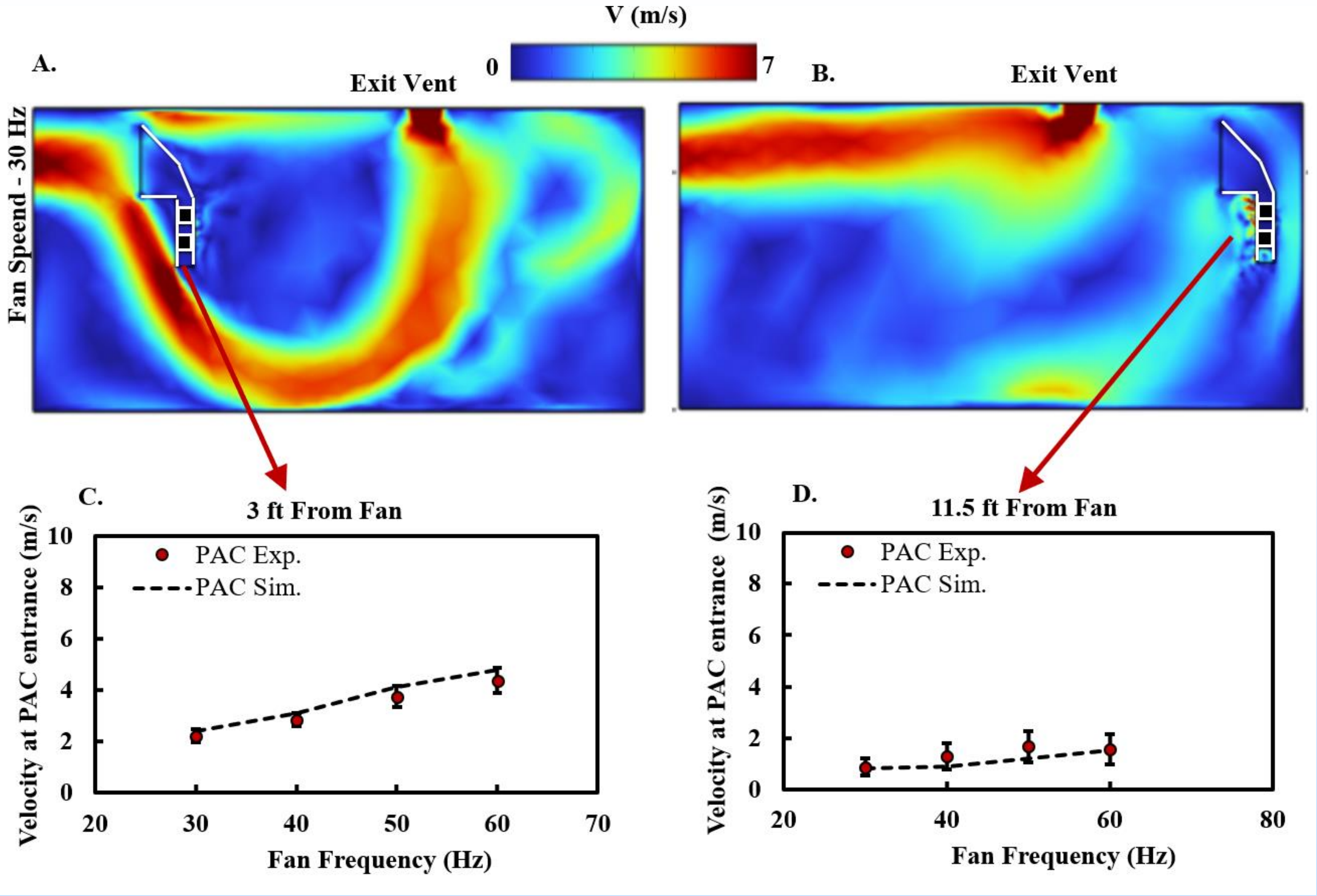




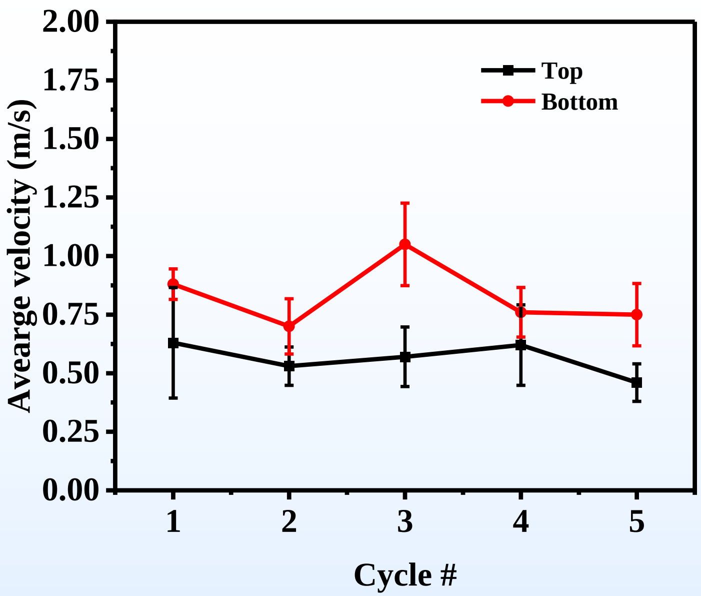
Wind Speeds without Passive Air Contactor (PAC)

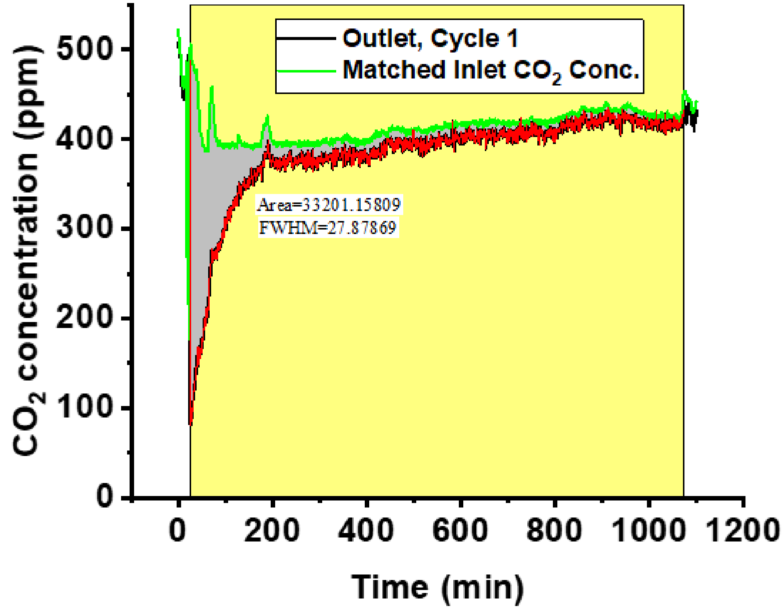


Linear Velocity inside DAC System



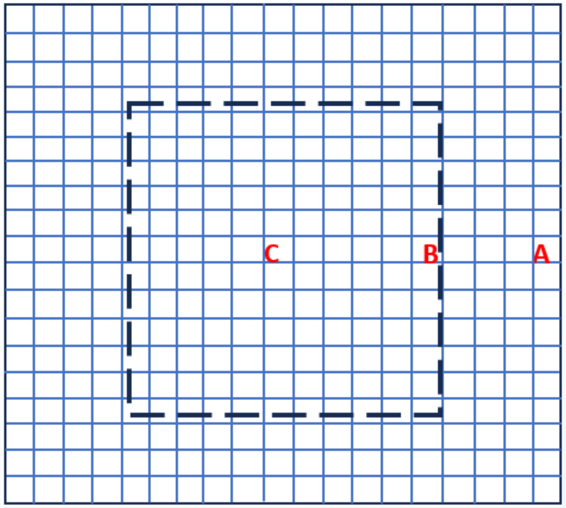
- ✓ **T: 0-40 C; RH: 20-80%**
- ✓ **Wind speeds: ~3-8 m/s**
- ✓ **LVs at monolith outlet: ~0.6-4 m/s**

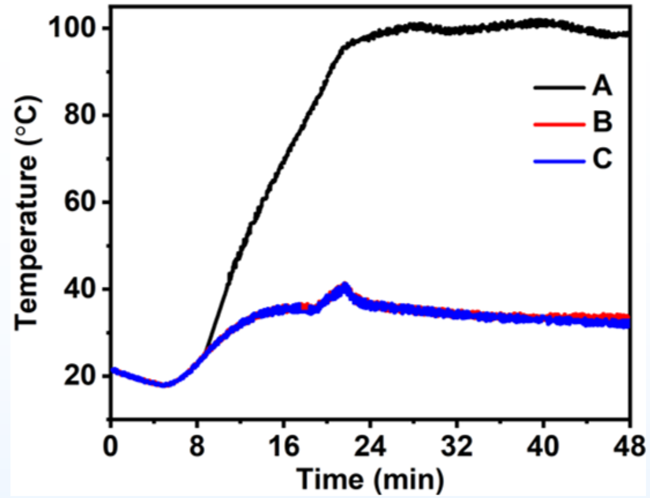




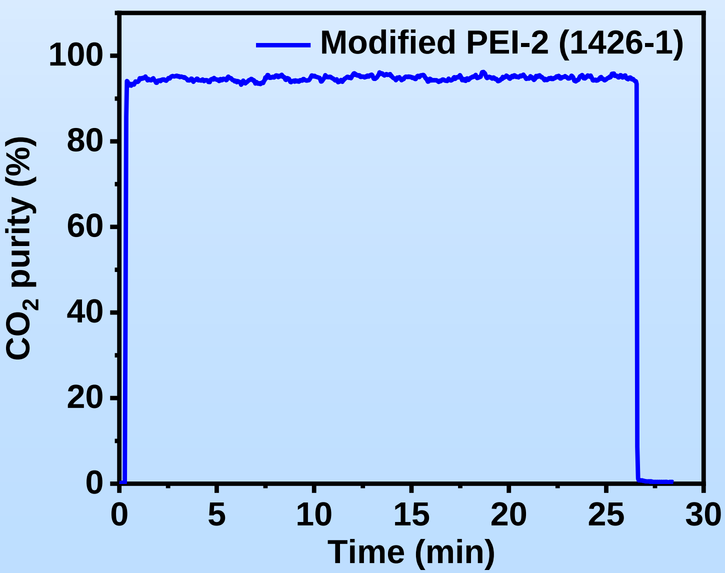
- ✓ **Single block with 64 cpsi**
- ✓ **RH:** ~50%; **Wind speed:** ~7 m/s; **Initial vacuum pressure:** -28” Hg
- ✓ **CO_2 purity**: 94 ±1%

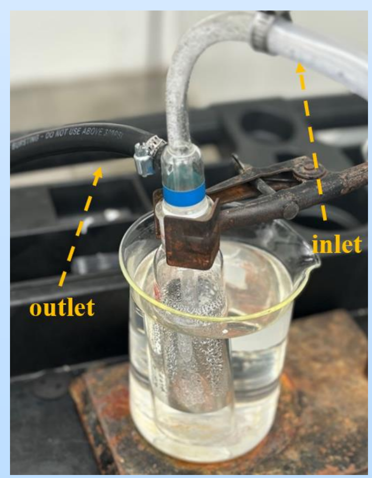
Cycle #	Avg. LV (m/s)	SV (h^{-1})	Pressure drop (Pa)	Adsorption T (°C)	Desorption capacity (mmol CO_2/g)
1	0.88	28,746	28	21	0.69
2	0.70	22,862	28	21	0.55
3	1.05	34,311	27	21	0.55
4	0.76	24,836	29	30	0.62
5	0.75	24,498	27	30	0.51

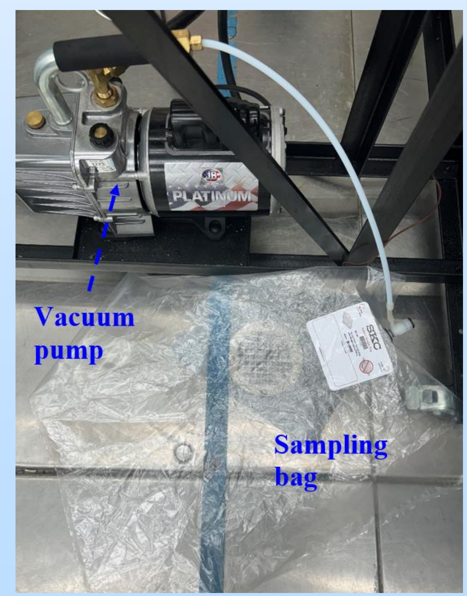




- ✓ Poor heat-transfer property of monolith is an issue for Joule heating.
- ✓ Total 2,025 cells (= 45 × 45); # of cells inside dashed line: 529 (= 23 × 23)
- ✓ Maximum recoverable CO_2 from outside dashed line = <74%
- ✓ Plan for desorption: vacuum convection oven







Lessons Learned

1. **Modified PEI** sorbent technology has potential for separating high-purity CO_2 at high working capacity with high CO_2 recovery from desorption at 85-90 °C.

2. Modified PEI sorbent technology has potential for switching from desorption to adsorption at ~50 °C without significant degradations.

3. Coating is critical when it needs to be used for structured system.

4. Poor heat-transfer property of cordierite is a hurdle for separation when non-steam direct injection-based desorption is used.

Plans for future testing/development

- Task 6: Performance evaluations
  - ✓ 100 cycles of small blocks with PEI and modified PEI-2 (weight, purity, degradation (MS, IR))
  - ✓ Large blocks

Parameters	Values
Block samples	Two modified PEI vs. PEI
T (°C)	0, 15, 30
RH (%)	20, 50, 80
Wind speed (m/s)	~3, 5, 7

- Task 7: TEA and LCA (UC + Trimeric)
  - ✓ Development of TCM and LCI
  - ✓ Assessment of DAC technology

- Future Plan
  - ✓ Hybrid monolith
  - ✓ Different structured DAC system with improved heat transfer properties
  - ✓ Partnerships for commercialization

Summary Slide

- Advanced sorbent technology from powdered form to structured form showing high CO_2 recovery and purity at 80-90 °C with resistance to degradations
- Successful development of sorbent-coated monolith for scalable DAC system
- Systematic and rational PAC design using CFD confirmed with experimental data
- Need for development of structured system with improved heat-transfer properties for scaled DAC system
- Open to partnerships for different structured system, scale-up, and commercialization