Computational Design and Process Intensification of \( \text{CO}_2 \) Absorbers

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Project Team
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Debangsu Bhattacharyya (WVU)

Acknowledgements: Many collaborators across CCSI\(^2\)
Objective: Design and optimize performance of novel, process-intensifying structured packing specific to solvent characteristics.

Approach: Interface with other CCSI² teams to leverage CCSI² expertise, utilizing ORNL capabilities in additive manufacturing and existing experimental infrastructure.
Absorber with intensified device

The intensified device provides intrastage cooling to maximize capture efficiency ► process intensification

Commercial packing
Device optimization relies on many components

- Experiments
- AM Prototyping
- Process Modeling
- ML / AI
- CFD Simulations
- SDoE

Optimized Device
EXPERIMENTS

Costas Tsouris, Gyoung Yang,
Josh Thompson, Aimee Jackson
Experimental activities are prepared to maximize utility of performance data for CFD model validation and candidate geometry evaluation

- Enhancement of existing 8-inch column for finer-grained measurements
  - Each flange has 4 ports for column performance measurements
  - Improved characterization of *operando* solvent properties (e.g., $\text{CO}_2$ concentration, amine concentration, water content, etc.)

- Designed reduced-size test section for more rapid simulations and experimental testing and prototyping
The 8-inch column has fine-grained measurement points

8-inch column at ORNL (Column A)

- Existing measurement points
- Added measurement points

- (a) HDPE solvent distributor
- (b) Commercial stainless-steel packing element with HDPE outer wall module
- (c) CO$_2$ absorption column setup
- (d) Installed intensified device
- (e) 3D printed intensified packing device
Performance data tools for model validation and candidate packing geometry evaluation are in place

Current measurement variables

On the column
– Pressure
– Temperature
– Gas flow rate & CO₂ concentration
– Solvent flow rate [column top & bottom]

In the lab
– Solvent CO₂ concentration
– Solvent density
– Solvent viscosity
– Solvent water content
– Solvent amine concentration
– Solvent–solid surface contact angle
Heat-transfer experiments were conducted to evaluate two intensified device geometries.

Experiments with the 8-inch diameter Column A

▶ Measured pressure drop, holdup, and residence time distribution of the solvent using intensified devices
Absorber column performance (1/2)

D=8” H=6.75” device for Column A (left) and D=12” H=16” device for Column B (right)

Heat-transfer experiments using Column A

Temperature profiles

No Cooling  Cooling

Run 11
Air-water system

[AMO-funded]
Future R&D Plan and Challenges

- Provide ground proof for iterative, progressive route to scale up absorber columns using commercially relevant and model-improved packing geometries
- Provide platform for computationally tractable validation data and rapid printed prototype testing

Virtual, reduced-size test section problems
- **Boundary flows** — handling boundary fluxes can cause numerical problems (solution stability)
- **Boundary conditions** — unknown spatial variations will be difficult to account for

Physical, reduced-size test section benefits
- **Exact validation target** — physical walls part of the simulation
- **Experimental operation ease** — less flow required
- **Tractable computational problem** — can run many tuning iterations using available resources in the limited time available

Later simulations will be run on larger sizes to discount wall effects
Options for reduced-size test sections (1/2)

**Option 1: New section in Column A**

- 8 in
- ~7 in
- H
- D

Each flange has ports for T&P measurements, fluids sampling, etc. for fine-grained validation data.

**Option 2: New benchtop column**

- H
- D

Currently, D = 2 inches is favored by the CFD team.

Sections can have nonequal heights etc.

Printing can allow fine control of surface roughness for potential interfacial area improvement.

Flows would be adjusted to compensate for different cross-sectional areas compared with the 8-inch column.
A reduced-size test section will be printed this year.
Validation data will be measured and transferred to the CFD team.
MODELING

System-level modeling, with detailed intensified absorber dynamics, to plan & guide future validation experiments

Josh Thompson (ORNL)
System-level modeling framework for design and planning

- Used to design ORNL 12” column [FEAA384] and plan experiments for ORNL 8” column [CCSI²].
- Leveraging to prepare for CCSI² experiments to support CFD validation.

\[ E = f(H_a, E_\infty) \]

\[ E = H_a \]

\[ E \text{: Enhancement factor} \]
\[ H_a \text{: Hatta number} \]

Thompson, Tsouris (2021). Rate-based absorption modeling for post-combustion CO₂ capture with additively manufactured structured packing. *Industrial & Engineering Chemistry Research* 60(41): 14845–14855. DOI: 10.1021/acs.iecr.1c02756
Comparison of two intensified devices

<table>
<thead>
<tr>
<th></th>
<th>Device 1</th>
<th>Device 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wall thickness</td>
<td>mm</td>
<td>0.733</td>
</tr>
<tr>
<td>Surface area</td>
<td>m²</td>
<td>2.132 (100%)</td>
</tr>
<tr>
<td>Coolant volume</td>
<td>L</td>
<td>0.784</td>
</tr>
<tr>
<td>Channel area</td>
<td>mm²</td>
<td>4408</td>
</tr>
</tbody>
</table>

Experimental conditions
- Gas flowrate from 400–700 LPM
- Liquid flowrate from 2.34–4.49 LPM
- Coolant flowrate from 0.96–2.25 LPM
- Liquid Temp from 50–70 °C
- Coolant temp from 7–22 °C

Modeling assumptions
- No reactions taking place – only air and water
- Temperature changes based entirely on enthalpy models of $\text{H}_2\text{O}$ and air
- MEA modeling correlations and heat and mass transfer equations used

Devices and HT experiments: AMO
Model validation runs: CCSI²
Model captures device heat-transfer performance

Experimental data from heat-transfer experiments shown above. Column contains normal Mellapak™ 250.Y sections with different intensified devices (gold boxes).
MODELING

Incorporation of MEA-property models from IDAES code into CFD simulation frameworks

Zachary Mills (ORNL) (OpenFOAM) and Yash Shah (NETL/Leidos) (ANSYS Fluent)
In consultation with WVU
**MEA mass transfer and thermochemical model**

**Mass Transfer model:**
- Interface assumed at equilibrium
- Henry’s law: \( \frac{c_l}{c_g} = He \)
- Reactions considered within the bulk liquid

<table>
<thead>
<tr>
<th>Gas</th>
<th>Interface</th>
<th>Liquid</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \text{CO}_2 )</td>
<td>( \text{CO}_2 )</td>
<td>( \text{CO}_2 + \text{MEA} \Leftrightarrow \text{Products} + \text{H}_2\text{O} )</td>
</tr>
<tr>
<td>diffusion</td>
<td>dissolution</td>
<td>chemical reactions</td>
</tr>
</tbody>
</table>

**Thermochemical properties:**
- Ideal gas property models for the gas phase
- MEA solvent properties extracted from the IDAES package
- Solvent reaction kinetics modeled using a two-reaction mechanism*:
  
  \[
  2\text{MEA} + \text{CO}_2 \Leftrightarrow \text{MEAH}^+ + \text{MEACOO}^- \\
  \text{MEA} + \text{CO}_2 + \text{H}_2\text{O} \Leftrightarrow \text{HCO}_3^- + \text{MEAH}^+
  \]

IDAES property package for MEA imported into ANSYS Fluent

Imported properties were verified to correspond exactly across the two frameworks (IDAES and Fluent; also, IDAES and OpenFOAM).

Properties and models incorporated include:

- **Instantaneous apparent/true species conversions**
- **Liquid-phase properties:**
  - Mixture density
  - Mixture viscosity
  - Mixture thermal conductivity
  - Mixture specific heat
  - Species diffusivities
- **Vapor-phase properties:**
  - Mixture density
  - Species & mixture viscosities
  - Species & mixture thermal conductivities
  - Species & mixture specific heats
  - Species diffusivities
- **Surface tension** (temperature & composition dependent)
- **Mass transfer** (liquid/gas) using Henry’s law
- **Reaction kinetics** (MEA–H₂O–CO₂ two-reaction mechanism)
CFD simulations with solvent thermochemical dynamics in new Volume of Fluids model of solvent layer in absorber device

Yash Shah (NETL/Leidos) (ANSYS Fluent) and Zachary Mills (ORNL) (OpenFOAM)

Simulations and visualizations shown here: Yash Shah
Simulation results: Interfacial and wetted areas

- Normalized interfacial area at pseudo-steady state: 0.928
- Normalized wetted area at pseudo-steady state: 0.314
- \((\Delta p)_{dry} = 4.754 \text{ Pa}\) calculated using a separate single-phase CFD simulation

⇒ Interfacial and wetted areas approach steady-state near \(t = 0.88\) seconds
⇒ Physical mass transfer and reaction rate kinetics were enabled after \(t = 0.88\) seconds and continued for another 0.1 seconds
Simulation results: solvent and gas-phase distributions
Simulation results: Liquid holdup & CO₂ absorption rate

Capture rate: 61.8%

Preliminary results from unvalidated model – work in progress
Future work

• Construct and operate rapid-prototyping, reduced-size test column section
  – CFD validation data
  – ML-identified candidate optimized geometries

• Scale up tests to 8- and 12-inch columns

• Consider effects of other solvents and/or materials of construction
Proposed pathway of dataflows & interfaces for application of modeling tools for optimized packing devices and columns

- Process model
  - 2-inch section fabrication
    - Validation data experiments
  - CFD model setup & verification
  - Validation simulations
  - Production simulations
    - ML surrogate model scoping
    - CFD surrogate model generation
    - Packing geometry optimization
  - Geometries from other subtasks?
  - Candidate evaluations
  - Printed coupons contact angles

- Reduced-scale column/section allows rapid print prototyping and experimental benchmarking of other candidate designs

- Candidate evaluations

- Iterate progressively from 2- to 8- to 12-inch sections
- Fewer evaluation experiments are anticipated with each increase in size

- Confidence in the validated CFD scale-up process would reduce uncertainty in predictions at larger scales where experiments or CFD would be limited or not possible.

- SDOE definition
- ORNL
- NETL
- LLNL
- WVU
- LANL
- PNNL
- CCSI²
For more information
https://www.acceleratecarboncapture.org/

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