Development of a Circulating Fluidized Bed for Flue Gas Carbon Capture using Solid Sorbent

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Overview

- A bench scale circulating fluidized bed system designated as the “Carbon Capture Unit” or “C2U” has been designed and built to evaluate the performance of sorbents on a solid substrate for removal of CO$_2$ from flue gases.
Program goals

• This project relates to the Existing Plants, Emissions and Capture (EPEC) Program within the Post-Combustion CO\textsubscript{2} Capture area and under the category of Solid Sorbents.

• Program goal: To develop fossil fuel conversion systems that can capture 90\% CO\textsubscript{2} while keeping the increase in cost of energy service below 35\%.*

Success criteria for sorbents: MATRIC* study

Initial performance:
\[30 - 50\% \text{ energy required for wet MEA process}\]

Minimum Delta Loading:
\[3.0 \text{ gmol CO}_2/\text{kg sorbent}\]

Temperature adsorption/desorption envelope:
\[40 - 110 \ C \ @ \text{ atm. press. with humidity}\]

Stability, durability and performance:
Sorbent must maintain its adsorption/desorption capability in the presence of water and other flue gases and maintain structural integrity through multiple cycles.

* Mid-Atlantic Technology, Research and Innovation Center (MATRIC), PROCESS ANALYSES AND R&D PLANS FORWARD FOR DRY-SORBENT-BASED PROCESSES FOR REMOVAL OF CO\text{2} FROM POWER PLANT FLUE GA S, July 19, 2006
Goals/objectives of C2U

• Flexible, inexpensive, small scale unit capable of validating CFD models including CO$_2$ adsorption and regeneration

• Desired Capabilities:
  – Evaluate heat transfer modes
  – Define mass transfer limits
  – Map hydrodynamic parameters
  – Validate sorbent kinetics
  – Quantify desired CO$_2$ loading on sorbent
  – Evaluate ability to isolate processes
  – Control process variables, such as mixing, flue gas composition, residence times, etc.
  – Evaluate reactor performance
Conceptual design

Concentrated CO₂

Loopseal#1
Separates flue gas from regenerated CO₂

Regenerator
CO₂ regeneration
Endothermic reaction

Heat added

Heat removed

Flue gas enters

Adsorber
CO₂ adsorption
Exothermic reaction

Sorbent without CO₂

Sorbent with CO₂

Cyclone
Separates solids from flue gas

Flue gas without CO₂

Loopseal#2
Separates flue gas from regenerated CO₂

Heat added?

Heat removed?

Flue gas without CO₂
C2U components

- Cyclone
- Diversion valve
- Concentrated CO₂ exit
- Regenerator
- Regenerator plenum
- Solids control valve
- L-valve
- Adsorber (5.5” ID)
- Adsorber plenum (Flue gas entrance)
- Loopseal #1 & plenum
- Loopseal #2 & plenum
- Collector
- Solids control valve
- 2” ID riser
- 5.5 to 2 in. ID transition
- Flue gas exit depleted CO₂
- Crossover - 1” ID
- Loopseal #1 & plenum
- Loopseal #2 & plenum
Design basis – Centerpoint of operation envelope

- **Sorbent**
  - Density: 2.0 g/cc
  - Diameter: 200 µm (Operation range: 70 to 400 µm)
  - Specific heat: 837 J/kg-K
  - Sorption capacity: 3.0 g-mol CO₂/kg sorbent

- **Flue gas composition**
  - Dry: 81.3 % N₂, 15.9 % CO₂, 2.8 % O₂, 0.0% H₂O
  - Wet: 68.1 % N₂, 13.5 % CO₂, 2.4 % O₂, 15.1% H₂O

- **Flue gas flow**
  - 3.5 x Uₘᵢᵢ in adsorber without cooling coils (116 slpm)
  - CO₂ flow: 18.4 slpm (1.26 x 10⁻² g-mol/s or 5.53x10⁻⁴ kg/s)
  - Equivalent to 5.0 kW (thermal) power plant. 1.75 kW with 35% efficiency
Design basis – Centerpoint of operation envelope

*Adsorber – 5.5” (0.14 m) ID, 18” (0.45 m) height, polycarbonate*

- **Sorbent circulation rate – with adsorber heat transfer**
  - CO₂ total flow: 5.53x10⁻⁴ kg/s (1.26x10⁻² g-mol/s)
  - 90% CO₂ capture
  - Sorbent loading: 3.0 g-mol CO₂/kg sorbent
  - Circulation rate:
    - \(0.9 \times (\frac{1.26 \times 10^{-2}}{3.0} \text{ g-mol CO}_2/\text{kg sorbent}) = 3.77 \times 10^{-3} \text{ kg/s} \quad (30 \text{ lb/hr})\)
- **Heat to be removed**
  - Heat of reaction
    - Adsorption heat of reaction (\(\Delta H = 1.51 \times 10^6 \text{ J/kg CO}_2\)):
      - CO₂ total flow: 5.53x10⁻⁴ kg/s (1.26x10⁻² g-mol/s)
      - 90% CO₂ capture
      - Heat produced: \(1.51 \times 10^6 \text{ J/kg CO}_2 \times 5.53 \times 10^{-4} \text{ kg/s} \times 0.9 = 752 \text{ J/s}\)
  - Sorbent sensible heat
    - Allow sorbent \(\Delta T = 30 \text{ C} \quad \text{(Enter @ 50 C leave @ 80 C)}\)
    - Sorbent specific heat 837 J/kg- C
    - \(Q_{\text{sorbent}} = 0.9 \times 5.53 \times 10^{-4} \text{ kg/s} \times 837 \text{ J/kg- C} \times 30 \text{ C} = 95 \text{ J/s}\)
    - Heat to be removed = 752 - 95 = 657 J/s
Design basis – Centerpoint of operation envelope

*Adsorber – 5.5” (0.14 m) ID, 18” (0.45 m) height, polycarbonate*

- **Required sorbent circulation rate - without adsorber heat transfer**
  - Heat produced by reaction 752 J/s
  - Sorbent circulation rate
    - Sorbent specific heat: 837 J/kg·C
    - Assume sorbent temp not permitted to exceed 80 C for adsorption, enters adsorber at 50 C (ΔT = 30 C)
    - Circulation rate: \((752 \text{ J/s})/(30 \text{ C} \times 837 \text{ J/kg·K}) = 3.0 \times 10^{-2} \text{ kg/s (237 lb/hr)}\)
  - Sorbent loading
    - \(0.9 \times (1.26 \times 10^{-2} \text{ g-mol/s})/3.0 \times 10^{-2} \text{ kg/s} = 0.4 \text{ g-mol CO}_2/\text{kg sorbent}\)
    - Some heat is removed by flue gas flow (approx. 10%) decreasing the required sorbent circulation rate calculated above
Measurements-Adsorber side

**Flue gas exit**
- Temp., press., gas flow rate, composition

**Adsorber**
- \(\Delta p\), temp, wall heat flux

**Adsorber plenum**
- (Flue gas entrance)

**Crossover - 1” ID**
- \(\Delta p\), temp

**Cyclone**
- \(\Delta p\), temp, wall heat flux

**Loopseal #2**
- \(\Delta p\), temp, wall heat flux

**Loopseal #2 plenum**

**2” ID riser**
- \(\Delta p\), temp, wall heat flux

**Transition**
- \(\Delta p\), temp

**Control:** Fluidizing gas flow and composition,
**Measure:** \(\Delta p\) across distributor, temperature

**Control:** Flue gas flow and composition,
**Measure:** \(\Delta p\) across distributor, temperature
Regeneration

\textit{Regenerator} – 5.5” (0.14 m) ID, 40” (0.45 m) height, \textit{polycarbonate with internal coils}

- \textbf{Heat addition for 100 \% CO}_2 \text{ regeneration}
  - Reverse amine reaction - 752 J/s
  - Heat substrate from 80 to 100 \degree C
    - Circulation rate: 3.77x10^{-3} \text{ kg/s} \quad Q_{\text{sorbent}} = 63 \text{ J/s} \quad \text{Total} = 815 \text{ J/s}
    - Circulation rate: 3.00x10^{-2} \text{ kg/s} \quad Q_{\text{sorbent}} = 501 \text{ J/s} \quad \text{Total} = 1253 \text{ J/s}
Heat transfer coils

Heat transfer within all reactors accomplished using two copper coils nested axisymmetrically.

Chilled water flows through the coils for cooling. Heat addition utilizes heated oil.

Heat transfer coefficient is determined using correlation from *Vreedenburg 1958 for horizontal tubes in a bubbling bed

\[
\frac{h_c D_i}{k_g} = 0.66 \Pr_g^{0.3} \left( \frac{\rho_s - \varepsilon}{\rho_g \varepsilon} \right)^{0.44} \text{Re}_D^{0.44} \quad \text{when} \quad \frac{\rho_s}{\rho_g} \text{Re}_p \leq 2050
\]

where

\[
\text{Re}_p = \frac{d_p \rho_g U_g}{\mu_g} \quad \text{Re}_D = \frac{D_i \rho_g U_g}{\mu_g}
\]

*Handbook of Fluidization and Fluid Particle Systems (Yang) p. 263
Heat addition by coils in regenerator

Using values at right and Vreedenburg correlation the following heat transfer coefficients are determined:

Heat transfer coeff of outer tube \( h \) = 188.6 W/m\(^2\)-K
\( hA_{outer} = 28.8 \text{W/K} \)

Heat transfer coeff of inner tube \( h \) = 211.8W/m\(^2\)-K
\( hA_{inner} = 17.52 \text{W/K} \)

Required Log Mean Temperature Difference

\[
\Delta T = \frac{Q}{hA_{outer} + hA_{inner}}
\]

For circulation rate \( 3.77 \times 10^{-3} \text{ kg/s} \) \( \Delta T = 17.6 \text{ K} \) since \( Q = 815 \text{ J/s} \)

For circulation rate \( 3.00 \times 10^{-2} \text{ kg/s} \) \( \Delta T = 27.0 \text{ K} \) since \( Q = 1253 \text{ J/s} \)

\( U_g = 2U_{mf} = 0.064 \text{ m/s} \)
\( \rho_{\text{gas}} = 1.28 \text{ kg/m}^3 \) @ operating pressure
\( \mu_{\text{gas}} = 1.81 \times 10^{-5} \text{ kg/m-s} \)
\( k_{\text{gas}} = 2.58 \times 10^{-2} \text{ W/m-K} \)
\( Pr = 0.713 \)
Void fraction = 0.516
Outer coil tube diameter = \( 1.27 \times 10^{-2} \text{ m} \)
Outer coil tube length = 3.83 m
Outer coil tube surface area = 0.229 m\(^2\)
Inner coil tube diameter = \( 9.67 \times 10^{-3} \text{ m} \)
Inner coil tube length = 2.55 m
Inner coil tube surface area = 0.116 m\(^2\)
Heat addition by coils in regenerator

Log mean temperature difference for a parallel flow heat exchanger

\[ LMTD = \frac{\Delta T_{x=0} - \Delta T_{x=L}}{\ln \left( \frac{\Delta T_{x=0}}{\Delta T_{x=L}} \right)} = \frac{(T_{h,i} - T_{c,i}) - (T_{h,o} - T_{c,o})}{\ln \left( \frac{T_{h,i} - T_{c,i}}{T_{h,o} - T_{c,o}} \right)} \]

- \( T_{h,i} \) - Temperature of oil at the inlet.
- \( T_{c,i} \) - Temperature of sorbent at the inlet.
- \( T_{h,o} \) - Temperature of oil at the outlet.
- \( T_{c,o} \) - Temperature of sorbent at the outlet.

Increase sorbent temperature from 80°C (\( T_{c,i} \)) to 100°C (\( T_{c,o} \)) and add regeneration energy

Using heated oil @ 4.0 liter/min (7.33x10\(^{-2}\) kg/s) sp. ht. = 1910 J/kg-K

\[ \Delta T_{oil} = \frac{Q}{\text{mass flow} \times \text{sp. ht.}} \]

Low circulation rate:

\[ \Delta T_{oil} = -5.8 \text{ °C} \quad \text{for LMTD} - 17.6 \text{ °C} \]

Oil inlet temp = 113.6 °C, Oil exit temp = 107.8 °C - Using LMTD equation:

High circulation rate:

\[ \Delta T_{oil} = -9.0 \text{ °C} \quad \text{for LMTD} - 27.0 \text{ °C} \quad - \text{sorbent circ rate } 3.00x10^{-2} \text{ kg/s} \]

Oil inlet temp = 124.0 °C, Oil exit temp = 115.0 °C
Heat removal by coils in Loopseal #1

Decrease sorbent temperature from 80 °C ($T_{h,i}$) to 50 °C ($T_{h,o}$)

Heat transfer coeff of outer tube (h) = 214 W/m²-K , $h_{A_{outer}} = 24.6$ W/K,  
Heat transfer coeff of inner tube (h) = 252 W/m²-K , $h_{A_{inner}} = 14.5$ W/K

$Q_{low\, circ} = 158$ J/s , LMTD = 4.0 °C , $Q_{high\, circ} = 1253$ J/s, LMTD = 32.1 °C

Using chilled water @ 4.0 liter/min (6.67x10⁻² kg/s)  
sp. ht. = 4181J/kg-K  
$\Delta T_{water} = Q/(\text{mass flow} \times \text{sp. ht.})$

Low circulation rate:  
$\Delta T_{water} = 0.6$ °C for LMTD - 4.0 °C  
Water inlet temp = 49.4 °C,  
water exit temp = 50.0 °C

High circulation rate:  
$\Delta T_{water} = 4.5$ °C for LMTD - 32.1 °C  
Water inlet temp = 33.2 °C,  
water exit temp = 37.7 °C
Measurements- Regenerator side

**Regenerator side**

- **Regenerator**
  - Δp, temp, wall heat flux
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp across distributor, temperature
  - Loopseal #1
  - Δp, temp, wall heat flux
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp, temperature

- **Regenerator plenum**
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp across distributor, temperature

- **L-valve**
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp, temperature

- **Regenerator standpipe**
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp, temperature

- **Regenerator heating coils**
  - oil inlet & outlet temp, flow rate
  - Concentrated CO₂ exit
  - Temp., press., gas flow rate, composition

- **Loopseal #1**
  - Control: Fluidizing gas flow and composition,
  - Measure: Δp across distributor, temperature
  - Loopseal #1 plenum
  - Loopseal #1 cooling coils
  - water inlet & outlet temp, flow rate
Conclusions

The design meets the criteria previously expressed

• Flexible, inexpensive, small scale unit capable of validating CFD models including CO\textsubscript{2} adsorption and regeneration

• Desired Capabilities:
  – Evaluate heat transfer modes
  – Define mass transfer limits
  – Map hydrodynamic parameters
  – Validate sorbent kinetics
  – Quantify desired CO\textsubscript{2} loading on sorbent
  – Evaluate ability to isolate processes
  – Control process variables, such as mixing, flue gas composition, residence times, etc.
  – Evaluate reactor performance