Acknowledgements:
…too many names to list…

Industrial Carbon Management Initiative (ICMI)

Project Review & Status

July 2012

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ICMI Research areas

Focus is on “industrial” applications: NG or coal boilers, process heat, chemical production, others. Technical results expected to benefit coal power as well.

Carbon Capture
Chemical Looping Combustion

Carbon Storage
Depleted Shale Fields

Carbon Utilization
Photocatalytic Conversion

CCUS for Industrial Applications

Industrial assessment and systems analysis
ICMI Research Areas

Focus is on “industrial” applications: NG or coal boilers, process heat, chemical production, others. Technical results expected to benefit coal power as well.
High-potential Industrial Applications

- CL industrial boilers
- CL for oil sands processing and production
- CO$_2$ sequestration in depleted shale gas reservoirs

ICMI Focus: Industrial Sources

Early industrial application... leading to electric power applications.

Reference for CO$_2$ Stationary Source Emissions by Category chart:
DOE’s Regional Carbon Sequestration Partnerships and NATCARB database.
U.S. Industrial Boiler Market (Natural Gas)

• 43,000 boilers in the U.S.
  – More than 50% are smaller than 3 MWt
• CO\textsubscript{2} emissions per boiler are comparable to some demonstration CCUS projects, or EOR wells
• Old infrastructure
  – For boilers > 3 MW\textsubscript{t}
    • 47% > 40 yrs old
    • 76% > 30 yrs old
  – Expected life 30 yrs
• NO\textsubscript{x} requirements
  – 30-80 ppm @ 3% O\textsubscript{2}
  – Larger units are lower

## CO₂ Separation Issues for Industrial Boiler Applications

<table>
<thead>
<tr>
<th>Technology</th>
<th>Issues</th>
</tr>
</thead>
</table>
| Flue-gas scrubbing (e.g. MEA) | • Capital investment for add-on  
                             • Chemical handling issues  
                             • Need economic cost studies |
| Membranes                   | • Low pressure flue gas  
                             • Potentially poor energetics  
                             • Need economic cost studies |
| Oxy-fuel                    | • Capital investment for ASU and exhaust gas recycle  
                             • NOx – O₂ purity trade-offs  
                             • CO₂ separation is simple  
                             • Need economic cost studies |
| Chemical Looping            | • Ultra-low NOx  
                             • CO₂ separation is simple  
                             • **Need economic cost studies**  
                             • **Need to validate technology** |
Where and how can chemical looping work?

**Industrial applications**
*(includes NG, smaller scale)*

**Power applications**
*(coal, 100+MW scale)*

Attributes:
- Fuel (NG, solid fuels)
- Size
- Cost
- Performance

System issues & configuration:
- Attrition
- Material supply & handling
- Heat exchanger/integration
- Sensors and control
- Emissions
- Carrier cost/supply & re-use

Components:
- Hydrodynamics
- Heat transfer
- Size/cost

Basic data:
- Carrier capacity
- Carrier reaction rate w/oxygen
- Carrier reaction rate w/fuel
- Carrier degradation

ICMI work elements provide the data and analysis.

Element 510 considered relevant industrial applications (next slides)
Defining the Application and Baseline for Economic Studies
# Industry - Capture Technology Matrix

<table>
<thead>
<tr>
<th>Refineries</th>
<th>Cement</th>
<th>Iron &amp; Steel</th>
<th>Oil &amp; Gas</th>
<th>Ethanol/Ethylene</th>
<th>Pulp &amp; Paper</th>
<th>Ammonia/Fertilizer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Process Heating - N4</td>
<td>Cement Kiln - N2</td>
<td>Traditional Blast Furnace - N5</td>
<td>O&amp;G Processing</td>
<td>Bioethanol via fermentation - N6</td>
<td>Kraft Mills - N5</td>
<td>Hydrogen Production</td>
</tr>
<tr>
<td>Steam/Utilities - N4</td>
<td>O&amp;G Processing Steam/Utilities</td>
<td>O&amp;G Processing Steam/Utilities - SAGD</td>
<td>Oil Sands Steam Production - Hydrogen</td>
<td>Ethylene</td>
<td>Steam and Heat</td>
<td>Hydrogen Production</td>
</tr>
<tr>
<td>Hydrogen Production</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>FCC Regeneration</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

### Potential
- Chemical Solvent
- Physical Solvent
- Sorbent
- Membrane
- Carbonate Looping
- Cryogenic Looping
- Oxyfuel
- Chemical Combustion
- Physical Solvent
- Membrane
- Chemical Solvent
- Oxyfuel

### Proven
- Chemical Solvent
- Physical Solvent
- Sorbent
- Membrane
- Carbonate Looping
- Cryogenic Looping
- Oxyfuel
- Chemical Combustion
- Physical Solvent
- Membrane
- Chemical Solvent
- Oxyfuel

### Preferred
- Chemical Solvent
- Physical Solvent
- Sorbent
- Membrane
- Carbonate Looping
- Cryogenic Looping
- Oxyfuel
- Chemical Combustion
- Physical Solvent
- Membrane
- Chemical Solvent
- Oxyfuel

### In Testing
- Chemical Solvent
- Physical Solvent
- Sorbent
- Membrane
- Carbonate Looping
- Cryogenic Looping
- Oxyfuel
- Chemical Combustion
- Physical Solvent
- Membrane
- Chemical Solvent
- Oxyfuel

### Capture not Required
- Chemical Solvent
- Physical Solvent
- Sorbent
- Membrane
- Carbonate Looping
- Cryogenic Looping
- Oxyfuel
- Chemical Combustion
- Physical Solvent
- Membrane
- Chemical Solvent
- Oxyfuel

### Notes
- N1: Will not be suited to retrofit -- new plant only
- N2: Pre-combustion not suitable due to lower radiant properties
- N3: Oxyfuel with CO2 removal via solvent
- N4: Post Combustion limited due to many point sources
- N5: Makes up majority of plants, ~70%
- N6: CO2 from fermentors only (no fuel) -- Produces relatively pure CO2
Potential Chemical Looping Application

- **Steam Production**
  - In any industrial or commercial facility where boilers are in use
  - Oil Sands production & processing, especially Steam Assisted Gravity Drainage (SAGD) very attractive
  - Oil & Gas production, especially where CO₂ could be used for EOR

- **Electric Power Generation**
  - Need to fully characterize size & complexity of the systems
  - Analysis coordinated with NETL studies of power systems
Industrial Boiler Steam Conditions

- **Oil Sands SAGD**
  - Saturated steam at 1000 to 1600 psi (Sat temp: 550°F to 610°F = 290 to 320°C)
  - 500,000+ lb/hr steam rate
  - Fueled by natural gas (could consider pet coke)

- **Oil & Gas Plant**
  - Saturated steam at varying pressure levels (LP [~50 psi] & MP [~300 psi] typical)
  - Variable steam rate
  - Fueled by natural gas

- **Refinery**
  - Saturated steam up to 900 psi pressure levels
  - 500,000+ lb/hr steam rate
  - Fueled by refinery gas

- **Recommendation for future systems analysis work**
  - Evaluate steam generation at 300, 600, 900 and 1500 psi levels
  - Natural gas fuel
  - Compare to SAGD and other industrial / commercial steam systems

Very different than power applications
Establishing a baseline case for an industrial boiler application with capture
Site Description and Conditions

- Unspecified location
- Generic conditions based on ISO specifications
- Site specific conditions can impact analysis, but comparisons are valid as long as design conditions are consistent across cases

<table>
<thead>
<tr>
<th>Elevation, (ft)</th>
<th>0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Barometric Pressure, MPa (psia)</td>
<td>0.10 (14.696)</td>
</tr>
<tr>
<td>Design Ambient Temperature, Dry Bulb, °C (°F)</td>
<td>15 (59)</td>
</tr>
<tr>
<td>Design Ambient Temperature, Wet Bulb, °C, (°F)</td>
<td>11 (51.5)</td>
</tr>
<tr>
<td>Design Ambient Relative Humidity, %</td>
<td>60</td>
</tr>
</tbody>
</table>
# Fuel – Natural Gas

## Natural Gas Composition:

<table>
<thead>
<tr>
<th>Component</th>
<th>Volume Percentage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methane (CH\textsubscript{4})</td>
<td>93.1</td>
</tr>
<tr>
<td>Ethane (C\textsubscript{2}H\textsubscript{6})</td>
<td>3.2</td>
</tr>
<tr>
<td>Propane (C\textsubscript{3}H\textsubscript{8})</td>
<td>0.7</td>
</tr>
<tr>
<td>n-Butane (C\textsubscript{4}H\textsubscript{10})</td>
<td>0.4</td>
</tr>
<tr>
<td>Carbon Dioxide (CO\textsubscript{2})</td>
<td>1.0</td>
</tr>
<tr>
<td>Nitrogen (N\textsubscript{2})</td>
<td>1.6</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>100.0</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>LHV</th>
<th>HHV</th>
</tr>
</thead>
<tbody>
<tr>
<td>kJ/kg</td>
<td>47,454</td>
<td>52,581</td>
</tr>
<tr>
<td>MJ/scm</td>
<td>34.71</td>
<td>38.46</td>
</tr>
<tr>
<td>Btu/lb</td>
<td>20,410</td>
<td>22,600</td>
</tr>
<tr>
<td>Btu/scf</td>
<td>932</td>
<td>1,032</td>
</tr>
</tbody>
</table>

## Natural Gas Cost:

**Baseline Studies:**
$6.55/\text{MMBtu}$, June 2007 dollars

**Updated Cost:**
$6.13/\text{MMBtu}$, June 2011 dollars

Assumes gas is delivered at 435 psig
Industrial Baseline Application Design

- **Steam Generator Capacity**
  - Case 1: 27,500 lb/hr (~10 MW Thermal)
  - Case 2: 275,000 lb/hr (~100 MW Thermal)
  - Steam is generated at 600 psi with 100°F of superheat
  - 80% boiler efficiency

- **Steam Generator Sparing Philosophy**
  - Assume no sparing

- **Reference Steam Generation Process**
  - Watertube Design (Characterization of the U.S. Industrial Commercial Boiler Population - large watertube boilers account for most steam production)

- **Carbon Capture**
  - Amine Scrubber
Industrial Reference Case Block Diagram

Capture Block
Industrial Reference Case Performance

- Heat rate was assumed to be natural gas feed and steam production was held constant on capture cases

<table>
<thead>
<tr>
<th>Units</th>
<th>10 MW&lt;sub&gt;TH&lt;/sub&gt;</th>
<th>12.4 MW&lt;sub&gt;TH&lt;/sub&gt;</th>
<th>100 MW&lt;sub&gt;TH&lt;/sub&gt;</th>
<th>124.4 MW&lt;sub&gt;TH&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>No Capture</td>
<td>Capture</td>
<td>No Capture</td>
<td>Capture</td>
<td>Units</td>
</tr>
<tr>
<td><strong>Auxiliary Load</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Boiler Feedwater Pumps</td>
<td>20</td>
<td>20</td>
<td>180</td>
<td>190</td>
</tr>
<tr>
<td>Amine System Auxiliaries</td>
<td>0</td>
<td>100</td>
<td>0</td>
<td>1,100</td>
</tr>
<tr>
<td>Circulating Water Pump</td>
<td>0</td>
<td>40</td>
<td>0</td>
<td>330</td>
</tr>
<tr>
<td>Ground Water Pumps</td>
<td>4</td>
<td>10</td>
<td>40</td>
<td>70</td>
</tr>
<tr>
<td>CO&lt;sub&gt;2&lt;/sub&gt; Compression</td>
<td>0</td>
<td>170</td>
<td>0</td>
<td>1,710</td>
</tr>
<tr>
<td>Cooling Tower Fans</td>
<td>0</td>
<td>20</td>
<td>0</td>
<td>170</td>
</tr>
<tr>
<td>Air Compressor</td>
<td>40</td>
<td>40</td>
<td>350</td>
<td>440</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td>64</td>
<td>400</td>
<td>570</td>
<td>4,010</td>
</tr>
</tbody>
</table>

| **Plant Performance** |                  |                       |                      |                        |
| Net Auxiliary Load | 64               | 400                   | 570                  | 4,010                  | kW<sub>e</sub> |
| **Net Plant Efficiency (HHV)** | 0.838            | 0.647                 | 0.838                | 0.647                  | Fraction |
| **Net Plant Efficiency (LHV)** | 0.928            | 0.717                 | 0.928                | 0.717                  | Fraction |
| Natural Gas Feed Flow | 685 (1,510)      | 852 (1,879)           | 6,848 (15,098)       | 8,522 (18,788)         | kg/hr (lb/hr) |
| Thermal Input (HHV) | 9,977            | 12,416                | 99,774               | 124,160                | kW<sub>th</sub> |
| Thermal Input (LHV) | 8,996            | 11,195                | 89,959               | 111,946                | kW<sub>th</sub> |
| 600 psia Steam Produced | 23,175           | 23,175                | 231,754              | 231,754                | lb/hr |
| 73.5 psia Steam Required | 0               | 7,798                 | 0                    | 77,978                 | lb/hr |
| Raw Water Consumption | 23,175           | 23,175                | 231,754              | 231,754                | lb/hr |

Notice the efficiency
Chemical Looping Application Analysis
10 MW\textsubscript{th} and 100 MW\textsubscript{th}
Key Model Assumptions Initially Applied

- Reducer reactor type:
  - Bubbling fluid bed/turbulent fluid bed
- Oxidizer reactor type:
  - Bubbling fluid bed/circulating bed
- Fluid bed gas-carrier contact: plug flow (optimistic)
- Carrier type: Fe$_2$O$_3$ on alumina support
- Carrier particle size: 0.15 mm
- Carrier reaction resistances: only shrinking grain resistance
- Solids transport: dilute pneumatic transport for bubbling bed case/none for circulating bed case
## Approximate Sizes

Based on existing data; subject to revisions with other carriers/reactor concepts

<table>
<thead>
<tr>
<th></th>
<th>Reducer</th>
<th>Oxidizer</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Bubbling Beds</td>
<td>Bubbling Beds</td>
</tr>
<tr>
<td>Natural Gas Input (MW\text{th})</td>
<td>10</td>
<td>100</td>
</tr>
<tr>
<td>Vessel diameter (ft)</td>
<td>4.3</td>
<td>10.7</td>
</tr>
<tr>
<td>Vessel height (ft)</td>
<td>43</td>
<td>38</td>
</tr>
<tr>
<td>Bed height (ft)</td>
<td>23</td>
<td>11</td>
</tr>
<tr>
<td>Bed outlet velocity (ft/s)</td>
<td>17</td>
<td>17</td>
</tr>
<tr>
<td>Cyclone number</td>
<td>1</td>
<td>4</td>
</tr>
<tr>
<td>Cyclone diameter (ft)</td>
<td>3.6</td>
<td>5.1</td>
</tr>
<tr>
<td>Cyclone height (ft)</td>
<td>13</td>
<td>20</td>
</tr>
<tr>
<td>Solids transport cyclone diameter (ft)</td>
<td>3.1</td>
<td>7.7</td>
</tr>
<tr>
<td>Solids transport cyclone height (ft)</td>
<td>11</td>
<td>34</td>
</tr>
<tr>
<td>Baghouse length and width (ft)</td>
<td>7</td>
<td>17</td>
</tr>
<tr>
<td>Baghouse height (ft)</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>HRSG diameter (ft)</td>
<td>2.0</td>
<td>6.3</td>
</tr>
<tr>
<td>HRSG length (ft)</td>
<td>20</td>
<td>20</td>
</tr>
</tbody>
</table>
### Process Performance

<table>
<thead>
<tr>
<th>CLC Design Concept</th>
<th>BUBBLING FLUID BEDS, PLUG FLOW GAS</th>
<th>CIRCULATING OXIDIZER, TURBULENT REDUCER PLUG FLOW GAS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural Gas Feed Rate (MWth, HHV)</td>
<td>10 100</td>
<td>10 100</td>
</tr>
<tr>
<td><strong>PROCESS HEAT BALANCE</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Natural gas energy input (MMBtu/hr, HHV)</td>
<td>34.12 341.20</td>
<td>34.12 341.20</td>
</tr>
<tr>
<td>Total product steam generation (MMBtu/hr)</td>
<td>26.72 270.19</td>
<td>27.37 284.98</td>
</tr>
<tr>
<td>Reducer vessel product steam generation (MMBtu/hr)</td>
<td>0.00 0.00</td>
<td>0.00 0.00</td>
</tr>
<tr>
<td>Oxidizer vessel product steam generation (MMBtu/hr)</td>
<td>15.26 152.60</td>
<td>16.78 167.80</td>
</tr>
<tr>
<td>Reducer CO2 offgas product steam generation (MMBtu/hr)</td>
<td>3.85 38.65</td>
<td>3.89 38.95</td>
</tr>
<tr>
<td>Oxidizer offgas product steam generation (MMBtu/hr)</td>
<td>7.61 78.94</td>
<td>6.71 78.22</td>
</tr>
<tr>
<td>Oxidizer offgas stripping steam generation (MMBtu/hr)</td>
<td>0.29 2.90</td>
<td>0.29 2.90</td>
</tr>
<tr>
<td>Vessel heat losses (MMBtu/hr)</td>
<td>0.94 4.39</td>
<td>1.87 4.94</td>
</tr>
<tr>
<td>CO2 product stream unburned fuel (MMBtu/hr, HHV)</td>
<td>0.47 4.66</td>
<td>0.47 4.65</td>
</tr>
<tr>
<td>Flue gas, CO2 product and vent streams sensible heat</td>
<td>5.70 59.07</td>
<td>4.12 43.73</td>
</tr>
<tr>
<td>Boiler Efficiency based on product steam (%)</td>
<td>78.3 79.2</td>
<td>80.2 83.5</td>
</tr>
</tbody>
</table>
Layout 100 MW CLC (110’ x 128’ x 50’)

PLAN VIEW

ELEVATION "A-A"

100 MW LAYOUT

BILL OF MATERIAL

CHEMICAL LOPPING REACTOR
MECHANICAL EQUIPMENT
EQUIPMENT ELEVATIONS AND DETAILS

SK-001
Where and **how** can chemical looping work?

*Industrial applications*  
(includes NG, smaller scale)

*Power applications*  
(coal, 100+MW scale)

**Attributes:**
- Fuel (NG, solid fuels)
- Size
- Cost
- Performance

**System issues & configuration**
- Heat and material balances
- Attrition
- Material supply & handling
- Heat exchanger/integration
- Sensors and control
- Emissions
- Carrier cost/supply & re-use

**Components**
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- Size/cost

**Basic data**
- Carrier capacity
- Carrier reaction rate w/oxygen
- Carrier reaction rate w/fuel
- Carrier degradation

Iterate with more information

ICMI work elements provide the data and analysis.

These data will enable CCSI scale-up simulation.
Detailed Modeling Tools

• Low-Fidelity Model
  – Excel based model used to validate basic material and energy balance of CLR
  – Includes pressure drop calculations and computed Heat & Material balance for at least five operating conditions
  Important to affirm that the solids circulate as desired.

• High-Fidelity CFD
  – “Cold Flow” simulations complete, awaiting experimental validation
  – “Hot Flow” simulations have been constructed
  – Gen 1 kinetics and 3 baseline operating conditions underway

CLR whole system – 3D, front view
Validating the Predictions: Laboratory Scale Chemical Looping Reactor (CLR)

Current Status: Being Installed at NETL

CLR Vessels Delivered to NETL

Project Structure

Air Reactor Bubble Caps
Validating the Predictions: Laboratory Scale Chemical Looping Reactor (CLR)

Current Status: Being Installed at NETL

- Cyclone C-1200
- Test Section C-1250
- Loop Seal R-1300
- Fuel Reactor R-1400
- L-Valve Housing R-1450
- Air Reactor R-1000
- Air Pre-heater and Tee H-1800 & H-1850
- Upper Riser R-1150
- Lower Riser R-1100

CLR Vessels Delivered to NETL Project Structure

- Air Reactor
- Bubble Caps
- Fuel Reactor
- Separation cyclone
- Riser
- Cross over
- Loop Seal
- L-Valve
- Solids Flow

Solid Volume Fraction

- 6.00E-01
- 3.80E-01
- 2.40E-01
- 1.52E-01
- 9.61E-02
- 6.08E-02
- 3.85E-02
- 2.43E-02
- 1.54E-02
- 9.74E-03
- 6.16E-03
- 3.90E-03
- 2.47E-03
- 1.56E-03
- 9.87E-04
- 6.24E-04
- 3.95E-04
- 2.50E-04
- 1.58E-04
- 1.00E-04
Non-Reacting Cold Flow Unit

- Used to simulate and characterize the behavior of solids transfer and the control of oxygen carrier particles.

- Measured characteristics: gas-particle velocity fields, 3-D solid-void fraction distributions, bubble size, bubble frequency.

- Geometry and flow match the hot unit except for the temperature.

- Acrylic construction allows for visual identification of the flow structures and use of advanced instruments such as high speed particle imaging velocimetry.

- Provides hydrodynamic validation data for various models and provides a similar system to explore control strategies.
Where and **how** can chemical looping work?

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*includes NG, smaller scale*  

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- Carrier degradation

Iterate with more information

ICMI work elements provide the data and analysis.
Oxygen Carrier Development

- **Carriers for CLR study have been identified and full report on screening study is available**
  - Hematite – natural ore
  - Cu-Fe/Al2O3 - synthetic material
    - Mixed-metal oxides developed at NETL
- **Vendors have been identified to provide materials**
- **Quality testing underway on vendor-supplied hematite**

### Oxygen Carrier Development

<table>
<thead>
<tr>
<th></th>
<th>Reduction rate (min⁻¹)</th>
<th>Oxidation rate (min⁻¹)</th>
<th>Oxygen transfer capacity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ilmenite</td>
<td>0.18</td>
<td>0.49</td>
<td>4.6</td>
</tr>
<tr>
<td>Hematite</td>
<td>0.33</td>
<td>0.52</td>
<td>10</td>
</tr>
</tbody>
</table>

Example of TGA cycle studies shows good stability and oxygen capacity.
Attrition Unit Shakedown Using Alumina Powder

Boring is Better!
Summary

• **Industrial Carbon Management Initiative:** technologies and validated simulation tools for carbon capture and storage from industrial sources:
  – Chemical Looping (CL) as a capture technology
  – Depleted shale gas reservoirs for CO₂ sequestration
  – Basic research in conversion of CO₂ to useful chemicals using light or waste heat

• **Research in progress covers**
  – Economic analysis of promising industrial CL applications
  – Development of oxygen carriers and reactor configurations
  – Validation of numeric models for detailed simulations & scale-up

• **Commercial and research interest is welcome!**
ICMI Reports (contact NETL)

- 2011 Annual Report on ICMI Project
- Literature Survey of Kinetic Parameters Relevant to Chemical Looping Combustion
- Chemical Looping Kinetic Rate Model
- Literature Review of Attrition Testing
- Literature Review of Solid-Solid Separation
- Evaluation of Commercially Available Solids Flow Sensors and Technologies for Chemical Looping Application
- Oxygen Carrier Development for Chemical Looping Combustion with Natural Gas Literature Review
- The Development of Applicable Oxygen Carrier Materials for Chemical Looping Combustion Using Methane as Fuel
- Modeling Lifetime of Corrodible Components Literature Review
- Hydrogen Production Screening Study
- Ca-Sorbent Development for Carbon-neutral Industrial Gas Production of Hydrogen Using Ca Looping
- Design Basis for Storage of CO₂ in Depleted Shale Gas Reservoirs
- CFBC Furnace Temperature and Other Considerations