Pressurized chemical-looping combustion of coal with iron ore as oxygen carrier in a pilot-scale unit

Shuai Zhang
Supervisor: Prof. Rui Xiao
School of Energy & Environment
Southeast University (SEU)
Contents

1. R&D of CLC at Southeast University (SEU)
2. Pressurized Pilot-Scale CLC Unit
3. Experiment and Results
4. Conclusion
5. Future Work at Southeast University
Technical roadmap

Technology

- Development of oxygen carriers
  - Reactivity test with TGA, bench scale reactor
  - Reactivity test for solid fuels
  - Performance in larger reactor
  - Performance in pilot scale reactor
  - Industrial demonstration of CLC technology

- Low cost materials
  - Nonmetal oxide: natural anhydrite (CaSO4/CaS)
  - Metal oxides:
    - Iron ores from Australia, Brazil, China, etc
    - Iron oxide scale from industry
    - Ilmenite

- Analysis of Ca-, Fe-, Ni-based oxygen carriers using gaseous fuels (CH4, CO, H2, etc) from the aspects of thermodynamic.

- Test of Fe- and Ca-based oxygen carriers with different solid fuels (coal, biomass) to investigate the effect of various influencing factors (temperature, pressure, etc) on the reactivity of oxygen carriers in atmospheric and pressurized bench scale reactors as well as a 10 kWth interconnected fluidized bed reactor.
Background

Major challenges for CLC of coal

- Low operating temperature (~1000 °C)
  - Difficult to couple with advanced power generation system
  - Low efficiency compared with traditional combustion
- Slow gasification rate, limiting step

Pressurized Chemical-Looping Combustion Combined Cycle (PCLC-CC) may be a solution

- High combustion efficiency of coal
- Potential higher system efficiency for power generation, steam turbine + gas turbine
- Low cost of CO₂ capture
Investigation of the reactivity and stability of three low cost Fe-based oxygen carriers with coal as fuel in a pressurized bench scale fixed bed reactor.

Pressure play a positive role in enhancing CO2 capture efficiency and carbon conversion.
Existing established technologies at Southeast University

- Pressurized Fluidized Bed Combustion-Combined Cycle
  
  R&D at Southeast University ~ 30 years
  
  - 1 MWt PFBC test facility (SEU-PFBC);
  - 15 MWt PFBC-CC pilot plant;
  - 2 MWt pressurized spout-fluid bed coal gasifier for 2G PFBC-CC
  - 2.5 MWt FBC pilot scale unit for oxy-fuel combustion (in progress)
Background

15MW PFBC-CC  2.5MW oxy-fuel FBC
Existing established technologies at Southeast University

- Chemical-Looping Technology R&D at Southeast University ~ 9 years
  - 1 kWth chemical-looping combustor;
  - 10 kWth chemical-looping combustor;
Background
Objective

- Demonstrate the feasibility of the new design of the PCLC continuous unit;
- Examine the potential of iron ore as a low-cost oxygen carrier for commercial coal-fueled CLC unit.
Features

- Due to the slow coal gasification process, 8m height of reaction zone of fuel reactor (FR) was adopted to provide sufficient time for coal gasification and reduction reaction.
- Compared with the FR operated in bubbling fluidization regime, FR operated in fast fluidization regime could provide a perfect gas-solid contact and a much more homogeneous mixture of oxygen carrier particles and coal particles.
Pressurized Pilot-Scale CLC Unit

- Preheater
- Coal feeder
- Data acquisition system
- Auxiliary facilities

PCLC combustor
## Chemical analysis of MAC iron ore

<table>
<thead>
<tr>
<th>Sample</th>
<th>Fe$_2$O$_3$</th>
<th>SiO$_2$</th>
<th>Al$_2$O$_3$</th>
<th>P$_2$O$_5$</th>
<th>MgO</th>
<th>SO$_3$</th>
<th>MnO</th>
<th>TiO$_2$</th>
<th>CaO</th>
</tr>
</thead>
<tbody>
<tr>
<td>MAC</td>
<td>91.59</td>
<td>3.17</td>
<td>2.33</td>
<td>0.288</td>
<td>0.256</td>
<td>0.202</td>
<td>0.197</td>
<td>0.129</td>
<td>0.092</td>
</tr>
</tbody>
</table>

## Analysis of Shenhua bituminous Coal

<table>
<thead>
<tr>
<th>Sample</th>
<th>Ultimate analysis/%</th>
<th>Heat Value</th>
<th>Proximate analysis/%</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C$<em>{ad}$ H$</em>{ad}$ O$<em>{ad}$ N$</em>{ad}$ S$_{ad}$</td>
<td>MJ·Kg$^{-1}$</td>
<td>FC$<em>{ad}$ V$</em>{ad}$ A$<em>{ad}$ M$</em>{ad}$</td>
</tr>
<tr>
<td>coal</td>
<td>70.56 4.79 9.58 1.04 0.71</td>
<td>26.02</td>
<td>54.48 32.2 7.81 5.51</td>
</tr>
</tbody>
</table>

## Operation conditions

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Test 1</th>
<th>Test 2</th>
<th>Test 3</th>
</tr>
</thead>
<tbody>
<tr>
<td>Operation pressure MPa</td>
<td>0.1</td>
<td>0.3</td>
<td>0.5</td>
</tr>
<tr>
<td>Coal feeding rate kg/h</td>
<td>7</td>
<td>7</td>
<td>7</td>
</tr>
<tr>
<td>Steam temperature (FR) °C</td>
<td>800</td>
<td>800</td>
<td>800</td>
</tr>
<tr>
<td>Air temperature °C</td>
<td>300</td>
<td>300</td>
<td>300</td>
</tr>
<tr>
<td>Bed temperature in FR °C</td>
<td>950</td>
<td>950</td>
<td>950</td>
</tr>
<tr>
<td>Bed temperature in AR °C</td>
<td>970</td>
<td>970</td>
<td>970</td>
</tr>
<tr>
<td>Hours of stable operation h</td>
<td>4.5</td>
<td>5.0</td>
<td>4</td>
</tr>
</tbody>
</table>
Experiment and Results

Test profile

Temperature and Pressure profile of Fuel reactor (FR)

0.1 Mpa

0.3 MPa
Experiment and Results

Outlet gas concentration of FR

0.1 Mpa

0.3MPa
Experiment and Results

Gas Concentration profile

![Graph showing gas concentration profile with different gases (CO2, CO, CH4, H2) against pressure in MPa.](image-url)
Experiment and Results

Carbon conversion

$$\eta_{FR, SF} = \frac{12 F_{C, FR}}{m_{SF} \times [C]_{SF}}$$

Gas conversion

$$\alpha_{FR, GC} = 1 - \frac{0.5[H_2] + 0.5[CO] + 2[CH_4]}{[CO_2] + [CO] + [CH_4]}$$

Carbon capture efficiency

$$\eta_{cc} = F_{C, FR} / \left( F_{C, FR} + F_{C, AR} \right)$$
## Experiment and Results

**Fly ash collected from Cyclone of FR**

### Fly Ash Characteristics

<table>
<thead>
<tr>
<th></th>
<th>0.1 MPa</th>
<th>0.3 MPa</th>
<th>0.5 MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>Time-averaged mass flow rate (kg/h)</td>
<td>1.35</td>
<td>2.66</td>
<td>3.18</td>
</tr>
<tr>
<td>Average particle size (μm)</td>
<td>48</td>
<td>57</td>
<td>69</td>
</tr>
<tr>
<td>Carbon content (%)</td>
<td>14.21</td>
<td>7.54</td>
<td>6.45</td>
</tr>
</tbody>
</table>
Experiment and Results

SEM analyses of used OC particles

0.1Mpa

0.3MPa

0.5Mpa
Conclusions

- The new design of the PCLC continuous reactors was demonstrated and a total duration of coal feeding of about 19 h and a total test of 13.5 h stable operation was realized;
- High CO2 Concentration and high carbon conversion could be obtained at relative high pressures;
- The potential of iron ore as a low-cost OC was well examined for commercial coal-fueled CLC unit;
- High temperature, high pressure chemical-looping combustion of coal is feasible and promising.
Future work

- Improvement of current PCLC unit and continue our research on this unit;
- Test of other low-cost oxygen carriers and prepare oxygen carriers with high reactivity and mechanical strength
- Multiphase CFD simulation of CLC process
- Design of Novel CLC reactors with the separation of ash and unreacted coal particles from oxygen carrier considered
- Building the relationship of coal rank and oxygen carrier with both reactivity and physiochemical properties of both coal char and oxygen carrier particles considered.
Thanks for your attention!