Abstract

To protect the environment changing, coal combustion must reduce its CO₂ emission by capture and storage. Chemical looping is a potentially high technology for coal combustion with CO₂ capture efficiently. A chemical-looping combustion system, using circulating fluidized bed which consist two main reactors, a fuel reactor and an air reactor. An oxygen carrier, typically a metal oxide, is employed to transfer oxygen from the air reactor to the fuel reactor, and circulates between these two reactors.

During 2012-2014, JCOAL, MHPS (Mitsubishi Hitachi Power Systems (2012-2013 Babcock-Hitachi)) and IAE(The Institute of Applied Energy) have a project funded by NEDO (New Energy and Industrial Technology Development Organization) to surveyed chemical looping technology development in the world, studied market needs of chemical looping combustion, investigated carrier costs and reactivity, etc., in order to reduce CO₂ separation recovery cost to 2,500 yen/ton of CO₂ or less. A small-scale reactor was used to study the behaviors of the direct reaction of coal with an oxygen carrier. It was found that the coal conversion efficiency increases with increasing iron level in the carrier, that the volume of unreacted CO gas increases when oxygen use in the carrier exceeds the range of Fe₂O₃ → Fe₃O₄, and that there is no notable surface melting that could obstruct particle circulation at 950°C or less.

A basic model of the CLC process was produced using the AspenPlus software and used to analyze the process under the conditions of a 250MWth plant. It found that the volume of circulating CLC carrier is roughly the same as the circulation of CFBC medium, and that inner desulfurization and ultra-low NOₓ combustion are possible.

Using the results of research and process analysis, a three-tower chemical looping coal combustion technology that consists primarily of an air reactor (AR), coal reactor (CR), and volatiles reactor (VR) was selected, and an conceptual design was produced for the reactor configuration and technical parameters.

Keywords: CO₂ capture, Coal, Combustion, Chemical looping

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1. Introduction

To protect the environment changing, coal combustion must reduce its CO₂ emission by capture and storage. Chemical looping is a potentially high technology for coal combustion with CO₂ capture efficiently.

A chemical-looping combustion system, which is schematically presented in Fig. 1, using circulating fluidized bed with consist two main reactors, a fuel reactor and an air reactor. An oxygen carrier, typically a metal oxide, is employed to transfer oxygen from the air reactor to the fuel reactor, and circulates between these two reactors.

![Schematic diagram of CLC](image)

The main reactions by using Fe base carrier in the fuel reactor and air reactor are:

**Fuel reactor:**  \[ \text{Fe}_2\text{O}_3 + \frac{1}{6}\text{C(Coal)} \rightarrow \frac{2}{3}\text{Fe}_3\text{O}_4 + \frac{1}{6}\text{CO}_2 \]

**Air reactor:**  \[ \frac{2}{3}\text{Fe}_3\text{O}_4 + \frac{1}{6}\text{O}_2(\text{Air}) \rightarrow \text{Fe}_2\text{O}_3 + Q \]

During 2012-2014, JCOAL, MHPS (Mitsubishi Hitachi Power Systems (2012-2013 Babcock-Hitachi)) and IAE(The Institute of Applied Energy) have a project funded by NEDO (New Energy and Industrial Technology Development Organization) to surveyed chemical looping technology development in the world (2012), studied market needs of chemical looping combustion (2013), investigated carrier costs and reactivity, etc., in order to reduce CO₂ separation recovery cost to 2,500 yen/ton of CO₂ or less (2014).

2. Market research

Fig. 2 shows CLC market as possible in a range of 100MWe -500MWe. From the perspectives of both engineering companies and suppliers of fluidized beds and other technologies used in CLC, the market research surveyed paper, cement, and other plants that use industrial boilers and would likely be early adopters of CLC. While CLC is a desirable technology for the future because it does not require energy to perform CO₂ separation and capture, there is a need to determine its safety and operational performance when used for power generation. Based on the findings of the research, it was also conducted on the use of CLC by IPPs and PPSs.
A study of market shown that the cost of CO₂ capture, 2,500 yen/t-CO₂ is a target for CLC development to achieve commercial viability in around 2030 (Fig.3).

3. CLC process concept design

A small-scale reactor was used to study the behaviors of the direct reaction of coal with an oxygen carrier and carrier reduction by CO gas. It was found that the coal conversion efficiency increases with increasing iron level in the carrier, that the volume of unreacted CO gas increases when oxygen use in the carrier exceeds the range of Fe₂O₃ → Fe₃O₄, and that there is no notable surface melting that could obstruct particle circulation at 950°C or less.

A basic model of the CLC process was produced using the AspenPlus software and used to analyze the process under the conditions of a 250MWth plant. It found that the volume of circulating CLC carrier is roughly the same as
the circulation of CFBC medium, and that inner desulfurization and ultra-low NOX combustion are possible (Fig.4)

Results of the Three Tower CLC process analysis

Analysis Conditions:
Boiler Size: 250MWth
Coal Supply: 36,655 kg/h
Inner De-Sulfur

Using the results of research and process analysis, a process that consists primarily of an air reactor (AR), coal reactor (CR), and volatiles reactor (VR) was selected, and an conceptual design was produced for the reactor configuration and technical parameters.(Fig.5).

4. Plan of the CLC development

We implemented identification and management of CLC technological development plan as well as a review of technological development roadmap, based on the results of technical and market research.
Regarding technological development plan, it is expected that CLC reactors will be constructed of circulating fluid beds and iron-based carrier cycles that have several tens of times the coal supply rate. Based on the above technical studies and market research results, we reviewed the technological development roadmap (Fig. 6).

**Phase I: A 6 years plan including 1 MW bench scale facility testing (NEDO project) have been started in 2015.**

![CLC development schedule of the plan](image)

**5. Conclusion**

Based on the results of three years research for technology progress over sea, CLC market, process concept, and the development target, a 6 years development plan was fixed.

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