Coal Direct Chemical Looping (CDCL) Process

Fanxing Li, Hyung Kim, Deepak Sridhar, Liang Zeng, Fei Wang, L. -S. Fan[1]
The Ohio State University, Columbus, OH. U.S.A.

Abstract

The Coal Direct Chemical Looping (CDCL) process is a novel technology that produces hydrogen from coal with in-situ CO_2 separation. The CDCL uses a composite iron oxide particle as an oxygen carrier for coal conversion. These particles are reduced by coal to capture CO_2 in the reducer. The reduced particles then are used in the second reactor to produce hydrogen through oxidation using steam. They can also be used to react with air for heat and power generation. The indirect combustion of coal with composite iron particles significantly enhances the efficiency of the hydrogen production process. The demonstration results carried out in a bench scale unit show more than 95% conversion of coal and more than 99% conversion of coal volatiles.

In this paper, the CDCL process is first reviewed. The demonstration results are then presented. Effect of sulfur and ash on the performance of the composite iron oxide particle is also discussed.

1. Introduction

Coal combustion power plant accounts for a third of the total CO_2 emission in the United States (Herzog, 1997). In order to capture and sequestrate the CO_2 from the coal combustion flue gas, significant energy and economic penalty have to be paid. For instance, CO_2 capture can reduce the energy conversion efficiency of a coal combustion power plant by up to 40% (NETL, 2007, Li, 2008). With projected increase in coal consumption for power generation, hydrogen generation, and fuel synthesis, novel coal conversion techniques that can reduce the CO_2 capture cost with improved energy conversion efficiency is highly desirable. Chemical-looping combustion, which integrates CO_2 capture in the fuel conversion scheme, is a promising approach for clean and efficient coal conversions (Ishida, 1994 and Mattisson, 2003).

In this paper, a novel coal direct chemical looping (CDCL) process which directly converts coal into hydrogen and/or electricity is presented. Experimental results obtained from a bench scale moving bed unit is reported and discussed.

2. Process Overview

The CDCL process, illustrated in Figure 1, utilizes composite Fe_2O_3 particles to convert coal into hydrogen and/or electricity at any relative proportions (Gupta, Velazquez-Vargas et al. 2004; Thomas, Fan et al. 2004; Fan, Gupta et al. 2007). In the CDCL process, composite Fe_2O_3 particles are introduced into the reducer along with pulverized coal and sub-stoichmetric amount of oxygen from the Air Separation Unit (ASU). With a desired gas-solid contacting pattern, coal is gasified in-situ and reacted

---

[1] Corresponding Author

with Fe_2O_3 particles. As a result, a mixture of metallic iron and Wuestite is produced along with a flue gas stream composed of CO_2, H_2O, and contaminants such as H_2S and elemental mercury. After condensing out the steam, the flue gas can be compressed and sequestrated. The reduced metallic iron/Wuestite particle from the reducer will then enter the oxidizer to react with steam to form hydrogen. The resulting Fe_3O_4 exiting the oxidizer will be subsequently combusted with air in the entrained flow combustor, which conveys the particle back to the reducer pneumatically while regenerating the particle to its original fully oxidized form. The heat released in the combustor will be partially carried by the exhaust gas, which can be used for steam or electricity generation.

Among the three main reactors involved in the CDCL process, the reducer is the most challenging since it involves complex solid-solid reactions scheme between the Fe_2O_3 composite particles and pulverized coal. Therefore, the process testing revolves around the validation of the concept of the reducer.

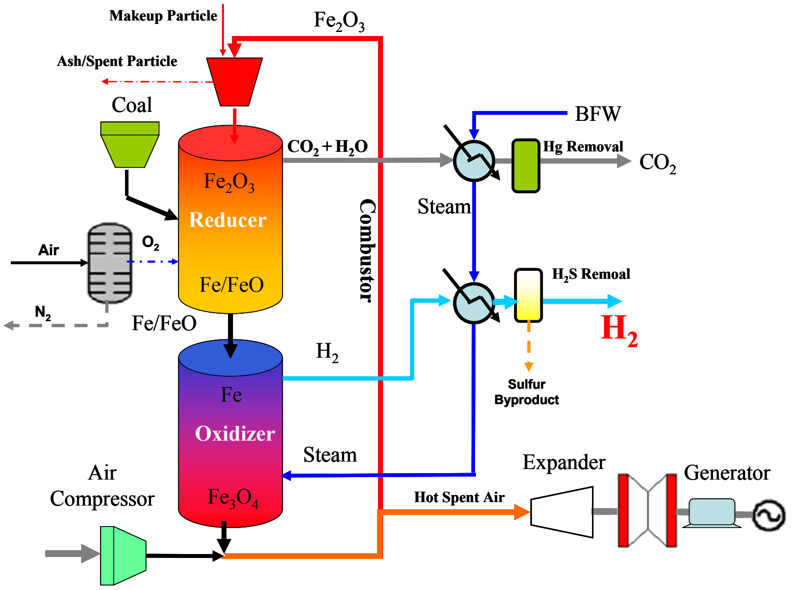

Figure 1 Schematic diagram of Coal-Direct Chemical Looping process

3. Experimental Setup

3.1 Reactor Setup

A bench scale demonstration unit for CDCL demonstration was designed and constructed at the Ohio State University. Figure 2 illustrates the schematic diagram of the unit. The unit was designed to handle solid flow rates up to 1.36 g/s (4.9 kg/hr) with gas flow rates up to 200 ml/s (12 l/min). The reacting section of the reactor has an I.D. of 1.6 inches and a height of 40 inches. In a typical demonstration, solid reactants such as the oxygen carrier particle and/or coal char are first loaded to the top funnel and then moved downwards steadily by the screw conveyor system. The gas composition along the axial positions of the reactor is constantly monitored using a Varian CP-4900 micro GC during the test and the solid can be taken from the solid sampling ports for further characterization after the experiment. Detailed information regarding the reactor setup is described in Gupta et al. (Gupta, Velazquez-Vargas et al. 2007)

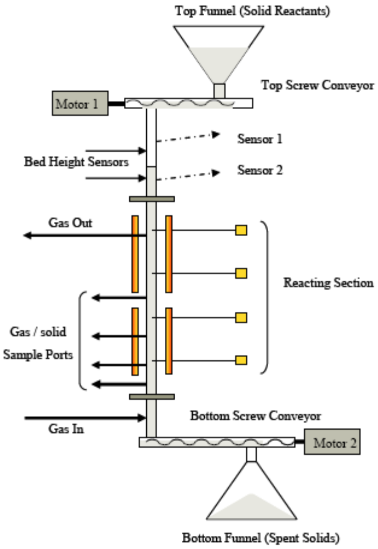

Figure 2. Schematic diagram of bench scale reactor

3.2 Experimental Procedures

Two experiments were carried out to demonstrate the feasibility of the proposed reducer configuration. The first experiment validates the conversion of methane, which represents one of the most stable form of coal volatile, using Fe_2O_3 composite particles. The second experiment validates the conversion of coal char with the presence of oxygen and Fe_2O_3 composite particles.

Before the methane experiment, the Fe_2O_3 composite particles were loaded in the top funnel. The particles were then moved downwards steadily by the screw conveyor system at a preset flow rate which is 31.5 g/min. Reactant gases were then introduced from the gas inlet located at the bottom of reactor. In the char conversion experiment, coal char and Fe_2O_3 particle mixture was loaded in the top funnel with oxygen introduced at a gas inlet located at the middle section of the reactor.

After the moving bed operation, the solid samples were taken from the various sampling ports for the further analysis using a carbon analyzer (CA) (UIC) and a thermogravimetic analyzer (TGA) (Perkin Elmer, Pyris 1). The carbon analyzer determines the carbon deposition and coal char conversion while the TGA verifies the conversion of particles through the weight change of the particle during air oxidation.

4. Results and Discussions

Figure 3 shows the gas conversions (red dotted line) and solid conversions (black solid line) at different ports.

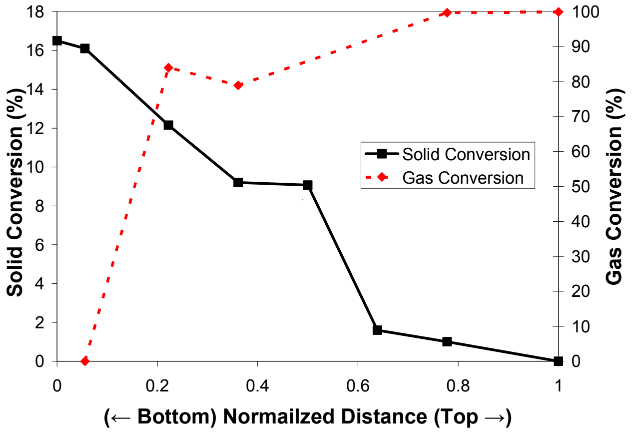

Figure 3. Gas and solid conversions at various solid sampling ports

As it is seen in Figure 3, around 16.5 % the solid conversion was achieved at the solid outlet whereas the upper parts of reactor observed less than 2% solid conversion. The solid conversion gradually increased as it approaches the bottom of the reactor. However, a methane conversion of nearly 100 % (99.89 %) was achieved at the gas outlet of the reactor, which corresponds to higher than 99% CO_2 concentration at the reactor outlet (dry basis). Figure 4 shows the gas concentrations at various gas sampling ports.

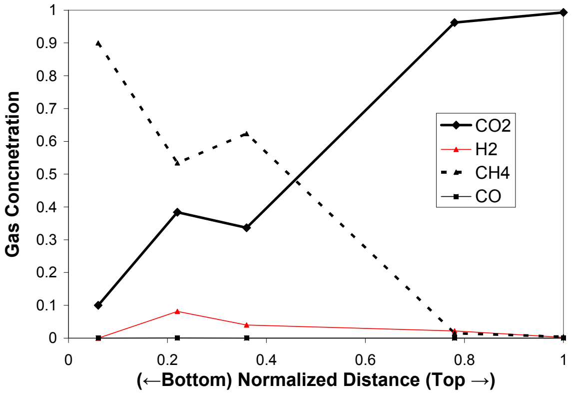

Figure 4. Gas concentrations at various gas sampling ports

The oxidation of coal char using the composite particles was also performed in the bench scale moving bed unit. The coal char obtained from bituminous (Pittsburgh #8) has been tested showing over 95% conversion as shown. The coal char conversion according to the normalized distance from the particle/coal mixture inlet is shown in Figure 5. Solid residence time of ~ 80 minutes was used.

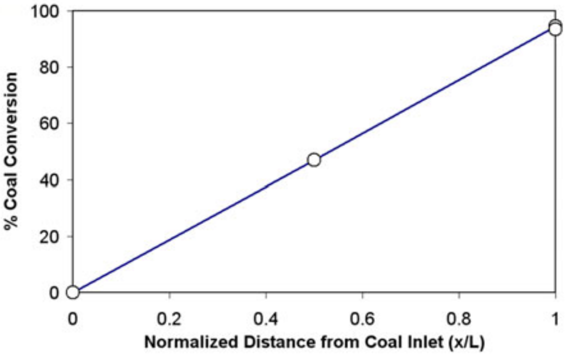

Figure 5. Coal char conversion in the presence of iron oxide composite particle and oxygen

The reduced Fe_2O_3 composite particles were further tested in an X-Ray Diffraction instrument (Scintag XDS2000) to determine the fate of sulfur. It was found that FeS was formed during the reducer operation. Further tests on the reactivity of the particle showed that the particle was fully recyclable after reacting with coal. More than 70% of the ash in coal was able to be separated from the particle using mechanical sieving method.

5. Conclusions

A novel coal direct chemical looping process that converts coal into hydrogen and/or electricity is discussed. Bench scale experiments show that the proposed reducer reactor is capable to convert around 99.9% coal volatiles and 95% bituminous coal char while reducing the Fe_2O_3 composite particles. At least a portion of the sulfur in coal was found to react with the Fe_2O_3 composite particle to form FeS. Moreover, ash can be separated from the particle using mechanical method. It is further determined that the Fe_2O_3 particle can be regenerated with air without losing its reactivity after reacting with coal. To conclude, the CDCO process concept is feasible according to preliminary testing results obtained from a bench scale units. Further improvements in coal char conversion, ash separation, and reduction in solid residence time are desirable.

6. Acknowledgement

This work has been supported by the Ohio Coal Development Office of the Ohio Air Quality Development Authority, U. S. Department of Energy, Ohio State University and an industrial consortium.

7. References

1. Fan, L.-S., P. Gupta, et al. (2007). Systems and methods of converting fuels. *PCT International Applications* WO 2007082089.
2. Gupta, P., L. G. Velazquez-Vargas, et al. (2004). Hydrogen production from combustion looping (solids-coal). *Proceedings of Clear Water Coal Conference*.
3. Gupta, P., L. G. Velazquez-Vargas, et al. (2007). "Moving bed reactor setup to study complex gas-solid reactions." *Review of Scientific Instruments* **78**(8)
4. Herzog, H.; Drake, E.; Adams, E. CO_2 Capture, Reuse, and Storage Technologies for Mitigating Global Climate Change. A White Paper; *U.S. Department of Energy*: Washington, D.C., January, 1997, U.S.DOE/DEAF22- 96PC01257
5. Ishida, M.; Jin, H. G., A New Advanced Power-Generation System Using Chemical-Looping Combustion. *Energy* (1994), 19, (4), 415-422.
6. Mattisson, T.; Jarders, A.; Lyngfelt, A. Reactivity of Some Metal Oxide Support on Alumina with Alternating Methane and Oxygen Application for Chemical-Looping Combustion. *Energy Fuels* 2003, 17, 643-651.
7. Thomas, T., L.-S. Fan, et al. (2004). Combustion looping using composite oxugen carriers. *US Patent Applications*. U.S. 11,010,648