Fuel 108 (2013) 370–384

Contents lists available at SciVerse ScienceDirect

Fuel

journal homepage: www.elsevier.com/locate/fuel

Coal direct chemical looping combustion process: Design and operation of a 25-kW_{th} sub-pilot unit

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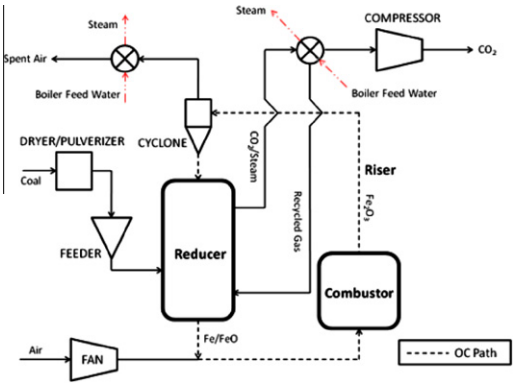
HIGHLIGHTS

- Successful long term (>230 h) demonstration of 25-kW_{th} CDCL sub-pilot unit.
- Demonstrated recyclability of oxygen carrier in the sub-pilot unit operation.
- 97% Sub-bituminous coal conversion.
- >99% CO_2 purity in reducer.
- No needs for solid recirculation device and additional carbon separation unit such as carbon stripper.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 26 August 2012
Received in revised form 9 December 2012
Accepted 10 December 2012
Available online 28 December 2012

Keywords:
Chemical looping
Coal
CO_2 capture
Moving bed
Pilot demonstration

ABSTRACT

The Coal-Direct Chemical Looping (CDCL) process using iron-based oxygen carriers has been developed as a coal conversion process with in situ CO_2 capture. The CDCL system cycles oxygen carriers between two reactors, a reducer reactor and a combustor reactor, in order to convert coal for electric power generation. The reducer reactor features a unique design of a gas–solid counter-current moving bed configuration to achieve the reduction of Fe_2O_3 particles to a mixture of Fe and FeO while converting the coal into CO_2 and steam. The combustor reactor is a fluidized bed that oxidizes the reduced particles back to Fe_2O_3 with air. The oxidation of iron is an exothermic reaction and the heat can be recovered for electricity generation. In the riser, the particles are pneumatically transported back to the reducer. An integrated 25-kW_{th} CDCL sub-pilot plant has been constructed and demonstrated, which is the first integrated chemical looping demonstration unit for the direct conversion of solid fuel with a circulating moving bed system. The design and operation experience of the 25 kW_{th} CDCL sub-pilot unit are reported in this paper. Specifically, the design criteria and operation conditions of the CDCL reactor system are first discussed which is followed by the construction of the sub-pilot unit. The tests with metallurgical coke and sub-bituminous coal resulted in 81% and 97% carbon conversions, respectively. Both tests yielded CO_2 purity greater than 99%, indicating the complete oxidation of volatile gases in the moving bed reducer. The gas analyses from the combustor outlet demonstrated a proper regeneration of oxygen carriers. A low CO_2 concentration in the combustor also confirms that there was no unconverted carbon transfer to the combustor and hence eliminated the need for an additional carbon separation device such as a carbon stripper. The demonstration results from the sub-pilot system substantiate the process concept of the CDCL, which is capable of processing coal continuously with reactor in situ CO_2 capture and the cyclic usage of oxygen carriers.

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http://dx.doi.org/10.1016/j.fuel.2012.12.038

Nomenclature

$A_{Reducer}$ cross-sectional area of reducer (m^2)
A_{Riser} cross-sectional area of riser (m^2)
C_i mole fraction of specific gas measured by the gas analyzer (–)
H_c higher heating value of coal (kJ/kg)
$\dot{m}_s$ solid circulation rate (kg/min)
$N_{C(Gasified)}$ gasified carbon including CO, CH_4 and CO_2 (mol/h)
N_C carbon flow rate: (Fuel flow rate) × (Carbon weight%) (mol/h)
$Q_{Carrier}$ normal gas flow rate of the carrier gas (L_n/min)
$Q_{Enhancer}$ normal gas flow rate of the enhancer gas (L_n/min)
$Q_{Combustor}$ gas flow rate in the combustor (L/min)
$Q_{out,Reducer}$ gas flow rate in the upper section of reducer (L/min)
$Q_{in,Reducer}$ gas flow rate in the lower section of reducer (L/min)
Q_{evol} gas flow rate of evolved gas from foal conversion in the reducer (L/min)
Q_{Riser} gas flow rate in the riser (L/min)
T_{red} reducer temperature (K)
$U_{mf,coal}$ minimum fluidization velocity of coal (m/s)
$U_{mf,OC}$ minimum fluidization velocity of oxygen carrier (m/s)
$U_{sg(out),Reducer}$ superficial gas velocity in the upper section of reducer (m/s)
$U_{sg(in),Reducer}$ superficial gas velocity in the lower section of reducer (m/s)
$U_{g,Riser}$ gas velocity in the riser (m/s)
$U_{t,OC}$ terminal velocity of oxygen carrier (m/s)
U_{OC} velocity of oxygen carrier in the riser (m/s)
$V_{combustor}$ design volume of the combustor (m^3)
V_n normal molar volume of an ideal gas at 0 °C and 1 bar of pressure (L_n/mol)
X carbon conversion in reducer
x_C elemental carbon fraction in coal (dry basis) (–)
x_H elemental hydrogen fraction in coal (dry basis) (–)
x_{H2O} coal moisture fraction (–)
x_O elemental oxygen fraction in coal (dry basis) (–)
Y_i normalized gas fraction in the reducer gas outlet (–)
α_s volume fraction of oxygen carrier in the combustor (–)
$\Theta_{Combustor}$ air demand of oxygen carrier for regeneration (L_n/min)
ρ_s particle density (kg/m^3)
$\tau_{R,Combustor}$ residence time of oxygen carrier in the combustor (min)
φ_c thermal operating capacity (kW_t)

1. Introduction

According to the Annual Energy Outlook 2011 by Energy Information Administration (EIA), coal is responsible for more than 40% of the global electricity generation and will remain a key fuel for base load power plants through 2035 [1]. Furthermore, in the electricity sector, coal-fired power plants contribute the largest pollutant emissions. Though the coal industry has made significant progress in reducing flue gas emissions such as sulfur oxides (SO_x), nitrogen oxides (NO_x), particulate matter (PM) and mercury (Hg), coal-fired power plants lack emissions control for carbon dioxide (CO_2), the most contributing greenhouse gas, associated with climate change. In order to control CO_2 emissions, a number of carbon capture technologies have been developed for retrofitting/repowering existing coal-fired power plants and for building a new generation of combustion systems. However, incorporating carbon capture technologies into power plants is generally associated with high capital and operating costs and large energy penalties. For example, the post-combustion carbon capture with a monoethanolamine (MEA) scrubber from the flue gas of a conventional pulverized coal (PC) power plant results in a 40–95% increase in cost of electricity (COE) due to high capital and operating costs and an approximately 10% decrease in plant efficiency [2–5]. An integrated gasification combined cycle (IGCC) for pre-combustion carbon capture is capable of achieving a lower decrease in plant efficiency than MEA process, between 5% and 9%, but the capital intensive gasifier and air separation unit (ASU), with costs reaching up to ~$2200/kW, poses a challenge to its commercial development [2,3].

Distinct from the pre- and post-combustion carbon capture processes, oxy-combustion technologies enable nearly 100% CO_2 capture by firing coal with nitrogen-free oxygen sources. The oxygen supply is provided either from an ASU, an oxygen transport membrane (OTM) or an oxygen carrier material in a chemical looping combustion (CLC) system. The ambient oxy-combustion with the ASU can increase the COE by nearly 46% [6]. Stand-alone OTM technologies can generate oxygen at a higher efficiency than can an ASU, but OTMs consume a large parasitic energy load due to an energy intensive air compression step. Alternatively, integrating the OTM with a coal boiler provides a better solution for the heat integration, however, the driving force between air and coal is lower as compared to the driving force required for a gas-fired OTM. Previous energy and exergy analyses have been conducted to compare a pressurized oxy-coal fluidized bed combustion (oxy-PFBC) process with a CLC process. The exergy loss for an oxy-PFBC is 37%, while a pressurized CLC process has an exergy loss of only 11.6%, demonstrating that CLC is significantly more efficient [7]. Therefore, the chemical looping technology can be an alternative approach to oxy-combustion.

The chemical looping technology indirectly combusts the fuel source with oxygen provided from the oxygen carrier to produce a sequestration-ready stream of CO_2 while generating heat for electricity production. The reaction path scheme separates the air from the fuel source and achieves in situ CO_2 capture, eliminating the need for the additional CO_2 separation unit. For the chemical looping concept, coal is preferred because it is an abundant, inexpensive and stable source of fuel. One way to utilize coal in chemical looping processes is through partial oxidation to syngas via a coal gasifier [8–10]. However, the coal gasifier operation requires the energy- and capital-intensive ASU. Thus, the direct utilization of coal in chemical looping processes has the potential to be a highly cost and energy effective carbon capture technology [8–15].

Over the last decade, the chemical looping technology has shown significant advancement from its concept to lab and bench scale testing (<5 kW_{th}), sub-pilot scale testing (5–50 kW_{th}), and pilot scale testing (50 kW_{th}–5 MW_{th}) [16–18]. Recently, many researchers have started to investigate the direct conversion of coal in the CLC process with various oxygen carrier types and reactor designs [19–28]. A 3-MW_{th} dual circulating fluidized bed CLC pilot process utilizing a $CaSO_4$ oxygen carrier is currently under demonstration by Alstom Power Inc. for the conversion of coal [19,20]. Further, ALSTOM is also developing a 1-MW_{th} CLC process in Europe under the Emission Free Chemical Looping Coal Combustion Process (ECLAIR). This process employs a fluidized bed reactor design with naturally occurring ilmenite and composite copper oxide particles as the oxygen carriers [21]. The University of Utah is developing a fluidized bed CLC process utilizing copper-based oxygen carriers [22,23]. Processes utilizing copper-based oxygen