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An investigation of steam production in chemical-looping combustion (CLC) and chemical-looping with oxygen uncoupling (CLOU) for solid fuels

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ABSTRACT

Chemical-looping combustion (CLC) and chemical-looping with oxygen uncoupling (CLOU) are being actively explored as solid fuel combustion technologies that have the potential to facilitate CO_2 capture. While CLC and CLOU have similarities operationally, there are some key differences. In particular, the CLC process requires a coal gasification step where coal is first broken down into a syngas with the use of steam or CO_2. The resulting syngas is then oxidized with the metal oxide to release energy. In the CLOU process the metal oxide releases oxygen that combusts the solid fuel, resulting in a lower residence time, as the coal gasification reactions are avoided. The CLC and CLOU systems were modeled with ASPEN Plus at a 10 MW_{th} scale, and the process streams were analyzed by ASPEN Energy Analyzer to determine the amount of industrial process steam that could be generated from CLC or CLOU. Both the air and fuel reactor were analyzed as two circulating fluidized beds, with metal oxide circulating between the two reactors. The air reactor, where metal oxide is oxidized, was fluidized with air. The fuel reactor, where the metal oxide is reduced, was fluidized with steam for CLC and recirculated CO_2 for CLOU. It was identified that the CLOU process had the potential to produce more steam, approximately 7920 kg/h, as compared to CLC (6910 kg/h).

1. Introduction

Recent interest in controlling CO_2 emissions has motivated research in carbon capture technologies for solid fuel combustion. Chemical-looping combustion (CLC) has emerged as a promising alternative technology for CO_2 capture. Research has been carried out in 0.3–140 kW_{th} pilot units (Lyngfelt, 2011) and 1–3 MW_{th} demonstration units are currently under construction (Lyngfelt, 2014). A typical CLC process consists of two interconnected fluidized beds which serve as a fuel and an air reactor. In the fuel reactor, the fuel is combusted with oxygen supplied by a circulating oxygen carrier, typically a metal oxide. The reduced oxygen carrier is then transported to the other reactor, the air reactor, where the reduced oxygen carrier is oxidized by air. The regenerated oxygen carrier is then carried back to the fuel reactor.

While chemical-looping combustion (CLC) and chemical-looping with oxygen uncoupling (CLOU) are very similar in configuration, they differ in the mechanism by which oxygen is accessed by the solid fuel (Mattisson, 2013). In the case of CLC the coal is gasified with steam, forming a syngas. The resulting syngas is then oxidized with the circulating oxygen

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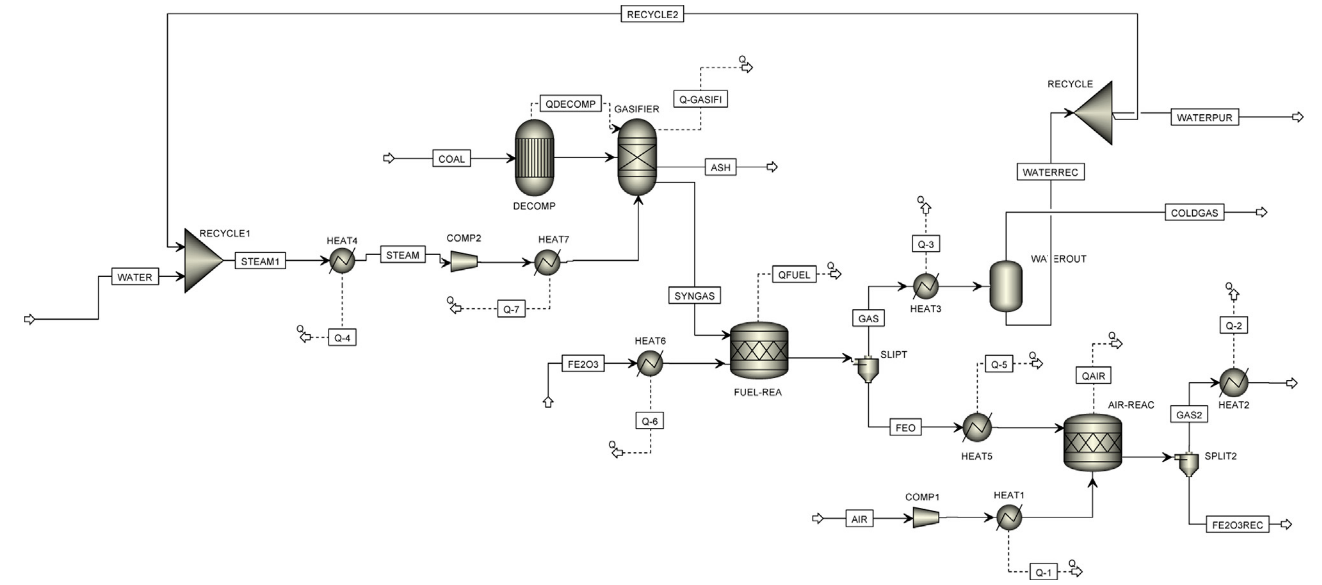

Fig. 1 – ASPEN Plus flow sheet of CLC process model.

carrier to form CO_2 and H_2O, accompanied with the release of energy. In the case of CLOU the oxygen carrier releases oxygen, which then combusts the coal. In CLOU the gasification step is avoided, resulting in a faster reaction time. A variety of materials have been explored for CLC processes and the CLOU process (Lyngfelt, 2011; Adanez et al., 2012; Mattisson, 2013; Imtiaz et al., 2013). In this particular study, the CLC process has been explored with an iron-based oxygen carrier Fe_2O_3 that releases oxygen by transitioning to Fe_3O_4. The CLOU process has been modeled using a copper-based oxygen carrier transitioning from CuO to Cu_2O (Sahir et al., 2014).

2. Overview of the CLC and CLOU process models

In this study, CLC and CLOU were modeled with ASPEN Plus to determine the material and energy balances (Sahir et al., 2014). The process stream data from those balances was then used in ASPEN Energy Analyzer to determine the steam production rate for each scenario. Wyoming Powder River Basin (PRB) coal was chosen as the feedstock basis, which is being utilized for a process development unit being built at the University of Utah (Lighty, 2012; Whitty, 2012). Specifications are given below and details can be found in Sahir et al. (2014).

2.1. *CLC ASPEN Plus process model*

The CLC ASPEN Plus model employed 60% Fe_2O_3 supported on Al_2O_3 oxygen carrier particles with a particle diameter of 150 μm. Within the CLC fuel reactor the following reactions take place:

$$3Fe_2O_3(s) + CO(g) \rightarrow 2Fe_3O_4(s) + CO_2(g) \quad (1)$$

$$3Fe_2O_3(s) + H_2(g) \rightarrow 2Fe_3O_4(s) + H_2O(g) \quad (2)$$

In the air reactor the oxygen carrier is oxidized by the following reaction:

$$4Fe_3O_4(s) + O_2(g) \rightarrow 6Fe_2O_3(s) \quad (3)$$

These aspects have been incorporated in the development of a process flow sheet shown in Fig. 1 (Sahir et al., 2014), and whose model blocks are explained below:

As mentioned in Sahir et al. (2014) the fuel reactor had three blocks (RGIBBS, RSTOIC, and RYIELD). While these are three separate blocks in the ASPEN Plus process model, in reality these reactions occur in a single fuel reactor representing a direct CLC process. The RYIELD block breaks the coal down into its constituent elements. The effluent stream from the RYIELD block is then fed into the RGIBBS reactor with the required steam, which generates the syngas. In the ASPEN process model the water consumption was based on an equal amount of carbon moles to water moles to affect the reaction $C_{(s)} + H_2O \rightarrow CO_{(g)} + H_{2(g)}$. The water requirements to produce steam for fluidizing the fuel reactor were 6.9 times larger than that required for the reaction. These were determined based on circulating fluidized bed calculations from Basu and Fraser (1991) and Lyngfelt et al. (2001). The reaction of the metal oxide particles with the syngas, represented by Reactions (1) and (2), was modeled in a RSTOIC block with 99.9% conversion of Fe_2O_3 to Fe_3O_4. Due to the endothermic nature of gasification the air reactor was operated at a higher temperature, 1050 °C, so that the oxygen carrier could transfer energy for the fuel reactor operation, at 975 °C, to maintain the gasification reaction in an autothermal operational mode. The residence time for the fuel reactor was 10 min, controlled by the char gasification.

The effluent gas/solid stream was then sent to a cyclone to separate out the solid particles from the gas. The gas was then cooled, and energy was recovered. Water was recycled to generate the steam for fluidization of the fuel reactor; a flash was used for this separation. The gas fed to the flash was cooled to 85 °C in order to condense the water from the other gases.

The reduced oxygen carrier particles were then sent to the air reactor; Fe_3O_4 was oxidized (Reaction (3)) with a stoichiometric conversion of 80%. Energy was recovered from the effluent gas of the air reactor. The regenerated oxygen carrier particles were separated with a cyclone and sent back to the fuel reactor. Air reactor residence time was 90 s.