Fuel Processing Technology 156 (2016) 124–137

Contents lists available at ScienceDirect

Fuel Processing Technology

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

Chemical-looping combustion with heavy liquid fuels in a 10 kW pilot plant

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ARTICLE INFO

Article history:
Received 8 August 2016
Received in revised form 18 October 2016
Accepted 24 October 2016
Available online xxxx

Keywords:
Chemical-looping combustion (CLC)
Liquid fuel
Vacuum residue
Calcium manganite
Ilmenite
CO_2 capture
Carbon capture and storage (CCS)

ABSTRACT

In this study, chemical-looping combustion was performed with highly viscous vacuum residue. A fuel reactor with a fuel-injection system for liquid fuels was designed and built for a chemical-looping reactor with the nominal fuel input of 10 kW_{th}. The gas velocities in the riser section and at the gas-distribution nozzles of this unit are comparable to those of industrial circulating fluidized-bed boilers. Reference experiments were performed with an ilmenite oxygen carrier and two different fuel blends that contained 40 wt.% and respectively 80 wt.% of vacuum residue in fuel oil 1. Fuel conversion was in line with that of experiments from an earlier campaign, where fuel oil 1 was used as fuel. The fuel contained a significant fraction of sulfur, but no SO_2 was detected in the flue gas of the air reactor.
More experiments were performed using an oxygen carrier based on calcium manganite. The oxygen carrier was exposed to fluidization at hot conditions (more than 600°C) for about 95 h, out of which fuel was injected during a total of 9.6 h. Undiluted vacuum residue, fuel oil 1 as well as different blends of these two were used as fuel. Furthermore, the parameters fuel flow, fuel-reactor temperature and air flow in the air reactor were varied to observe trends in fuel conversion. The experiments were carried out with a fuel flow corresponding to 4.0–6.2 kW_{th} and an oxygen carrier-to-fuel ratio of about 1300–2000 kg/MW_{th} (fuel-reactor bed mass per thermal fuel-power). With undiluted vacuum residue as fuel and a fuel-reactor temperature of 1000 °C, up to 93% of all carbon leaving the fuel reactor was in the form of CO_2. Carbon leakage from fuel reactor to air reactor was usually below 1% for all fuel types tested, but no SO_2 was detected in the off-gas from the air reactor. The reactivity of the calcium manganite-based material decreased over the course of the experiments, which is likely due to sulfur poisoning. No defluidization or agglomeration problems were experienced over the course of the experimental campaign.

1. Introduction

1.1. Chemical-looping combustion

Chemical-looping combustion (CLC) is a method of using carbon-based fuels for production of heat with inherent separation of CO_2. CLC can be part of a CCS (carbon capture and storage) scheme with the purpose of reducing the impact of carbon dioxide from carbon-based fuels on the climate of the Earth. Continued utilization of fossil fuels at a large scale will require implementation of CCS if the UN and EU endorsed temperature limits are to be met [1,2].

In chemical-looping combustion, an active bed material, the oxygen carrier, is cyclically exposed to oxidizing and reducing atmospheres, respectively air and fuel. While in air, the oxygen carrier takes up oxygen and subsequently passes it to the fuel. This can be achieved by circulating the oxygen-carrier particles continuously between two fluidized-bed reactors. The net reaction of this process is the same as for combustion in air, i.e., the heat produced in chemical-looping combustion is the same as in combustion in air. The difference is that in chemical-looping combustion the fuel is never mixed with nitrogen from the air, so that the off-gas from the fuel reactor ideally consists of only carbon dioxide and steam. The latter can be condensed and the sequestration of CO_2 does not require any input of energy. Recent literature concerning developments, advancements and operational experience in different chemical-looping applications has been published by Adánez et al. [3], Lyngfelt [4] and Nandy et al. [5].

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http://dx.doi.org/10.1016/j.fuproc.2016.10.027

1.2. Liquid fuels in chemical-looping combustion

The use of liquid fuel in stationary combustion processes is far less common than the use of coal or gas. Because of their high energy density, liquid fuels are mostly used in transport applications. In an oil refining process, a large variety of end products can be generated, which differ greatly in quality and price. The bottom residues of a vacuum distillation process, also called vacuum residues, are upgraded if economically feasible. Such low-grade heavy-oil products could pose an interesting and feasible option for thermal processes, especially if the process is located close to or integrated in a refinery process, where there is a constant need for heat and steam. Difficulties in the use of heavy-oil products are related to their high viscosity and to high amounts of sulfur and other impurities, such as heavy metals. A recent overview about chemical-looping combustion of liquid fuels has been published by Rydén [6].

2. Experimental details

2.1. 10 kW chemical-looping combustion reactor system

Fig. 1 shows a schematic of the reactor system with all major sections shown, including gas and fuel inlets and gas exits. The unit is based on interconnected fluidized beds and is similar to the design originally presented by Lyngfelt et al. [7]. In the riser section, there is a fast-fluidized regime, whereas in the loop-seals and the fuel reactor there is a bubbling regime. In the air reactor the fluidization regime is on the transition between bubbling and fast fluidized. The unit was heated externally to maintain sufficient temperature in the system and compensate for radiation losses. Such losses are inherent in small-scale units like the one used here, i.e., such with a high ratio of surface area to volume. Additionally, the air that is fed to the air reactor is preheated to 1000°C. The heating cables are wrapped around the riser section, the cyclone, the upper loop-seal and the fuel reactor, and covered with insulation material. More details about the reactor system can be found elsewhere [8].

The lower section of the fuel reactor was specifically designed for direct injection of liquid fuel, see Fig. 2. The height of this section is about 165 mm and the bed diameter is 150 mm. The total bed height in the fuel reactor is 275 mm. Steam is used to fluidize the oxygen-carrier particles at a superficial gas velocity that corresponds to about 7–20 times the minimum fluidization velocity, u_{mf}. Fuel is introduced via an exchangeable injection nozzle with an orifice diameter of 0.25 mm. The injection nozzle and the fuel jet are surrounded by a flow of steam, which has the purpose of cooling injection system and fuel jet as well as preventing oxygen-carrier particles from entering the injection system. To reduce heat transfer from the hot particle bed to the injection system, the injection nozzle is located as far away as possible from the bed, while being as close as necessary for the fuel jet to reach the bed. In the fluidized bed reactor, 31 rods with a diameter of 6 mm are arranged in an equidistant pattern, which are referred to as internals. These internals are used to limit the growth of gas bubbles and to reduce the risk of formation of gas channels, which would reduce the fuel–oxygen carrier contact and therefore decrease fuel conversion. The projected open area that results from the pattern of the internals corresponds to 54% of the total cross-sectional area of the fuel reactor. A plate with 55 holes, which have a diameter of 0.5 mm an are arranged in an equidistant pattern, acts as gas distributor in the bottom of the fuel reactor. The porous quartz, which is located below the hole plate, was intended to be used as gas distributor in an earlier design phase, but became redundant when the hole plate was introduced.

The amount of oxygen carrier used in the 10 kW unit depends on the bulk density of the oxygen-carrier particles. In the configuration used for injection of liquid fuel, the bed mass is in the range of 15–25 kg, out of which 5–10 kg are in the fuel reactor.

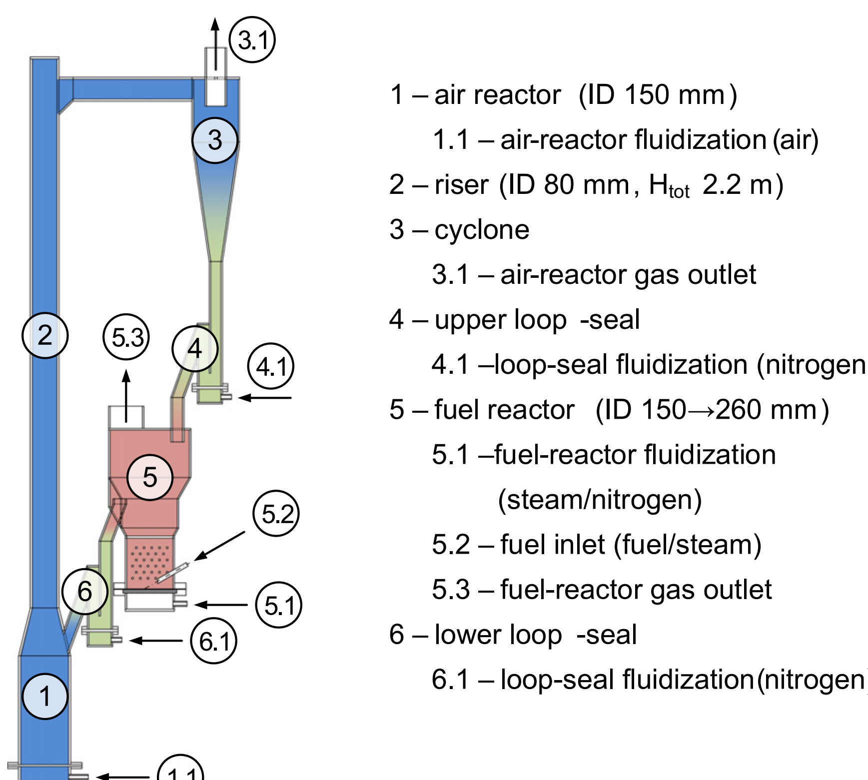

Fig. 1. Schematic of the 10kW chemical-looping combustion reactor.