Particle Size Optimization for Syngas Chemical Looping Process

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Abstract

The syngas chemical looping (SCL) process developed at the Ohio State University can efficiently convert coal derived syngas into hydrogen with in-situ CO_2 capture. In this novel process, two moving bed units are used to convert syngas into hydrogen through the assistance of an oxygen carrier particle. A third unit, an entrained bed combustor, is used to convey the oxygen carrier particles to complete the redox (reduction-oxidation) cycle. The reaction kinetics and thermodynamics of the combustor are well understood. Major challenges for the entrained bed design lie in the clear understanding of the hydrodynamics as well as the optimization of the oxygen carrier particle size. A desirable particle should be easy to convey in the third reactor while not being fluidized in the two moving bed units.

In this paper, a systematic approach is adopted to study the entrainment behavior of the oxygen carrier particles. The configurations and operating conditions for all three reactors in the SCL process are considered. A set of particle sizes are first determined based on theoretical estimation and then tested in the sub-pilot scale SCL demonstration reactor constructed at OSU. The hydrodynamics of particles conveying in the entrained bed reactor are studied. The required conveying gas velocities for the particles are then determined using the entrained bed combustor. The optimized particle size range for the operation of all three reactors is then determined. Key information for the SCL reactor demonstration such as particle attrition is also obtained.

Introduction

Energy is of great importance to the modern economy. Although renewable energy sources such as solar, wind, hydro, and biomass have been widely used, their contribution to the overall energy demands can be limited within the foreseeable future due to higher costs, geological constraints, and intermittent issues. Despite high crude oil and natural gas prices, fossil fuels is expected to provide a major portion of the overall world energy consumption in the next several decades [1, 2]. Abundantly available throughout the world, coal can play a more important role in the overall energy mix, especially when the crude and natural gas prices are volatile.

In this paper, a novel syngas chemical looping (SCL) process is studied. The SCL process converts coal derived syngas into hydrogen, which is an important feedstock for oil refining, fertilizer production, and liquid fuel synthesis. The process consists of three main units: (1) a moving bed reducer, (2) a moving bed oxidizer; and (3) an entrained bed combustor. The two moving bed units convert syngas into hydrogen while the entrained bed combustor conveys the iron based oxygen carrier particles to complete the redox (reduction-oxidation) cycle. The reaction kinetics and thermodynamics in the combustor are well understood [3]. The major challenge for the combustor design resides in the in-depth understanding to the hydrodynamic properties of the oxygen carrier particles. In this paper, the

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pneumatic conveying properties and attrition rates of the particle obtained from a sub-pilot scale SCL unit are discussed. Such information can help determine important SCL process operation parameters such as optimum particle size, shape, and fresh particle makeup rate.

Experimental Setup

Figure 1 is the schematic diagram of the sub-pilot scale SCL demonstration unit located at OSU west campus SCL demonstration lab. Figure 2 is the photography of the demonstration unit. The demonstration unit consists of three main reactors: a reducer (heating section A), an oxidizer (heating section B), and a combustor (conveying system). In the SCL reactor operation, oxygen carrier particles fed from the top of the reducer moves through the reducer and the oxidizer by gravity. Valve systems and rotary solid feeder assemblies are used to assist solid movement while avoiding gas leakage. After being discharged from the oxidizer outlet, the partially regenerated oxygen carrier will be introduced to the combustor through a slant stainless steel tube and then conveyed vertically back to the reducer inlet with air to complete the redox/solid conveying cycle. The combustor, air inlet, and slant tube are connected by a "Y" shaped tube. A programmed gate valve is mounted on the slant pipe to control the particle feed to the vertical section of the combustor. A distributor made of Hastelloy X is installed at the bottom of the combustor to ensure even distribution of the air. The main section of the combustor is a vertical pipe with a 2 in ID and a height of 20 ft. A 90^0 elbow and a horizontal pipe are connected to the top of the vertical pipe with a cyclone connected on the other end of the horizontal pipe. The cyclone allows the separation of the fines and ensures that larger particles will be re-introduced to the reducer. All the parts for the combustor are made of 304L stainless steel. An air compressor, with a capacity of ~30 l/s at 180 psig, is connected to the bottom of the distributor to provide air for both particle conveying and oxygen carrier combustion purposes.

During the reactor operation, particles are loaded into the slant pipe from the bottom of the oxidizer with the programmed ball valve. After the particles in the declined pipe are accumulated to a preset amount, all the particles are dumped into the bottom section of the vertical combustor by opening the programmed gate valve for short period of time (< 10s). After the programmed ball valve is closed, the air source from the compressor is introduced into the combustor to convey all the particles at the bottom of the vertical pipe into the reducer inlet. Exhaust air coming out of the cyclone will then be introduced to a powder separator for the separation of fine powders before ventilation. The powder separator is not shown in Figure 1.

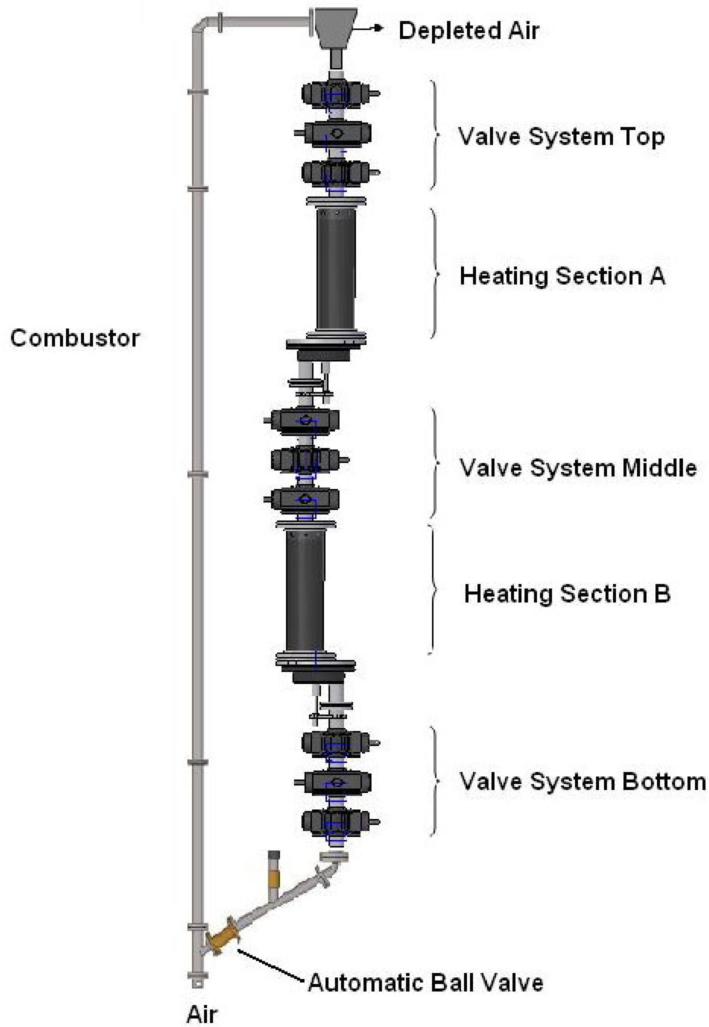

Figure 1. Schematic diagram of the units for the SCL process

Figure 2. Picture of the actual units for the SCL process in west campus SCL demonstration lab at OSU

Results and Discussion

Pneumatic Conveying of Particles

The pneumatic conveying system consists three parts: (1) vertical flow through the combustor pipe, (2) change in direction through a 90^0 elbow, and (3) horizontal flow. The flow pattern in the gas-solid pipe is a dilute suspension flow with a very low particle concentration. Several sets of particles are selected to test the combustor operation. The particles are pellets processed from iron oxide and inert support with a density of $2500\, kg / m^3$. A 3kW rotary pelletizer (ZP-35) is used to fabricate pellets with a production rate of up to 15 kg/hour. The pellets have a cylindrical shape with a diameter of 5 mm and a thickness of 1-5 mm depending on the setting of the pelletizer. Before testing, the pellets are sintered at 900 ^{0}C for more than 20 hours to achieve better physical and chemical properties.

It was determined through experiments that the terminal velocity for the pellets with a diameter of 5 mm and a thickness of 1.5 mm, or type A particles, is around 9.3 m/s. For this type of particles, the pneumatic conveying system needs to be operated at a gas velocity ~1 m/s higher than its terminal velocity since horizontal pneumatic transport requires higher gas velocity than vertical pneumatic transport. This due mainly to the higher drag forces and the various frictions in the horizontal conveying step [4]. However, the effects of the drag forces and the frictions in the horizontal pneumatic transport are not significant since the length of the horizontal part is relatively short. It was also determined that the terminal velocity for the pellets with a diameter of 5 mm and a thickness of 4.5 mm, or type B particles, is around 15 m/s. For type B particles, the pneumatic conveying system can be operated at a gas velocity slightly higher than its terminal velocity due to the better flow properties of the particle in the horizontal pipe. The higher terminal velocity and better flow properties of type B particles can be explained by it near sphere shape which allows for a higher fluidity performance, lower effects of drag forces, and less friction than type A particles.

Particle Attrition

In order to reduce the particle purging rate, it is desirable for the oxygen carrier particles to maintain its chemical reactivity and physical integrity for multiple cycles. Since particles are in vigorous motion in the combustor, the attrition rate of the particle is an important parameter for the SCL combustor design and particle optimizations. Particle attrition affects the flow performance and economics of the SCL process. A low attrition rate of the particles is highly desirable. Due to the turbulent movements at high gas velocities, the frequent particle-particle/ particle-wall collisions during the pneumatic conveying step in the combustor and the particle separation step in the cyclone play an important role for particle attrition, which is defined as the degradation of particles due to mechanical stress [5]. Abrasion and fragmentation of the particles are the main modes of the attrition [6, 7]. The attrition effects due to the combustor and the cyclone are measured in the SCL process. For the attrition test in combustor, particles are loaded at the bottom of the vertical pipe and then conveyed out of the horizontal part of the combustor. These particles are subsequently collected using a bucket. The particles are then sieved and weighted according to different size fractions. All the particles are then well mixed and reloaded into the combustor for the next conveying cycle. To test the attrition in both the combustor and the cyclone, a similar testing procedure is adopted. The only difference is that the particles are collected after the cyclone. The attrition rates for both type A and type B particles were test with 20 runs.

Figure 3 and Figure 4 shows the combustor attrition test results for type A particles and type B particles respectively. A superficial gas velocity of 10 m/s is used to convey the type A particles. From Figure 3, the particle attrition rate is 0.56% per cycle when the cutoff is set to be 600 μm, i.e., particles smaller than 600 μm will be purged out from the system. A superficial gas velocity of 15 m/s is used to convey the type B particles. From Figure 4, the particle attrition rate is 0.23% per cycle when the cutoff is set to be 600 μm. The particle attrition rate is around 0.38% per cycle when the cyclone is used for the test at the same experimental condition, which means an additional attrition of 0.15% was introduced due to the addition of the cyclone. The type B particles with 5 mm diameter and 4.5 mm thickness perform better on attrition than type A particles due to its close-to-spherical shape. Such a shape leads to reduced effects of the drag forces and the various particle-particle and particle-wall frictions. As can be seen, both type A and type B particles possess excellent physical strength.

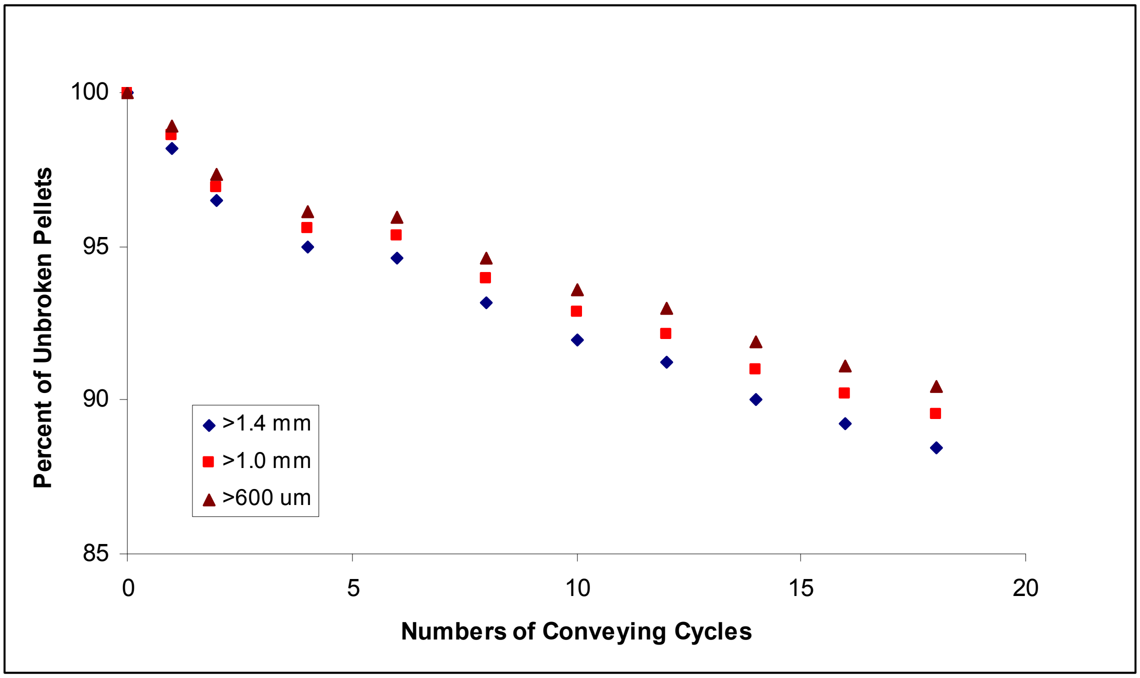

Figure 3. Attrition test results using Type A particles with 5 mm diameter and 1.5 mm thickness

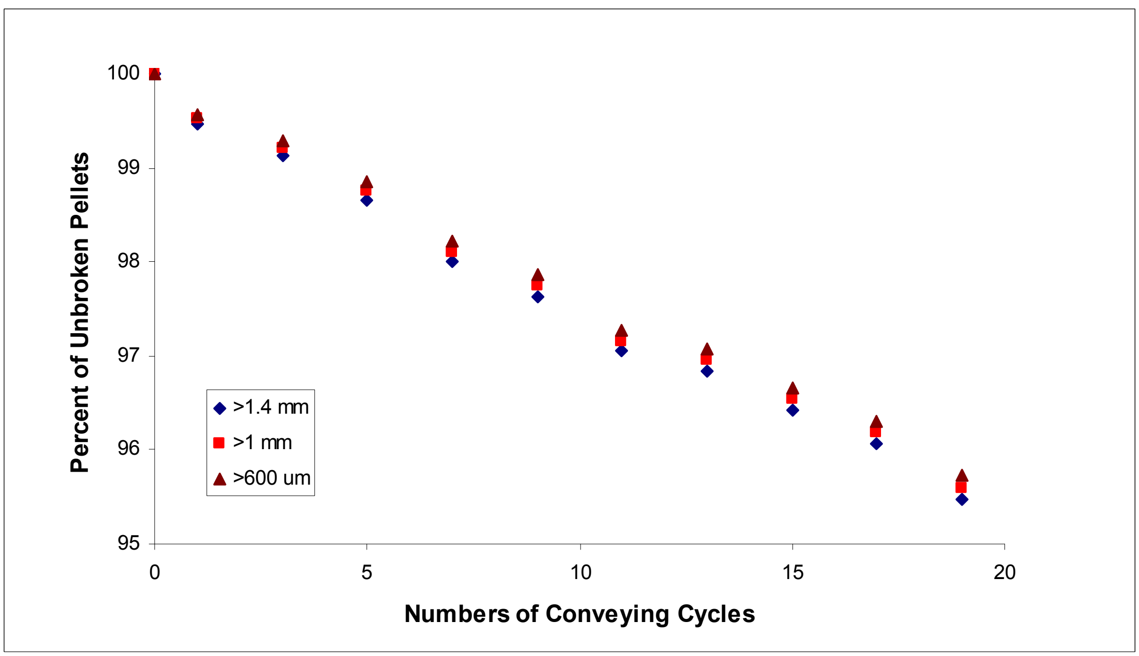

Figure 4. Attrition test results using Type B particles with 5 mm diameter and 4.5 mm thickness

Conclusion

The syngas chemical looping (SCL) process developed at the Ohio State University can convert coal derived syngas into hydrogen with in-situ CO_2 capture. Two sets of particles with different sizes are selected and tested in the entrained bed reactor constructed at OSU. The hydrodynamics of particles conveying in the entrained bed reactor are studied. It was found that the particles can be pneumatically conveyed at a linear gas velocity of 10 m/s and 15 m/s respectively. Particles with 5 mm diameter and 4.5 mm thickness show much lower attrition than particles with 5 mm diameter and 1.5 mm thickness. Both types of particles possess excellent physical strength and can potentially be recycled for multiple cycles without losing its physical integrity.

Acknowledgements

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