Transient 1-d model simulation of a packed-beds chemical-looping combustion reactor of syngas with ilmenite

MODEL SIMULATIONS OF A PACKED-BED CHEMICAL-LOOPING COMBUSTION REACTOR OF SYNGAS

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Summary

A packed-bed chemical-looping combustion (CLC) reactor is developed using syngas as fuel with ilmenite as an oxygen carrier. The aim of the current study is to analyze, design and optimize the performance of this novel reactor concept using numerical simulations. Transient 1-D reactor models of the fuel and air reactors have been developed to investigate the dynamic temperature and concentration profiles along the length of the reactor. Simulation studies show that a very high temperature air stream can be produced efficiently with packed-bed CLC using syngas combustion with ilmenite with intrinsic capture of CO_2. On the other hand, due to the relatively low reduction rates, much more attention has to be paid to fuel slip during the reduction cycle.

Keywords: Chemical-looping combustion (CLC), Ilmenite, syngas, Modeling and simulation

Introduction

Chemical-looping combustion (CLC) has proven to be an attractive alternative for conventional combustion technology to integrate the power production with inherent capture of CO_2. Traditionally CLC has been studied in fluidized beds. Recently, our group has demontrated the possibility to carry out the CLC of methane in dynamically operated packed beds [1]. In this concept, the oxygen carrier (OC) particles are alternately exposed in the fuel and air reactors with syngas and air as feed gas respectively. The exhaust gas from the fuel reactor contains only CO_2 and H_2O while the flue gas from the air reactor contains high temperature N_2 and some un-reacted O_2 stream. Thus, CO_2 and N_2 are inherently separated [1]. In this study, we analyze the use of ilmenite as possible oxygen carrier in CLC with syngas (derived by coal gasification) via detailed particle and reactor models. The models are used to design a lab-scale reactor for the proof-of-principle of CLC of syngas with packed-bed technology with ilmenite as OC.

Theory and principle

Ilmenite has been used as oxygen carrier material where hematite (Fe_2O_3) and rutile (TiO_2) are assumed to be the active metal oxide and the inert support, repectively. The hematite in the combustion process with syngas, participates in both reduction and oxidation reactions by the following general reactions;

$$Fe_2O_3 + CO \rightarrow 2FeO + CO_2 \qquad \Delta H > 0$$
$$Fe_2O_3 + H_2 \rightarrow 2FeO + H_2O \qquad \Delta H < 0$$
$$4FeO + O_2 \rightarrow 2Fe_2O_3 \qquad \Delta H > 0$$

In the packed-beds CLC reactor (see Figure 1), the oxygen carrier particles are alternately exposed to:

Reduction Step: Oxygen carrier is reduced with syngas combustion and CO_2 is captured during the reduction cycle.
Oxidation step: Oxygen carrier is regenerated and high temperature depleted air is produced for power production during the oxidation cycle.

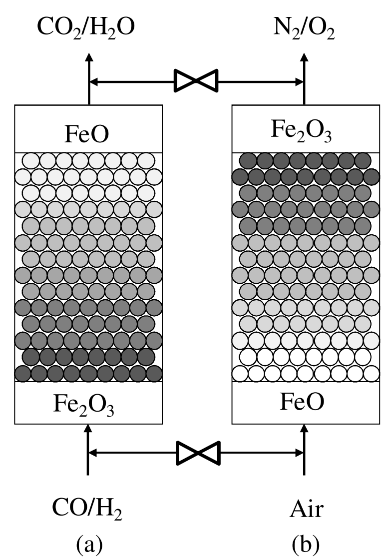

Figure 1. Schematic view of the CLC process principle; (a) reduction step (b) oxidation step.

Base case simulations: Effects

In the current study, a detailed reactor model is used for the combustion of syngas using ilmenite as oxygen carrier. It is assumed that the ilmenite is activated so that hematite (Fe_2O_3) and wustite (FeO) are considered to be the most oxidised and reduced form of ilmenite, respectively, together with rutile (TiO_2). Furthermore, the reaction kinetics have been taken from literature [2] and the initial bed temperature has been taken the same as the feed gas temperature. The reactor model has been used for the design, optimization and scale-up of a pilot scale packed-bed CLC reactor.

The effect of the reactor bed and feed gas temperature, the active content of ilmenite in the oxygen carrier, the conversion of syngas and transient axial temperature and concentration profiles during both oxidation and reduction steps have been theoretically investigated.

It is shown in Figure 2 that an increase in mass fraction of the active content in the oxygen carrier (*i.e.* hematite) increases the net temperature rise within the reactor during the oxidation and reduction cycles for CO combustion only. However reduction of hematite with H_2 alone is an endothermic reaction and thus a slight temperature decrease is observed.

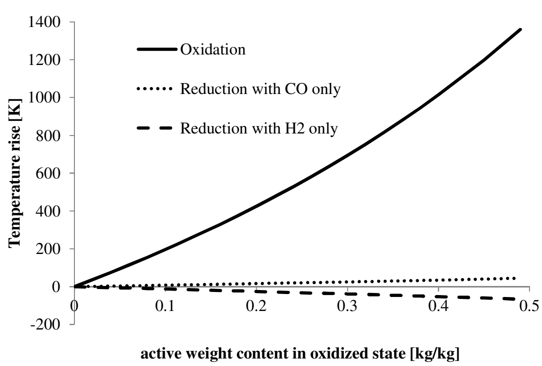

Figure 2. Tempereature rise during oxidation and reduction cycles; T_{ox} = 873 K, T_{red} = 1273 K

The reduction step was afterwards simulated for 0.40 mass fraction of hematite with the remaining as rutile (TiO_2) in ilmenite at the feed gas and reactor bed temperature of 1270 K. Figure 3 shows the OC concentrations profile for different components of the fuel during the reduction cycle keeping the flow rate of fuel gas constant. It has been envisaged that addition of steam, CO_2 and inert, in particular, increases the OC stability although complete conversion of fuel components into CO_2 and H_2O become possible at the exit of the reactor.

It can also be shown that the net temperature rise is only influenced by the mass fraction of active content of the oxygen carrier, while other parameters like air flow rate and length of the reactor only influence the cycle time during the oxidation cycle whithout altering the temperature change.

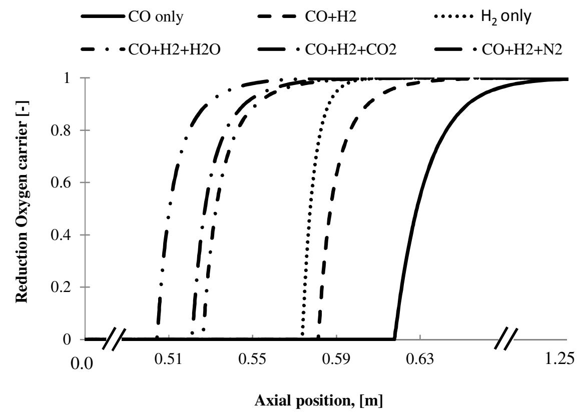

Figure 3. OC concentration profile in the fuel reactor during the reduction cycle. T_{red} = 1273 K, P = 5 bar

Conclusion

A 1-D model has been developed to design a packed-bed CLC reactor and validated afterwards for the syngas combustion with ilmenite. The proposed reactor model computes the net temperature rise, ilmenite conversion, CO_2 and steam production during the repetitive reduction and oxidation cycles and proposes the design parameters for the efficient and optimum performance of a packed-bed CLC reactor of syngas with ilmenite which becomes the basis for the scale-up of the reactor for power production with intrinsic capture of CO_2.

Acknowledgement

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