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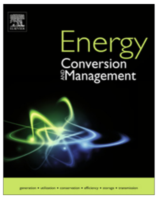

Simulation of an integrated gasification combined cycle with chemical-looping combustion and carbon dioxide sequestration

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ABSTRACT

Chemical-looping combustion is an interesting technique that makes it possible to integrate power generation from fuels combustion and sequestration of carbon dioxide without energy penalty. In addition, the combustion chemical reaction occurs with a lower irreversibility compared to a conventional combustion, leading to attain a somewhat higher overall thermal efficiency in gas turbine systems. This paper provides results about the energetic performance of an integrated gasification combined cycle power plant based on chemical-looping combustion of synthesis gas. A real understanding of the behavior of this concept of power plant implies a complete thermodynamic analysis, involving several interrelated aspects as the integration of energy flows between the gasifier and the combined cycle, the restrictions in relation with heat balances and chemical equilibrium in reactors and the performance of the gas turbines and the downstream steam cycle. An accurate thermodynamic modeling is required for the optimization of several design parameters. Simulations to evaluate the energetic efficiency of this chemical-looping-combustion based power plant under diverse working conditions have been carried out, and a comparison with a conventional integrated gasification power plant with precombustion capture of carbon dioxide has been made. Two different synthesis gas compositions have been tried to check its influence on the results. The energy saved in carbon capture and storage is found to be significant and even notable, inducing an improvement of the overall power plant thermal efficiency of around 7% in some cases.

1. Introduction

A clear evolution of power generation toward clean energy has been taking place for some time. However, fossil fuels are still being widely used and will continue to be for the next years. A number of technologies for achieving a significant reduction in CO_2 emissions from oxidation of fuels in the medium term are being investigated. One of the most promising is carbon capture and storage (CCS).

In practice, CO_2 can be captured by mainly three different alternatives: (a) "post-combustion", via amine chemical absorption [1]; (b) "pre-combustion", in the case there is a previous fuel decarbonization to a mixture of H_2 and CO_2 [2]; (c) "oxy-combustion", in which the fuel is oxidized by oxygen instead of by air [3], and then nearly pure CO_2 is obtained by cooling the combustion gases and removing the condensed water.

A key factor determining the real potential for widespread deployment of any of these techniques is their energy cost. Both pre- and post- combustion capture are based on separation by membrane or other conventional separation technologies, any of which entail a high energetic cost. Pre-combustion, when possible, is generally more advantageous because CO_2 is less diluted in the fuel than in the combustion gases, but its cost is still high.

Oxy-combustion would seem a good alternative, given that high-concentration CO_2 is obtained in the gases. However, the oxygen required for the process must be produced in a previous stage of air-separation, also highly energy-demanding [4].

In this context, research has been taking place in chemical-looping combustion (CLC), an alternative technique first proposed in [5]. With CLC, fuel and air are physically isolated. This leads to a higher energetic efficiency, since instead of separating O_2 from the air as in the case of oxy-combustion, it is extracted chemically from air by contact with an oxygen carrier that is oxidized. The resulting oxide is then conveyed to the fuel reactor; there it delivers the oxygen to the fuel and, once in its original form it returns to the air reactor and the process starts again.

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Nomenclature

ASU	air separation unit
CC	combined cycle
CLC	chemical-looping combustion
CCS	carbon capture and storage
CR	conversion ratio (%)
IGCC	integrated gasification combined cycle
LHV	lower heating value (kJ/mol)
TIT	turbine inlet temperature (K)
ΔH°_{298}	standard enthalpy of reaction at 25 °C/298.15 K (kJ/mol)
T_{red}	temperature at reduction reactor (K)
p_r	pressure at CLC reactors (bar)
η_{th}	power plant thermal efficiency
$W_{CLC\text{-}GT}$	gross gas turbines power (kW)
W_{ST}	gross power by the steam turbines (kW)
W_{aux}	auxiliaries power consumption (kW)
m_{bm}	biomass flow rate (kg/s)

CLC applied to gas turbine systems has been analyzed in the literature, considering mainly methane as fuel. For instance, Ref. [6] analyzes a methane-fueled combined cycle of gas and steam turbines with CLC, [7] studies the viability of non-conventional oxygen carrier for CLC of methane, [8] provides a model-based evaluation at pre-commercial stage of a combined cycle and [9] gives a theoretical exploration of chemical-looping hydrogen (CLH) generation with methane as fuel, which is a CLC variation. Another interesting CLC variant known as chemical-looping with oxygen uncoupling (CLOU) is explored in reference [10]. Some other fuels such as methanol have been proposed as well [11]. There also exist energetic and exergetic analyses of CLC gas turbines fueled with synthesis gas (syngas). Reference [12] gives second-law of a CLC-syngas combined cycle and [13] provides further simulation results of CLC-syngas with different oxygen carriers, but energy savings in the capture of CO_2 are not quantified.

There is also very interesting prior work on the potential of integrating CLC with combined cycles, like the analysis of the trigeneration system proposed by [14], although it has not been sufficiently explored. Combining CLC and IGCC in particular, could achieve highly efficient power generation together with nearly zero greenhouse gas emissions. This work focuses on this technique.

A recent previous work analyzed the energetic performance of a CLC combined cycle (CC) syngas power plant in detail, estimating the energy savings in CO_2 [15]. The present work tries to extend that analysis to a complete CLC-based IGCC power plant considering the integration of the gasification stage with power production by a CLC combined cycle and CO_2 sequestration and storage.

2. Chemical-looping combustion

The CLC conception is schematically shown in Fig. 1. Two reactors substitute the conventional combustion chamber in order to avoid a direct contact between air and fuel. The first reactor is usually called "air reactor" or "oxidation reactor". There an oxygen carrier, denoted here generically by "Me" (as it is typically a metal oxide), is put in contact with air from which it takes some amount of oxygen by being chemically transformed into an oxidized form "MeO":

$$\mathrm{Me} + \frac{1}{2}\mathrm{O_2} \rightarrow \mathrm{MeO} \qquad (1)$$

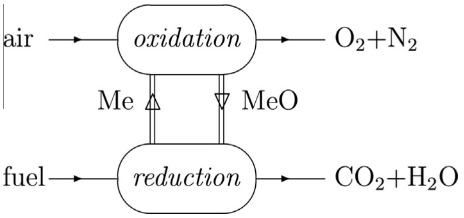

Fig. 1. Chemical-looping combustion concept.

Afterward, the oxygen carrier "MeO" is relocated to the second reactor, the "fuel reactor" or "reduction reactor". Then, "MeO" reacts with fuel getting back its original chemical form "Me" and oxidizing the fuel according to:

$$(2n+m)\mathrm{MeO} + \mathrm{C}_n\mathrm{H}_{2m} \rightarrow (2n+m)\mathrm{Me} + m\mathrm{H_2O} + n\mathrm{CO_2} \qquad (2)$$

$$\mathrm{MeO} + \mathrm{CO} \rightarrow \mathrm{Me} + \mathrm{CO_2} \qquad (3)$$

As a result, the gaseous stream at the reduction reactor outlet is a mixture of only carbon dioxide and water, from which CO_2 can be easily isolated by cooling and condensation of water. The stream at the oxidation reactor outlet is depleted air, *i.e.* mainly N_2 and O_2.

The principal benefit of CLC is to obtain the carbon dioxide that results from the fuel oxidation in a relatively pure form instead of diluted in air or any other non-condensable gas. This implies that there is not need to consume energy for CO_2 capture and the only energy needs for sequestration of CO_2 would be the compression power consumption up to the storage pressure. The energy savings in this step is very important.

Furthermore, if the oxygen carrier is chosen properly, also the power production can be increased compared with a conventional combustion system. In effect, if one or both of the reactions (2), (3) are endothermic, CLC can act as a sort of "chemical heat pump". For instance, these endothermic reactions can be enforced to take place at low/medium temperature supplying all the required heat from a medium temperature source, *e.g.* the exhaust gases stream from a gas turbine. Since the amount of heat released in both reactions (reduction and oxidation) must equal the fuel's heat of combustion (Hess' law,), the oxidation reaction must release more heat than a conventional combustion. Thus, the amount of heat obtained at high temperature can be increased. It is well known that the exergy content of heat is higher as the temperature of the source increases, so in this way more power can be attained for the same amount of fuel. This is equivalent to say that that the exergy destruction due to the irreversibility associated with the chemical reactions that occur in the whole of both reactors in the case of CLC is lower than the irreversibility in a conventional combustion, as noted and quantified in [13].

A solution often proposed for implementing the CLC scheme shown in Fig. 1 consists in fluidized bed reactors. If solid particles are prepared fine enough, a suitable contact area between them and gases can be achieved so that chemical reactions occur satisfactorily. It is common to add inert material particles to improve their stability and other physical properties or some catalyst [6].

3. Description of the study and methodology

A whole CLC-based IGCC power plant with CO_2 sequestration has been simulated, including calculations related to the gasifier, CLC gas turbine system, steam cycle and CO_2 separation and