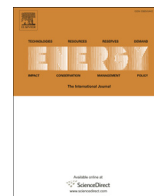




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Thermodynamic analysis of a dual power-hydrogen production system based on chemical-looping combustion

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ABSTRACT

Chemical-looping hydrogen generation (CLHG) is a chemical-looping combustion variant that allows simultaneous production of power and hydrogen. Additional integration of systems would allow the recovery of waste heat with extra advantage. A thermodynamic analysis from the exergy method point of view of an integrated syngas-fueled CLHG cycle is carried out with the aim of contributing to the conceptual understanding and development of CLHG systems. The analysis gives place to an optimization of the cycle performance in a range of working conditions. The proposed system shows a very interesting potential for trigeneration of power, hydrogen and process heating with notable overall efficiency.

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1. Introduction

The global warming phenomenon that is currently taking place should be mitigated by reducing the emissions of greenhouse gases (GHG) to atmosphere [1]. As it is well known, a large part of the anthropogenic GHG emissions is due to the combustion of fossil fuels in transport and power generation. Until a transition to the use of new clean sources of energy is eventually achieved, a possible option for reducing the impact of power generation from fossil fuels is the carbon capture and storage (CCS). A review on several alternatives for this goal can be found in Ref. [2]. Nevertheless, the main available techniques (pre-combustion carbon capture [3], post-combustion carbon capture [4] and oxy-combustion [5]) imply severe energy penalties related to the difficulty of separation of pure gases from a mixture of them. The excessive increment of the power production cost makes very hard in practice the implementation of this kind of steps.

Chemical-looping combustion (CLC) is a promising different approach that intends to make possible the carbon capture without

large energy consumption. First proposed by Ishida and Jin [6], and subsequently patented in USA [7], CLC is defined as a thermochemical process where fuel oxidation is carried out in an indirect way by means of an intermediate agent that actuates as oxygen carrier between two separated reactors: i) a fuel reactor, where the oxygen carrier is reduced oxidizing the fuel, and ii) an air reactor, where the oxygen carrier is oxidized in air. Overall, the fuel experiences the same chemical transformation as a conventional combustion, with the fundamental advantage of segregating the oxidation products CO₂ and H₂O apart from air, and thus the only non-condensable gas in the output flow is CO₂. Interesting research has been carried out on the application of CLC to gas turbine systems for power generation by several authors. For instance, ref. [8] gives an analysis of a gas-steam combined cycle fired by methane with CLC [9]; provides a pre-commercial evaluation of a CLC combined cycle scaled plant. Also CLC gas turbine systems based on alternative fuels such as methanol have been proposed [10]. In Ref. [11] an interesting exergy analysis of a CLC gas turbine system considering methane and syngas as fuels and nickel and iron oxides as oxygen carriers is presented.

Some other recent works focus on CLC combustion of syngas in gas turbine systems [12] exploring a possible integration of combined cycle power plants with carbon or biomass gasification. A

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complete integrated gasification combined cycle power plant based on CLC is analyzed in Ref. [13].

An attractive variation of CLC is the so-called chemical-looping hydrogen generation (CLHG). An interesting theoretical exploration of CLHG can be found in Ref. [14]. The main idea is to split the combustion into three stages instead of into two, introducing a third reactor in the chemical looping, as shown in Fig. 1. The complete reduction of the oxygen carrier takes place in the fuel reactor (FR), and its oxidation takes place in two steps, partially in a steam reactor (SR) and completely at the air reactor (AR). Some heat is released in the AR, which can be introduced in a power generation cycle, typically a gas turbine power system. Also, a mixture of steam and hydrogen is obtained as output from the SR, allowing for obtaining pure and already pressurized hydrogen just by condensation of water. Furthermore, the outcome of the FR is a blend of almost only H_2O and CO_2 , so the carbon dioxide can be easily separable as well. Both power and hydrogen can be generated in this way simultaneously from a fossil fuel, and the carbon dioxide that results from the fuel's oxidation can be sequestered with a high potential of energy savings.

In this context, a combination of the two previous ideas, *i.e.* the integrated gasification of coal upstream to a syngas-fueled gas turbine system, and the CLHG concept itself, is explored very convincingly by Ref. [15]. A complex system is modeled and both, energy and exergy analysis are provided.

A similar approach is followed in the present work. Primarily, the exergy method is followed here to carry out a thermodynamic theoretical analysis of a CLHG system fueled by syngas obtained from coal gasification. In this paper there are several novelties. The CLHG cycle design and configuration has its own particularities. Several thermodynamic aspects are taken into account for this study that are disregarded or not mentioned in other works, in particular the influence of the key thermodynamic conditions of the cycle, such as the CLHG reactors pressure and equilibrium temperatures of the FR and the SR, on the behavior of the main cycle components, the complex intertwined energy balances and chemical equilibrium in reactors and the quantification of the exergy flows in the system, among others. This thermodynamic theoretical analysis allows a subsequent step for the optimization of the cycle performance. Thus, this work is intended to contribute to the conceptual understanding and development of combined power and hydrogen chemical looping generation with high efficiency.

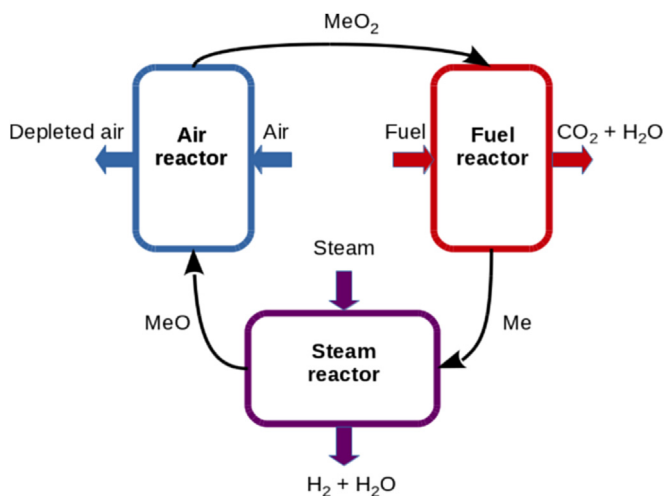


Fig. 1. Representation of the CLHG concept.

2. Proposed CLHG system

2.1. Cycle description and operating conditions

The proposed CLHG system is schematically presented in Fig. 2. Its most important components are enumerated as follows:

- Two gas turbines. GT1 is the main gas turbine and generates power from the expansion of the depleted air stream. Additionally a second gas turbine GT2 is integrated to expand the pressurized $\text{H}_2\text{O}/\text{CO}_2$ stream obtained as output from the FR for extra power production.
- A heat recovery steam generator (HRSG). It is fed by the streams at both gas turbines outlets for additional power production in a steam turbine cycle (ST). This part of the system is considered conventional and has been modeled in a simplified way instead of resolving the details.
- The three looping reactors FR, SR and AR. They are pressure-linked, since the oxygen carrier describes a closed loop. The SR is fed by a steam extraction from the HRSG.
- A compression stage for carbon dioxide and hydrogen streams. Both gases are obtained blended with water, so previously to the compression a condensation of water is required.
- Similarly to the $\text{H}_2\text{O}/\text{CO}_2$ stream, the $\text{H}_2\text{O}/\text{H}_2$ stream could be reheated and expanded in a third gas turbine to obtain some extra work. This is the case in the CLHG system proposed by Ref. [15]. On the contrary, here we have preferred to consider a trigeneration scheme where some amount of process heating can be supplied by the $\text{H}_2\text{O}/\text{H}_2$ stream, prior to the water condensation and the later H_2 compression.

There are some other cycle components: air compressor (AC), fuel compressor (FC), carbon dioxide compressors (CDC), hydrogen compressors (HC) (a two-stage compression is assumed for both cases), air filter (AF) and several condensers for water extraction from gaseous streams.

The cycle parameters are summarized next: Ambient temperature 15°C (288.15 K), ambient pressure 1 atm (1.01325 bar) and air molar composition resulting from a value of 60% relative humidity (RH): N_2 77.26%, O_2 20.78%, H_2O 1.01%, Ar 0.93% and CO_2 0.03%. Pressure drop at the air filter 0.01 bar. Isentropic efficiency of all compressors: 0.845. Isentropic efficiency of gas turbines: 0.895. Syngas input conditions are taken as provided by the gasifier after cleanup, 153.4°C (426.58 K) and 27.24 bar (taken from Ref. [16]). A 4% pressure drop in each reactor is adopted. Heat losses of 0.5% of the fuel's lower heating value (LHV) in the AR and 0.2% in both, FR and SR. Pressure drop of 3.5% in the HRSG. Temperature of exhaust air at HRSG outlet is the highest among the dew point and 90°C (363.15 K). The compression pressure for carbon dioxide and hydrogen has been set to 85 bar, allowing storage or transport as high-density supercritical fluid. These operating conditions are considered to be within typical ranges in combined cycle power plants.

The behavior of this hybrid power- H_2 generation CLHG plant is evaluated by simulations in a range of operating thermodynamic conditions. For that purpose, the temperature attained at the AR (denoted here by T_{AR}) and the pressure of the looping reactors (denoted here by p_{R}) are varied along a range of values to evaluate their influence on the cycle performance. An analysis for the optimization of some other thermodynamic parameters is provided.

2.2. CLHG reactors

The chemical-looping scheme adopted here is a three-reactor system previously proposed for CLHG from methane [17] and

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