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TECHNICAL ANALYSIS OF THE USE OF BIOGAS PRODUCED IN THE STATE OF PARANÁ AS FUEL FOR HYDROGEN AND SYNGAS PRODUCTION THROUGH CHEMICAL LOOPING

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Abstract. *The sugar-alcohol industry is a significant economic driver in Brazil, known for its outstanding biomass generation that can be utilized as a sustainable energy resource. Several scholars suggest that generating biogas from waste produced by this sector can be a practical alternative, as it facilitates the recuperation of energy from these materials. Chemical looping is a highly promising carbon dioxide (CO_2) capture technology in energy production due to its inherent CO_2 separation capabilities. The classical three-reactor chemical looping process, which is also known as TR-CLH, consists of the Fuel Reactor (FR), Steam Reactor (SR), and Air Reactor (AR). The process has the potential to co-produce syngas and high-purity hydrogen, but presents low yields and purity, achieving only 0.200 mole fraction under adiabatic conditions. The aim of this study is to simulate the technical feasibility of biogas as a fuel in Three-Reactors Chemical Looping Hydrogen (TR-CLH) through Aspen Plus software. It is expected to produce syngas as a gaseous fuel primarily for obtaining hydrogen or liquid fuels while capturing CO_2. Syngas consists mainly of hydrogen (H_2) and carbon monoxide (CO), with a significant amount of CO_2 and methane (CH_4).*

Keywords: *Chemical Looping, Biogas, Syngas production, CCS.*

1. INTRODUCTION

Over the past few years, the energy generation sector has been dedicated to implementing more efficient technologies due to the high consumption of hydrocarbon resources, which are primarily responsible for damaging the atmosphere through the significant emission of greenhouse gases. The rise in Earth's temperature, which implies reduced ice mass at the poles and habitat changes for various species, is attributed to a substantial increase in carbon dioxide concentrations since the industrial era's inception. According to the EPA (2022), the average annual concentration rose from 280 ppm in the late 1700s to 414 ppm in 2021.

The process of capturing carbon dioxide can be accomplished through biological, physical, or chemical means. However, Wennersten et al. (2015) found that these methods only demonstrate the effects of CO_2 reduction over extended periods and are insufficient to counteract CO_2 emissions. To address this, carbon capture and storage (CCS) technologies have been developed to mitigate CO_2 gas emissions by reducing its concentration in the atmosphere. Among the CCS technologies, to this date the most developed are:

1) pre-combustion removes the carbon dioxide after producing a synthesis gas composed of carbon monoxide (CO) and hydrogen (H_2); provided from the fuel conversion involved (Saldívar Esparza et al. 2017).
2) oxy-combustion; involves the modifying of the combustion process to obtain a high CO_2 concentrated fuel gas, to burn with almost purely oxygen (O_2) gas (being 95%) (Saldívar Esparza et al. 2017).
3) post-combustion; is a process that enables low carbon dioxide concentrations given the possibility that it can be performed by absorption (physical and chemical), adsorption, cryogenic separation and membrane separation (Saldívar Esparza et al. 2017).

Chemical Looping (CL) is a highly efficient and flexible process that distinguishes itself from other CCS technologies due to its ability to perform various combustion processes. Also known as Chemical Looping Combustion (CLC), Labiano (2017) proposes that biofuels or biomass could be used as primary sources for heat and energy production, or the production of synthesis gas and/or hydrogen gas using reforming techniques. This makes it one of the most favorable alternatives for energy production systems with negative emissions of carbon dioxide gas. The CL technology transfers oxygen from air to fuel via an oxygen carrier (OC), typically a metal oxide. This avoids direct contact between air and fuel (Flores 2014). According to Serrano (2018), this process typically utilizes two reactors; one for oxidation where a

release of energy occurs from the exothermic reaction involved in the metal's oxidation, converting it to a metal oxide. At the second reactor, known as the reduction reactor, the mixture of metal oxide and energy that is released from the first reactor is received. At this stage, the process generates carbon dioxide and water as a result of the reduction reaction in the presence of the fuel.

Metal oxides are often used as oxygen carriers in conjunction with inert materials. Various metal oxides have been suggested as potential candidates for this role in the literature. Flores (2014) observed that among these contenders, those containing nickel, copper, iron, and manganese have demonstrated the most promising characteristics. Throughout the process, it assumes the most crucial role, however, it is also the most complex aspect of this technique due to its expensive production at an industrial scale, (Penthor 2017).

Labiano (2017) reported that over the past two decades, the Chemical Looping (CL) technology has undergone significant evolution, with thirteen pilot plants developed for gaseous fuels, three for liquid fuels, and eighteen for solid fuels. The use of biofuels like biomass, biogas, or bio-oils offers a distinct advantage of negative carbon dioxide emissions. In this process, the gas is captured and stored at the outlet of the reduction reactor, and eventually absorbed for the biofuel production that involves biological processes.

Biogas is a gaseous mixture generated by the anaerobic digestion of organic materials. It consists mostly of methane (CH_4) and carbon dioxide (CO_2) with low concentrations of hydrogen sulfide and ammonia (Zanette 2009). In 2021, approximately 811 biogas plants were counted in Brazil, indicating a high potential for biogas production in the area. According to CIBiogás (2021), 755 of these plants were operational, accounting for 93%. Additionally, 44 were in the implementation process (5%), and 12 were undergoing reform (2%). This marks a 20% growth compared to the 2020 numbers of 675 plants, resulting in an annual production of at least 2.8 billion of biogas. For the development of the proposed analysis, the potential of sugar cane residues to produce biogas in the state of Paraná, cited by the author Penteado (2022).Three-Reactors Chemical Looping Hydrogen (T-R CLH) involves three reactors being; the fuel reactor (FR), the steam reactor (SR) and the air reactor (AR). T-R CLH technology is considered to be the best method for carbon dioxide capturing at the fuel reactor, where it allows SYNGAS and/or hydrogen to be produced; depending on the oxygen carrier feed flows by modifying such (Wang et al. 2023). Chiesa et al. 2008 offers a general description of the process:

1) At the fuel FR, iron oxides (Fe_2O_3), present in the oxygen-rich hematite state, such species are frequently reduced to ferrous oxide (FeO); such occurs by oxidizing the fuel generating an endothermic reaction. Next the outflow stream presents water and carbon dioxide, this simplifies the geological storage of carbon dioxide after the condensation process of the water.
2) In the steam reactor, most of the reduced species (FeO) reacts with the steam present forming magnetite (Fe_3O_4) and hydrogen gas, being the final products of this process. Given that the reaction is exothermic, a large steam excess is needed in order to be able to obtain an acceptable oxidation of the metal, therefore, the exhaust steam gas is a mixture composed of hydrogen and water.
3) As noted, most of the ferrous oxide undergoes in reaction, therefore, at the air reactor, magnetite (with traces of unreacted ferrous oxide) fully oxidizes into iron oxide (Fe_2O_3). Finally, the highly exothermic reaction manages to keep thermal equilibrium throughout the system, where air with low concentrations of oxygen (lean air) is discharged from the reactor (p. 2).

Various researchers have assessed solid fuels for hydrogen production using methods related to this study. For instance, Varon Cardona et al. (2022) introduced pellets as fuel for the process and reported a proportional relationship between hydrogen gas production and oxygen carrier oxidation. The oxidation temperature has an impact on the reaction time, as demonstrated in the study.

Another instance of Chemical Looping hydrogen production was demonstrated in a study by Isarapakdeetham et al. (2020) utilizing ethanol as a fuel. The focus of their research was to examine the impact of using Ce^{4+} and La^{3+} in Nickel and Aluminum oxygen carriers to analyze their behavior alongside ethanol over five cycles.

The purpose of this study is to assess the viability and functionality of the T-R CLH method, comprising three reactors, for generating hydrogen and synthesis gas utilizing biogas as a high-potential fuel source in the state of Paraná.A fundamental inquiry was conducted using pertinent sources and Aspen Plus simulation software to obtain outcomes. The inquiry examined the impact of diverse variables such as reactor temperature, water and air supply flows, fuel, and oxygen carriers.

2. PROCESS DESCRIPTION

2.1 Three Reactors Chemical Looping Hydrogen

The process comprises three reactors: the fuel reactor (FR), steam reactor (SR), and air reactor (AR). Each reactor undergoes multiple reactions that are either exothermic or endothermic, depending on the involved reactants (Khan and Shamim, 2016). Figure 1 depicts a scheme demonstrating the process for production of hydrogen and SYNGAS, along with the inlet and outlet molar flows for each stage.

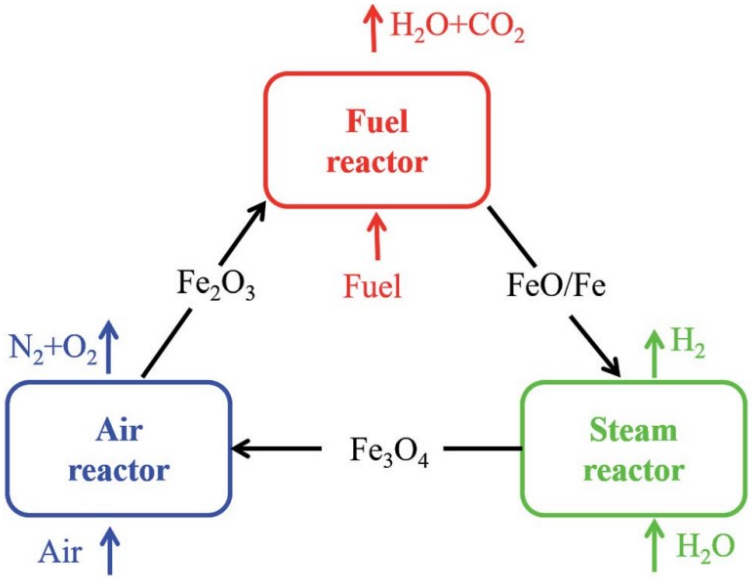

Figure 1. Three Reactors Chemical Looping Hydrogen Road map (Xu et al. 2019)

Along the process, four reactions take place which represent the stages of reduction, oxidation and steam methane reforming provided by the authors Chiesa et al., 2008; where positive energy values refer to energy released in the form of heat, and the negative convention for such results refers to energy consumption.

$$4Fe_2O_3 + CH_4 \rightarrow 8FeO + 2H_2O + CO_2 - 356{,}5\ kJ/kmol \quad (1)$$

$$8FeO + \frac{8}{3}H_2O \rightarrow \frac{8}{3}Fe_3O_4 + \frac{8}{3}H_2 + 199{,}3\ kJ/kmol \quad (2)$$

$$\frac{8}{3}Fe_3O_4 + \frac{2}{3}O_2 \rightarrow 4Fe_2O_3 + 314{,}6\ kJ/kmol \quad (3)$$

$$CH_4 + \frac{2}{3}H_2O + \frac{2}{3}O_2 \rightarrow \frac{8}{3}H_2 + CO_2 + 157{,}4\ kJ/kmol \quad (4)$$

Between the reactors, the oxygen carrier (OC) is responsible for transferring oxygen (Khan & Shamim, 2016). Therefore, the efficiency of Chemical Looping processes depends on this parameter. As a result, the following features must be present: chemical stability, ability to be fluidized, strong affinity to react with inlet gases, stability at high operating temperatures, resistance against agglomeration, and low environmental impact (Mattisson et al. 2001).

2.2 Simulation

The thermodynamic analysis was carried out by simulating it in Aspen Plus V11® with the use of the Peng Robinson equation of state along with Boston-Mathias modification as the thermodynamic model. This technique, according to Wang et al. (2023), is commonly employed in other related works. For the T-R CLH process, biogas was selected as the primary fuel. The composition of the biogas consists of 60% methane CH_4 and 40% carbon dioxide CO_2. The effects of other present substances in small concentrations such as hydrogen sulfide and ammonia on reactor balance were disregarded. Iron oxide Fe_2O_3, also known as OC, undergoes reduction by reacting with biogas in the fuel reactor (FR) to obtain ferrous oxide FeO, iron Fe, and a gas stream labeled “SYNGAS” (CH_4, H_2, CO_2, O2, CO, H_2O) as products. The product stream then passes through a cyclone to separate the solid and gas flows. Inside the steam reactor, iron and ferrous oxide undergo oxidation by the steam present, resulting in the production of magnetite (Fe_3O_4) and release of hydrogen gas. Like the fuel reactor, the product obtained in the steam reactor undergoes an ideal separation between the gas and solid phases. Eventually, the magnetite is oxidized within the air reactor (AR), converting it back into iron oxide Fe_2O_3 and producing a flow of flue gas or air with low concentrations of oxygen as a product.

The FR, SR, and AR reactors were simulated using the RGibbs simulation model with Gibbs energy minimization in the calculations. Three solid-gas separators were used to separate solid particles from the gas phase. These were simulated with the SSplit model (Wang et al. 2023). The required temperatures of the fluids (water and air) were obtained. Two simulated heat exchangers were used with the Heater model. An isobaric process was conducted at a pressure of 1.2 bar through two compressors. The parameters were set as follows: the fuel reactor (FR) was kept at a constant temperature, while the steam (SR) and air (AR) reactors were kept adiabatic.

The biogas inlet flow is 1 kilomole per hour, at a temperature of 25°C and atmospheric pressure. The water flow rate ranges from 10 to 14 kilomoles per hour, while the air flow rate ranges from 8 to 16 kilomoles per hour. These flow rates start at a temperature of 25°C and atmospheric pressure, and are subsequently heated to enter the reactor at a temperature of 150°C. The iron oxide feed rate ranges from 2 to 8 kilomoles per hour. The process flow diagram is shown in Figure 2.

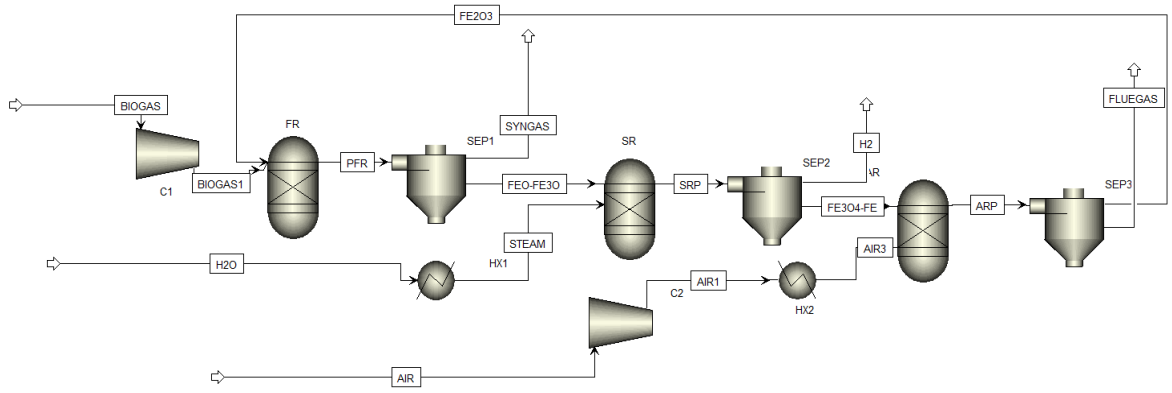

Figure 2. Three Reactors Chemical Looping Hydrogen flowsheet simulation in Aspen Plus.

3. ANALYSIS AND RESULTS

The development of the proposed thermodynamic analysis was achieved through adjusting operational parameters in the initial simulation, including fuel reactor temperature and feed molar flows of air, water, and oxygen carrier. Various alterations were made to the system, resulting in differing molar flows of produced hydrogen and SYNGAS composition ratios, as well as a flue gas whose temperature varied throughout the process. Wang et al., (2023) proposed an equation to determine the purity of the Syngas produced during the process:

$$Syngas\ purity = \frac{f_{CO} + f_{H_2}}{f_{SYNGAS}} \times 100 \quad (5)$$

f_{SYNGAS} corresponds for the sum of the mole fractions of the gas components produced (CO, CO_2, CH_4, H_2), the numerator refers to the sum of the mole fractions of hydrogen gas and carbon monoxide composing the SYNGAS. Zhang et al. (2020) investigated the parametric conditions for the autothermal behavior of the Three Reactors Chemical Looping Hydrogen process. They determined these conditions based on the Fe_2O_3/fuel molar ratio in the reduction reactor and the ratio between steam input to the oxidation reactor and fuel input to the reduction reactor, where natural gas served as the fuel. The authors then presented energy and exergetic balances, concluding that an autothermal process can produce hydrogen. In this study, we have conducted a basic analysis while assuming that the energy consumed in the fuel reactor comes from the temperature of the flue gas.

3.1 Fuel Reactor Temperature

This variable was selected based on the findings of authors such as Varon Cardona et al. (2022) and Maicas (2015), who presented results regarding the impact of temperature on the reduction reactor. The values adjusted in this study align with those cited by Maicas and colleagues, as they pertain to biogas as a fuel source. The authors cited found a reduction in reaction time with an increase in reactor temperature, operating within the range of 650 to 850°C for biogas.

The fuel reactor (FR) is where the reduction of ferrous oxide (Fe_2O_3) to ferrous oxide (FeO) and magnetite (Fe_3O_4) occurs, resulting in the release of SYNGAS in small concentrations. The TR-CLH process simulation involved maintaining the initial settings for other parameters, with the FR starting at 550°C and then increasing at a rate of 50°C until reaching 950°C.

During this stage it is important to emphasize that the molar flows of SYNGAS and hydrogen gas remained constant with values of 2.609 and 10 kmol/h, while the molar flow with respect to the FLUE GAS decreases substantially as there is an increase in the temperature, such can be seen represented in Figure 3. In Figure 4 the composition of the hydrogen outlet flow of the steam reactor (SR), demonstrating the high increase of the molar fraction of the highly pure hydrogen gas in regard to the increase of temperature inside the fuel reactor (FR) until achieving its maximum point being 750°C with a value of 0,200, afterwards the decrease in this fraction occurs. The composition for the outflow corresponding the SYNGAS from the FR is presented in Figure 5, where the mole fractions of carbon monoxide and hydrogen gas appear

to decrease gradually with the increase in the FR temperature, up to 750°C Starting from this temperature such parameters start to spike. For the SYNGAS purity, such achieved its highest point at 550C with a value of 31.1158% whilst at 750°C it reached its lowest value of 19.004%. Figure 6 provides the temperature analysis respectively of the SYNGAS outflow inside the fuel reactor and the FLUE GAS in the air reactor, where it is seen that between the temperature ranges from 550°C to 700°C in the FR; the FLUE GAS keeps up a high temperature, allowing consider an autothermal character of the process, where starting the 750°C, the temperature for the flow in respect to the SYNGAS is greater than that of the FLUE GAS flow.

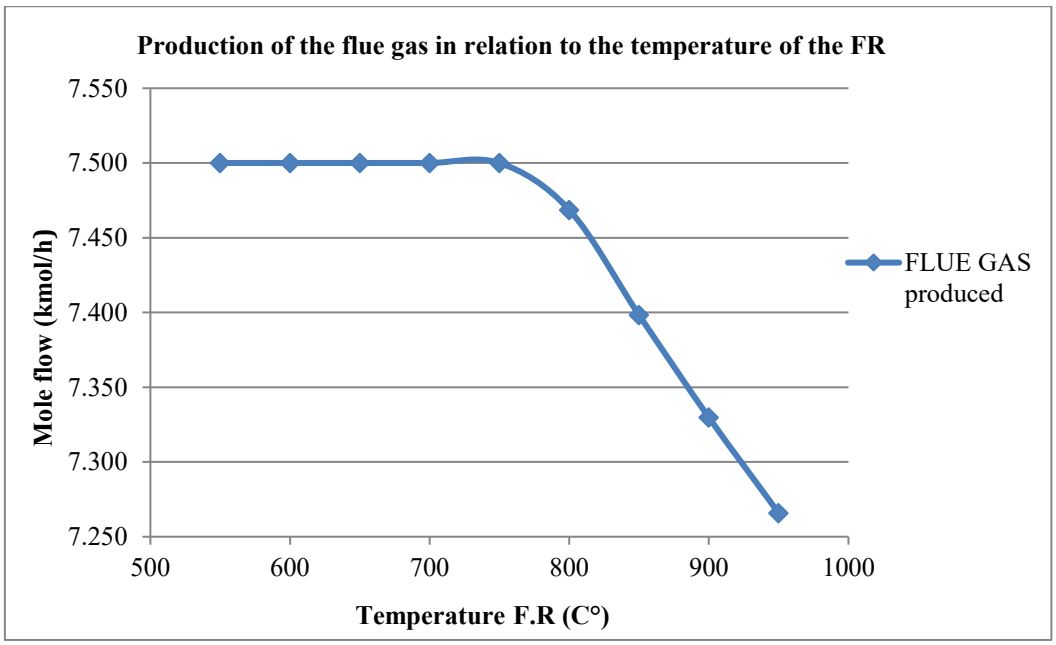

Figure 3. Produced FLUE GAS molar flow in the relation to the FR temperature.

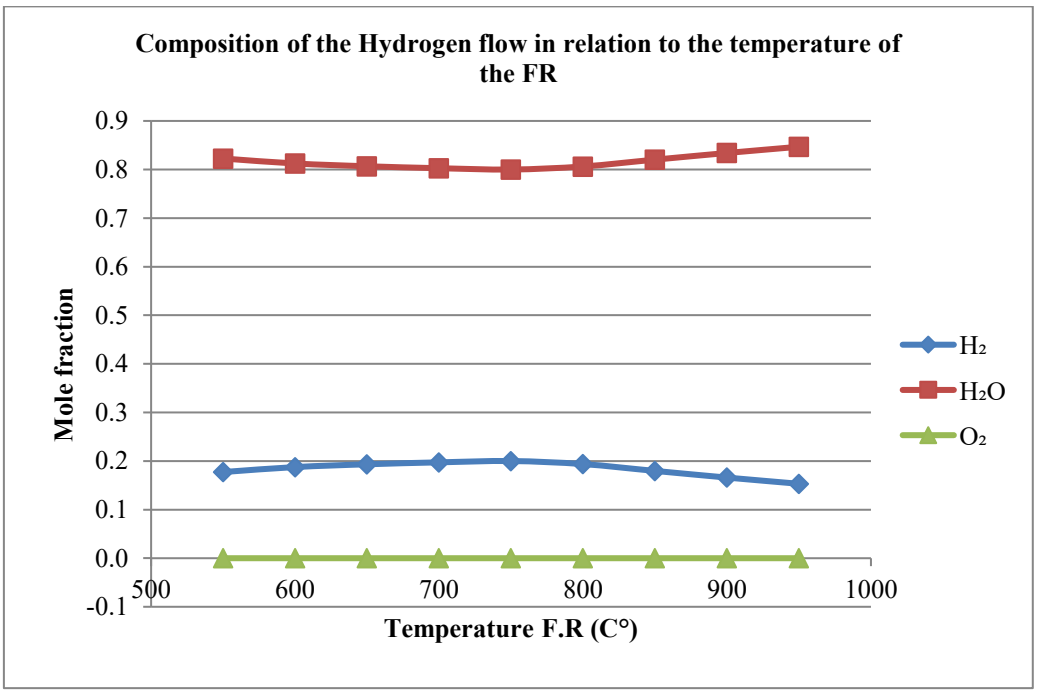

Figure 4. Hydrogen outlet flow composition in the Steam Reactor (SR)

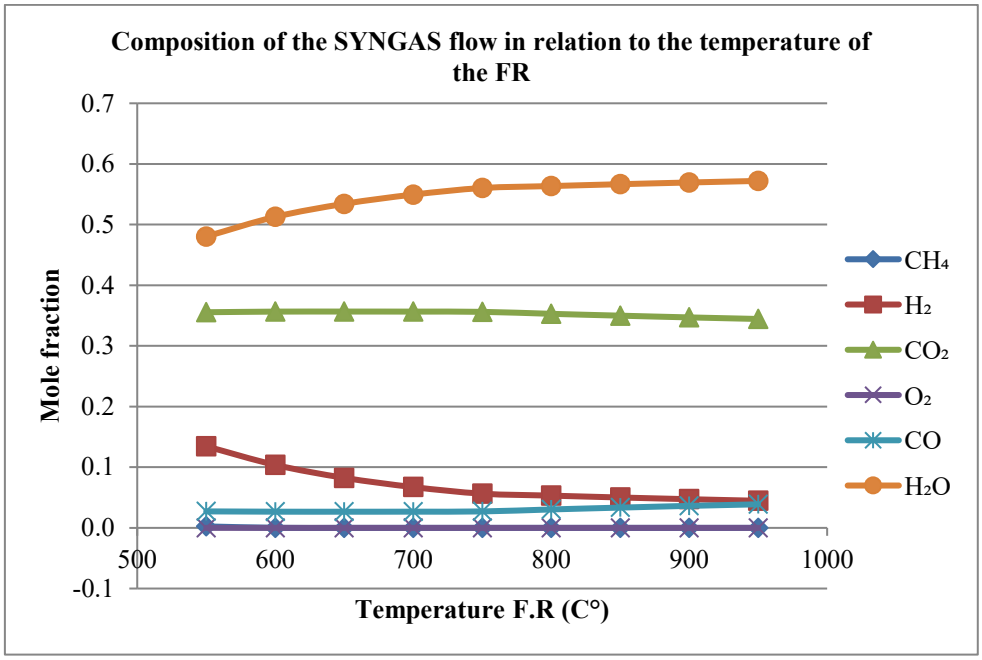

Figure 5. SYNGAS outlet flow composition inside the Fuel Reactor (FR)

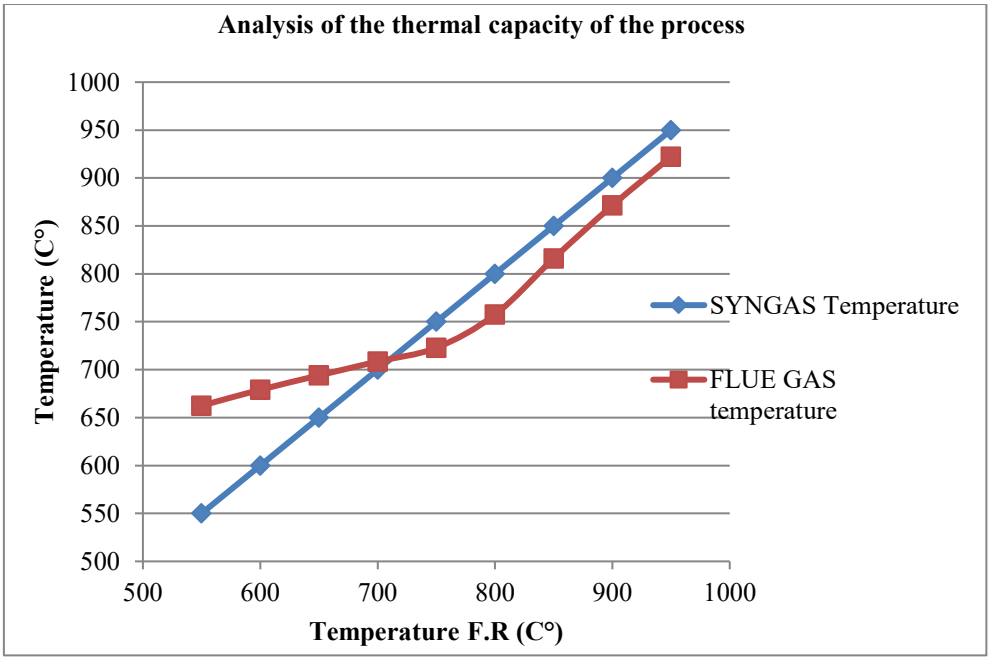

Figure 6. Temperature analysis corresponding the outlet flows of the FR and AR respectively in relation to the FR temperature

3.2 Feed molar flows

Air Flow

For the fuel reactor the temperature was maintained constant at a value of 550°C, setting the water feed molar flow at 10 kmol/h, oxygen carrier (Fe_2O_3) feed molar flow at 2 kmol/h, the simulation was executed increasing the air feed flux, being fundamental for the oxidation reaction taking place at the air reactor, at a 1 kmol/h rate. In Figure 7 it can be evidenced that the alteration of this parameter affected the temperature of the FLUE GAS produced in the air reactor, yet it can be perceived that the increase in air flow causing the FLUE GAS to decrease, where this temperature presents itself higher than the operating temperature of the fuel reactor, until the molar air flow surpasses 13 kmol/h. For the hydrogen and SYNGAS outflows no variations or fluctuations were reported.

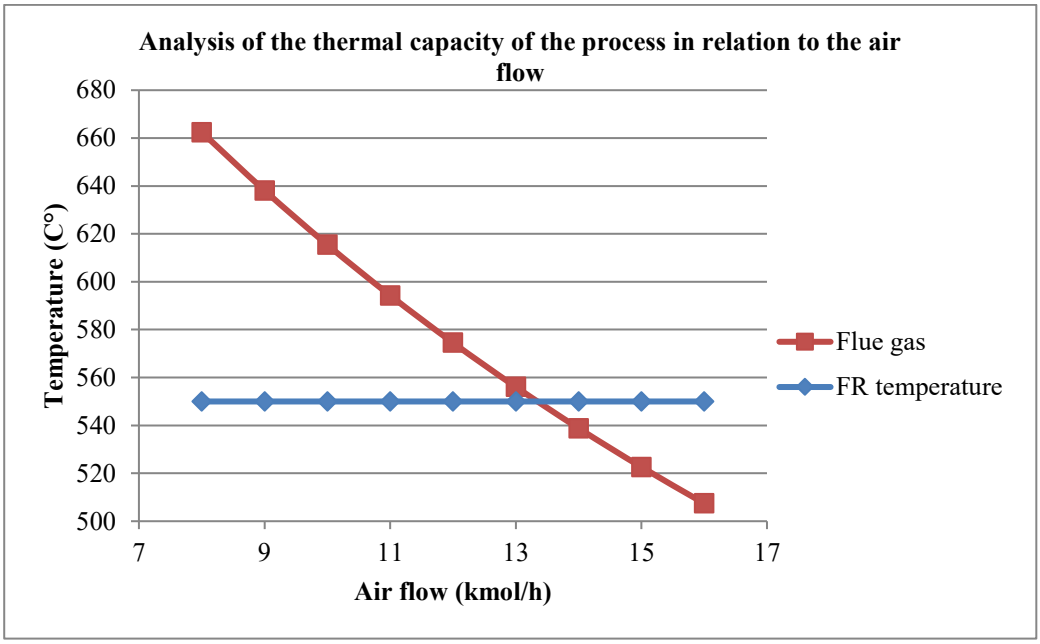

Figure 7. Analysis of the FLUE GAS temperature behavior in relation to the operating temperature of the FR by the increase of the molar feed flow of the air.

Water Molar Flow

At this stage initial parameters were maintained to perform the simulation, then water feed flow rate was increased at 0.5 kmol/h in the steam reactor (SR). The composition of the hydrogen flow with respect to the outlet of the SR is shown in Figure 8, at such region it is observed a decrease in the mole fraction of the hydrogen gas corresponding to the product; therefore the highest mole fraction of hydrogen achieved corresponded to 0.177, obtained setting a water feed flow of 10 kmol/h. Figure 9 shows the analysis regarding the FLUE GAS outlet temperatures and the fuel reactor operating temperature, it can be noted that an increase in respect to the water flow rate leaned in small ratios to the decrease of the gas temperature, still and all the species maintains a higher temperature than the reactor temperature at all simulated instants, making it possible to carry out an analysis minding an autothermal behavior of the process.

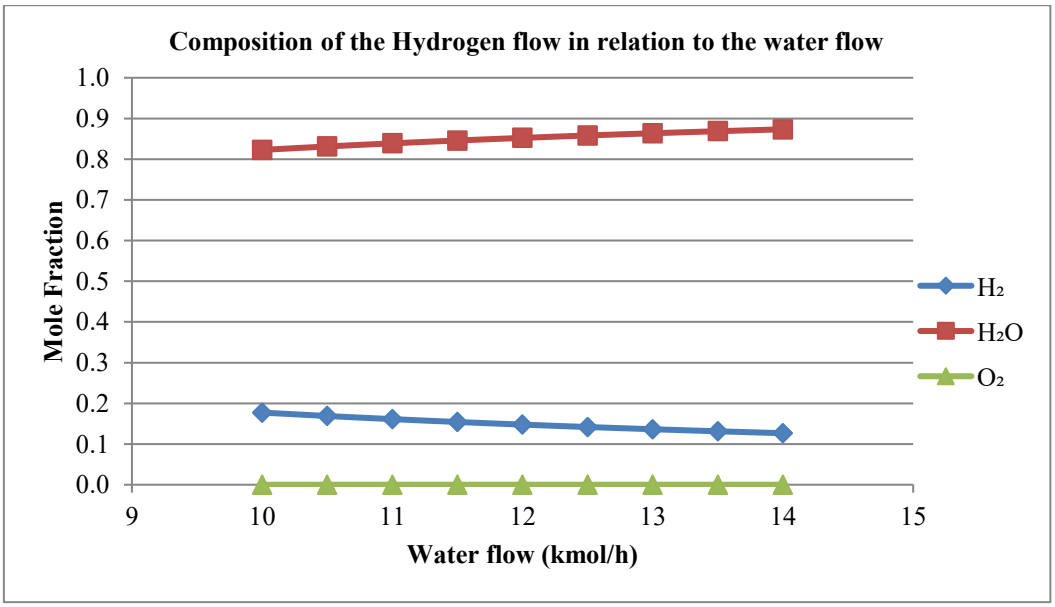

Figure 8. Composition of the produced hydrogen flow in relation to the increase in water feed flow.

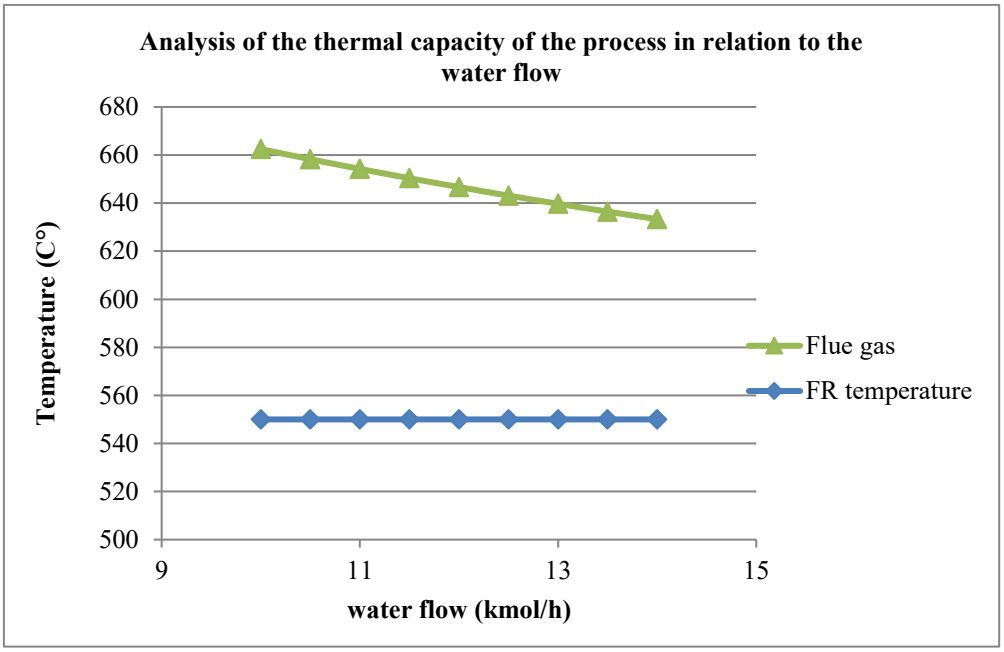

Figure 9. Temperature analysis obtained corresponding to the FLUE GAS in relation to the FR temperature by the increase on water feed flow.

Oxygen carrier flow

For the following analysis the initial parameters settings of the simulations were settled, whilst the iron oxide feed flow rate increased from 2 to 8 kmol/h. Such variation caused the hydrogen mole fraction to lower at the outflow in the steam reactor (SR), gradually decreasing almost to zero, as shown in Figure 10. The analysis of the obtained FLUE GAS temperature comparing it to the fuel reactor operating temperature is presented in Figure 11, an increase in the oxygen carrier inlet flow leans to an increase of the FLUE GAS outlet temperature; reaching its maximum point (834,02°C) corresponding to a molar flow of 6.782 kmol/h of Fe_2O_3. It is perceived that the temperature of the gas product does not decrease, meaning it has a higher temperature than the operating temperature of the fuel reactor; therefore, an analysis of the autothermal behavior of the process is feasible.

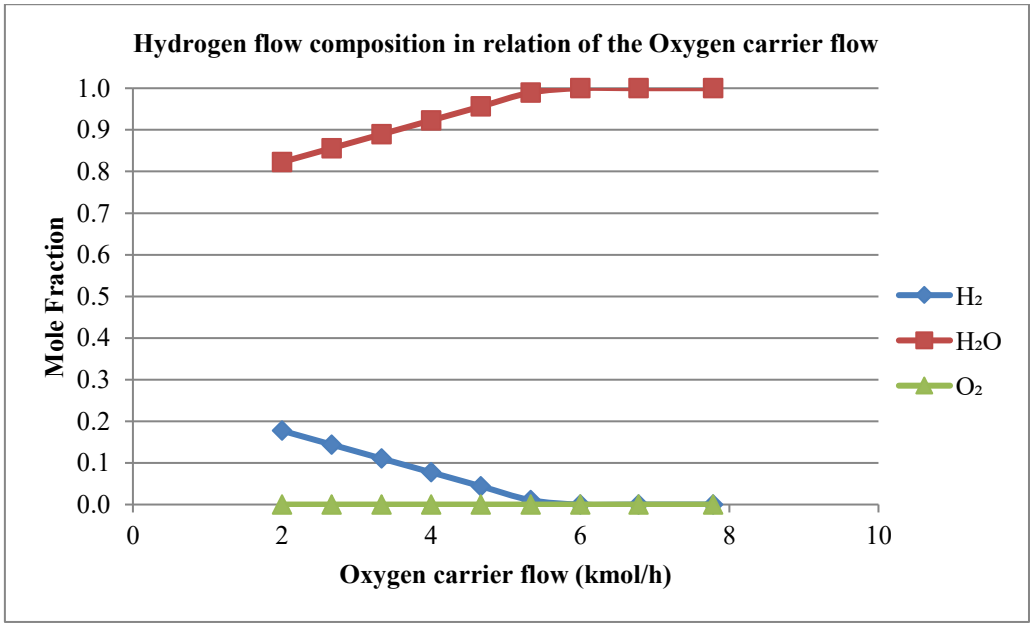

Figure 10. Composition of the hydrogen flow produced in relation to the increase in the feed molar flow of iron oxide.

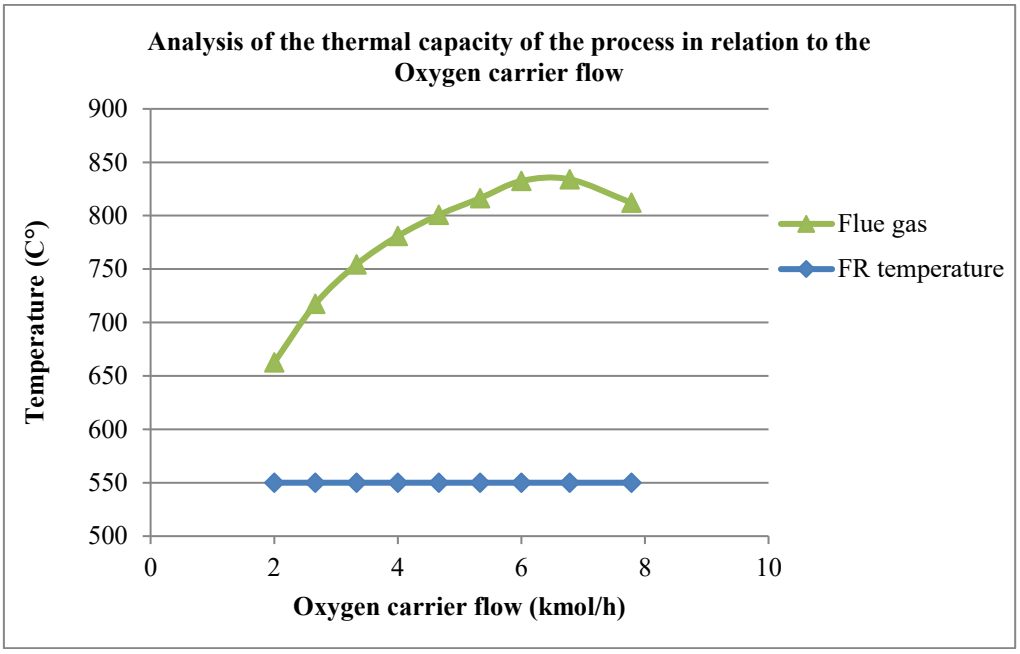

Figure 11. Process thermal analysis.

4. CONCLUSIONS

Based on the parametric analysis carried out, it is feasible to conclude that trying to achieve a high hydrogen molar composition at the steam reactor outlet flow decreases the purity regarding the SYNGAS flow at the fuel reactor outlet flow along with the autothermal capacity of the process.

As for the fuel reactor temperature, it was a parameter that presented lots of differences with respect to the output flows, showing the highest hydrogen molar composition, lowest SYNGAS purity along with the lowest autothermal capacity, at its critical point (750°C).Under the simulation parameters that were set, the Three Reactors Chemical Looping Hydrogen process presents a favorable autothermal capacity altering such parameters as the feed flows, an important characteristic in energy terms, since it can represent advantages in production costs.

5. ACKNOWLEDGEMENTS

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7. RESPONSIBILITY NOTICE

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