Simulation of an Integrated Gasification Combined Cycle with Chemical-Looping Combustion and CO_2 sequestration

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ABSTRACT

Chemical-looping combustion allows an integration of CO_2 capture in a thermal power plant without energy penalty; secondly, a less exergy destruction in the combustion chemical transformation is achieved, leading to a greater overall thermal efficiency. This paper focus on the study of the energetic performance of this concept of combustion in an integrated gasification combined cycle power plant when synthesis gas is used as fuel for the gas turbines. After thermodynamic modelling and optimization of some cycle parameters, the power plant performance is evaluated under diverse working conditions and compared to a conventional integrated gasification combined cycle with precombustion capture.

Energy savings in CO_2 capture and storage has been quantified. The overall efficiency increase is found to be significant and even notable, reaching values of around 7%. In order to analyze the influence of syngas composition on the results, different H_2-content fuels are considered.

KEYWORDS

Chemical-looping combustion, CO_2 capture and storage, Gas turbine system, Efficient power generation systems, Integrated gasification combined cycle, Synthesis gas.

INTRODUCTION

Since fossil fuels are widely used for power generation, the effort required to achieve a real reduction of greenhouse gas emissions is enormous. However, it is possible to attain a short and medium term substantial reduction through sequestration of the CO_2 produced in fuels oxidation. The carbon capture and storage (CCS) could thus facilitate the transition to the use of new sources of clean energy.

There are basically three ways to perform CO_2 capture in practice: a) "post-combustion" capture via amine chemical absorption [1,2]; b) "pre-combustion" capture, in the case of a previous fuel decarbonisation to a mixture of H_2 and CO_2 takes place [3]; c) "oxy-combustion", in which the fuel is burned into oxygen instead of into air [4], and then nearly pure CO_2 is obtained after condensation of water.
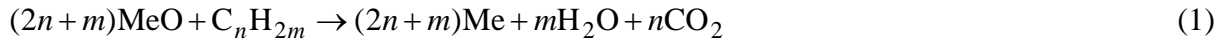
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The first two strategies, *i.e.*, pre- and post-combustion capture, involve a high energy penalty, since they are based on conventional separation techniques such as separation by membrane or absorption, adsorption and cryogenic separation methods. When pre-combustion is possible, as in integrated gasification combined cycles (IGCC), it is energetically cheaper due to the fact that CO_2 is less diluted in the fuel than it is in the flue gases; but energy consumption is still important. In the case of oxy-combustion, the energy penalty occurs in the air separation unit rather than in the separation of CO_2 from flue gases.

The alternative technique of chemical-looping combustion (CLC) was first proposed by [5], and afterwards several researchers have contributed to the development of this technology. Nevertheless, regarding CLC applied to gas turbine systems, most efforts have been dedicated to the study of methane as fuel [6,7,8], although alternative fuels such as methanol have been proposed as well [9]. This work focuses on the analysis of the energetic efficiency of a CLC gas turbine system fuelled by synthesis gas (syngas hereinafter), in the base of a comparison with a similar gas turbine system with conventional combustion. There is also prior work on energetic analysis of a CLC gas turbine system with syngas as fuel [10,11] but in both cases only the gas turbine cycle is analyzed instead of a combined gas-steam cycle power plant, and the energy savings in the capture of CO_2 are not quantified.

CHEMICAL-LOOPING COMBUSTION DESCRIPTION

The idea of the CLC system is illustrated in Fig. 1. The gaseous fuel is introduced into the reduction reactor and put in contact with an oxygen carrier, typically a metal oxide, here denoted generically as "MeO". The fuel is then oxidized and the metal oxide is reduced, reacting according to:

$$(2n+m)\text{MeO} + \text{C}_n\text{H}_{2m} \rightarrow (2n+m)\text{Me} + m\text{H}_2\text{O} + n\text{CO}_2 \tag{1}$$
$$\text{MeO} + \text{CO} \rightarrow \text{Me} + \text{CO}_2 \tag{2}$$

Thus, the output stream from reduction reactor contains a gaseous mixture of CO_2 and H_2O, so that the only non-condensable gas in that flow is CO_2. The reduced metal oxide "Me" is then transferred to the oxidation reactor where it is oxidized in presence of air, according to:

$$\text{Me} + \tfrac{1}{2}\text{O}_2 \rightarrow \text{MeO} \tag{3}$$

As a result, at the outlet of this reactor oxygen-depleted air is obtained, *i.e.*, mainly a stream of N_2 and O_2 in a certain proportion.
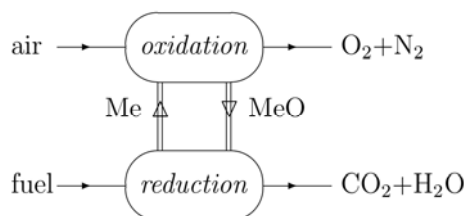

Figure 1. Chemical-looping combustion concept

The major advantage of CLC lies on the fact that the carbon dioxide that results from the fuel oxidation is not diluted with air or any other non-condensable gas, but is obtained in a

relatively pure form. This removes the need to invest energy for separation and the only energy needs for sequestration of CO_2 is that due to the compression up to the storage pressure. In addition, it is known that if a suitable oxygen carrier can be found in order to one or both of reactions (1),(2) become endothermic, the overall exergy destruction occurred in the oxidation of fuel is lower than in a conventional combustion [11], what leads to attaining a greater cycle thermal efficiency.

The CLC scheme in Fig. 1 can be implemented in practice in different ways, depending on the oxide's physical characteristics, the type of reactor and the operating conditions [6]. Typically fluidized-bed reactors are used, in which the metal oxides "float" as fine solid particles, guaranteeing a sufficient contact area for the chemical reactions with oxygen (oxidation) and with fuel (reduction) take place. It may be added some inert material or any catalyst to improve the physical properties and stability of the metal oxide particles and chemical kinetics, although this is not investigated in this work.

DESCRIPTION OF THE STUDY

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 compression up to storage pressure. A schematic flow diagram of the power plant is depicted in Fig. 2.

For the gasifier system, we have selected an entrained-flow slagging gasifier with convective cooler (Shell type). This option has been preferred as it is actually used to carry out a co-gasification of coal and biomass [12], and furthermore, regarding preliminary calculations, this is the gasifier that provides a better energetic performance. As a result, synthesis gas consisting mainly of CO and H_2, together with significant amounts of CO_2 and certain quantities of carbonyl sulphide (COS) and hydrogen sulphide (H_2S) is produced. As this chemical process is highly exothermic, the necessary cooling of the gasifier takes places with an important amount of steam generation. This raw syngas is taken to powder filtration, COS is hydrolysed to $CO_2 + H_2S$ and, after cooling, the sulphur is removed by chemical absorption.

Since temperature of flue gases at CLC outlet is high enough, a conventional three-pressure levels steam cycle with reheat has been implemented.

The overall energetic performance of such a power plant is evaluated. Different fuels have been tried in order to somehow evaluate the influence on syngas H_2-content in the thermal efficiency of the CLC system.

The CO_2 is sequestrated by condensation of water in that stream and compression up to storage pressure is carried out in two stages with intermediate cooling. Final pressure has been assumed 100 bar, so that storage or transport as high-density supercritical fluid is allowed. In addition, a small quantity of this quite pure CO_2 is reinjected in the cycle for fuel preparation instead of N_2. This is preferred, since residual N_2 contents in syngas are then reduced saving energy for compression.

To better illustrate its behaviour, the CLC-based gas turbine cycle has been separately represented in Fig 3. The main gas turbine is the one referred to as GT1, where depleted air is expanded from the oxidation pressure to around atmospheric pressure. In order to maximize the power production, two ideas for an optimized components configuration taken from previous works have been incorporated:

a) Introduction of an air pre-heater, in order to take advantage of the solids heat capacity to increase the air mass flow through GT1. This idea is proposed by [9].

b) The oxidation and reduction reaction must be pressure-linked due to the chemical looping followed by the oxygen carrier. As suggested by [10], a second gas turbine (GT2) is introduced to convert into work the pressurized CO_2 and H_2O mixture generated in the reduction reaction. This work is maximized by heating this stream from the higher temperature available heat source, the oxidation reactor.
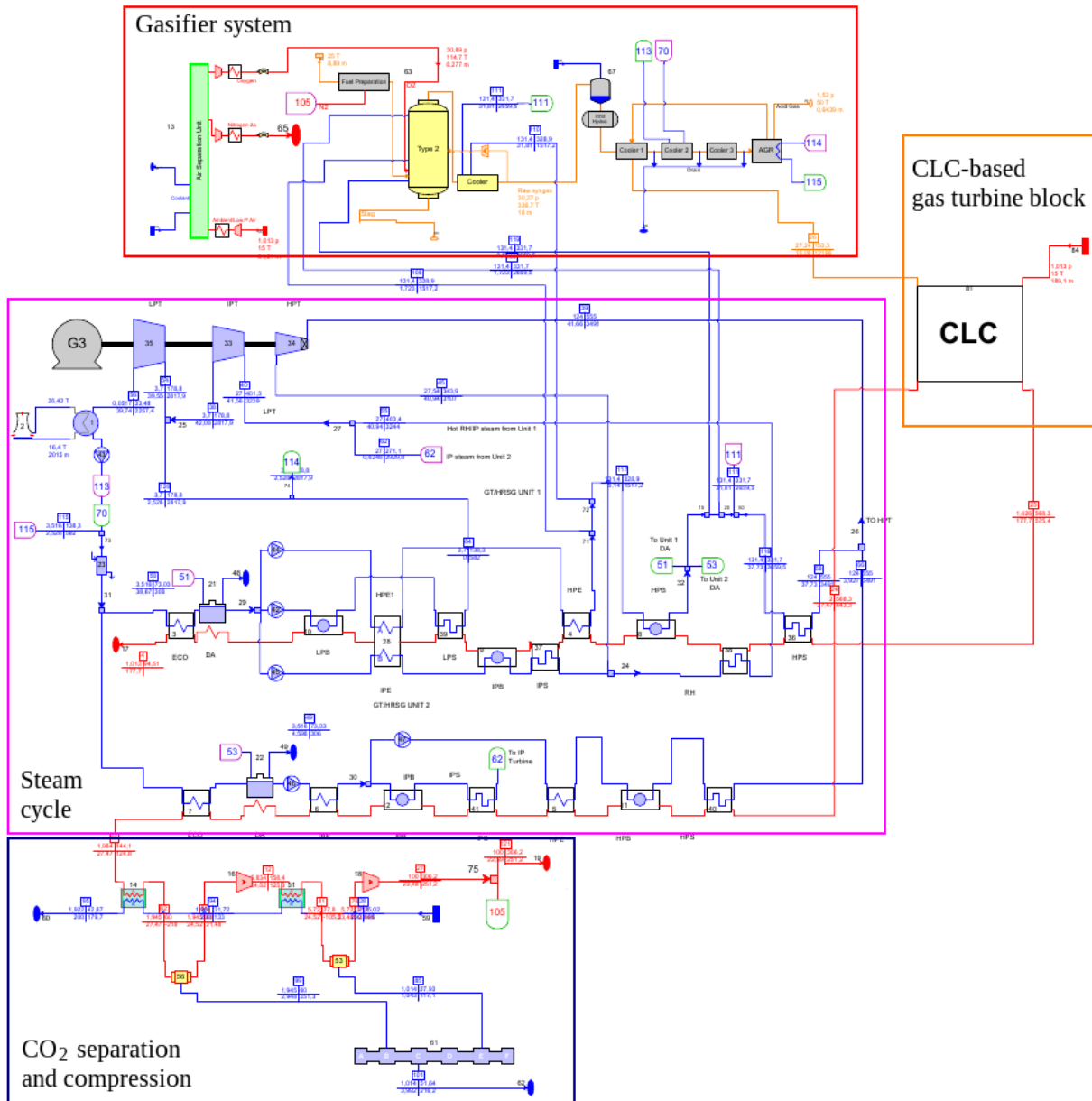

Figure 2. Integrated gasification combined cycle with chemical-loping combustion

As previously mentioned, if one or both of the reduction reactions (1),(2) is/are endothermic and they take place at low/medium temperature, then it would be possible to supply the needed heat from a medium temperature source, which in this case has been assumed to be the exhaust gases stream from the gas turbines. According to Hess' law, the amount of heat released in both reduction and oxidation reactions must be equal to the fuel's heat of combustion, which implies that the oxidation reaction presents a heat of reaction higher than

the conventional combustion. As a result, more heat is released at high temperature in comparison with a conventional combustion. CLC acts as a "chemical heat pump" transforming energy with lower exergy content into energy with higher exergy content. In other words, the overall exergy destruction due to irreversibility in chemical transformations involved is lower with CLC than with conventional combustion.
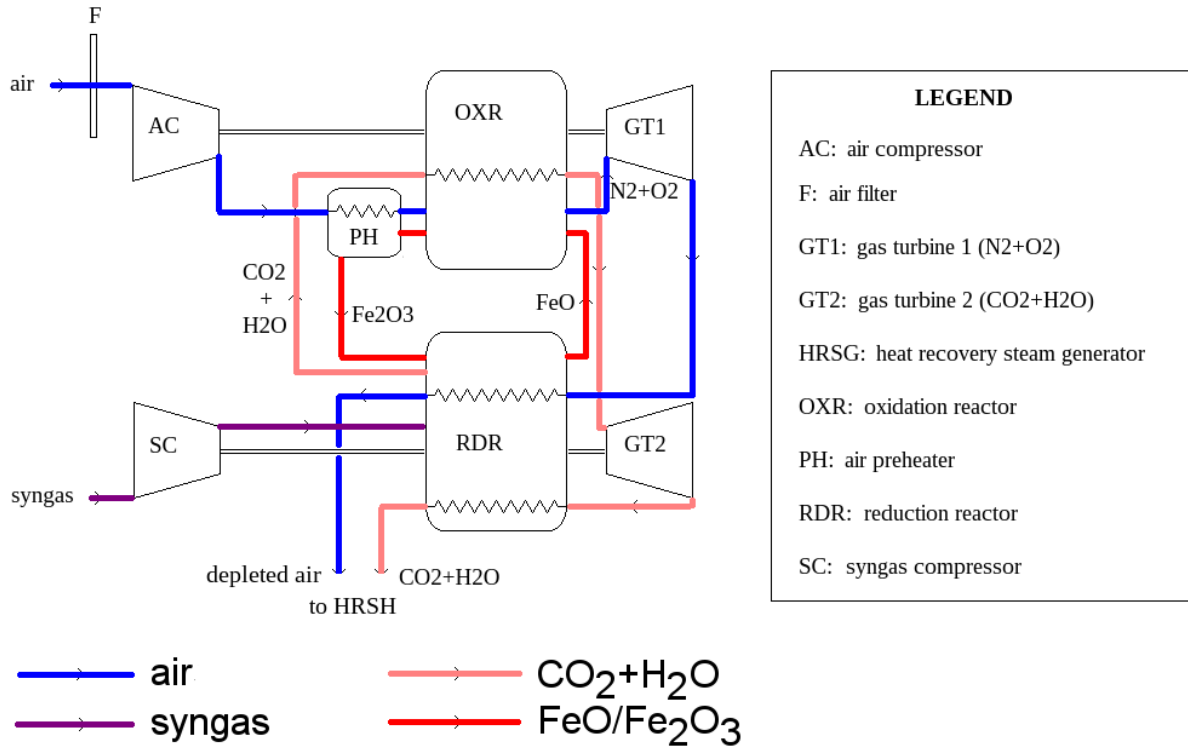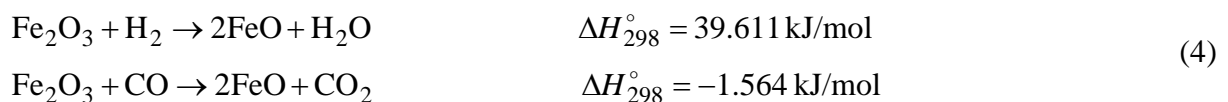

Figure 3. Diagram of CLC gas turbine cycle

Oxygen carrier

A few metal oxides have been proposed to act as oxygen carriers in CLC; typically nickel, iron, copper and manganese oxides, e.g. NiO/Ni, Fe_2O_3/Fe_3O_4, CuO/Cu, CuO/Cu_2O, Mn_2O_3/Mn_3O_4, *etc.* For the case of methane/natural gas as fuel, the first two options are the most frequently used. However, for the case syngas as fuel, which is the case in an IGCC, the main reactions occur with carbon monoxide and hydrogen. Only the pair F_2O_3/FeO from among those tested meets the requirement to provide an endothermic chemical transformation at the reduction reactor:

$$Fe_2O_3 + H_2 \rightarrow 2FeO + H_2O \qquad \Delta H^{\circ}_{298} = 39.611\,\text{kJ/mol}$$
$$Fe_2O_3 + CO \rightarrow 2FeO + CO_2 \qquad \Delta H^{\circ}_{298} = -1.564\,\text{kJ/mol}$$

(4)

In (4) ΔH°_{298} represents the standard enthalpy of reaction at 25 °C (298.15 K).

In the oxidation reactor, the resulting chemical reaction to close the loop is:

$$2FeO + \tfrac{1}{2}O_2 \rightarrow Fe_2O_3 \qquad \Delta H^{\circ}_{298} = -281.438\,\text{kJ/mol} \qquad (5)$$
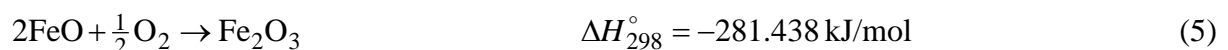
This value is to be compared with the lower heating value (LHV) of H_2 (241.827 kJ/mol) and CO (283.002 kJ/mol). Therefore, the pair F_2O_3/FeO has been selected as oxygen carrier for this study.

Inert material has been reported necessary to achieve the appropriate physical characteristics of the oxides solid particles in relation with their stability facing high changes of temperature. Among the several oxides that have been proposed, ZrO_2 has been selected since [11] referred that YSZ (ZrO_2 stabilized by yttria) seems to act as a catalyst in reactions (4). The ratio between inert material and oxygen carrier has been assumed 0.27 mol ZrO_2 per mol FeO, an intermediate value between those given in [10] and [6] (0.2 and 0.34 respectively). ZrO_2 does not take part in any reaction and just acts as heat transporter between the two reactors.

Fuels

Two different fuels have been considered for this study, in order to compare the influence of the syngas hydrogen content in the CLC performance. Waste biomass substrates of different origin are chosen for that purpose. Since biomass combustion is considered CO_2-neutral itself, the net CO_2 emissions would be negative when CO_2 sequestration is carried out in an only-biomass-fuelled power plant. In practice, biomass can be used in co-gasification with coal in a significant proportion [13]. The ultimate analysis compositions and LHV of the substrates under study is given in Tab. 1.

Table 1. Composition and LHV of substrates under study

Substrate	Ultimate analysis (% weight)								LHV
	O	N	H	C	S	Cl	ash	moisture	(kJ/kg)
wood waste	33.51	0.69	5.19	44.02	0.07	0.11	6.21	10.20	15 665
rice straw	33.92	0.80	4.79	35.20	0.17	0.00	17.19	7.93	15 809

Methodology

The power plant represented in Fig. 2 has been simulated with THERMOFLEX software [14], by Thermoflow Inc. It is a modular program that allows the user to assemble a power plant model from different industrial components. Despite THERMOFLEX is a powerful comprehensive, stand-alone tool, it does not provide a particular module for a CLC-based system. A specific interface has been programmed in order to generate a bidirectional runtime link between THERMOFLEX and a user-defined module for modelling CLC. For that purpose, a thermodynamic model for the simulation of the cycle shown in Fig. 3 has been developed relying on PATITUG library, an own software for the analysis of thermodynamic cycles developed by the Applied Thermodynamics Group of the Technical University of Madrid. PATITUG is oriented to the design, analysis and optimization of thermodynamic cycles and their elements, and consists of a number of modules that can be conveniently assembled to calculate thermodynamic properties of all fluid streams involved in a cycle. Different models are included to handle pure substances, mixtures and chemical transformations. A variety of equations of state, as ideal gas, virial gas, Lee-Kesler equation and IAPWS-IF97 for water, and different expressions for the specific heat at nil pressure can be selected. Recent published works have been carried out by the Thermodynamics group of professors at ETSII-UPM in base of this library. A deeper description of PATITUG can be found in [15,16].

In this simulations the following equations of state have been selected for gases:

- IAPWS-IF97 equation of state for water where temperature exceeds the water boiling temperature at that pressure.
- The virial gas equation of state truncated after the second term for gaseous water in the cycle points where temperature is below the water boiling temperature at that pressure (but exceeds the water boiling temperature at water's partial pressure in that point, so we find water in gaseous state) and for all non-condensable gases when their specific volume is at least twice its critical specific volume.
- The Lee-Kesler's equation of state for the rest of the cases, *i.e.* non-condensable gases with specific volume is lower than twice its critical specific volume, and supercritical fluids.

For solids, the functional dependence of molar enthalpy and entropy with temperature has been taken from the NIST Chemistry Webbook [17]. A slight correction to account the variation of enthalpy with pressure has been applied.

Cycle optimization

Some parameters dictate the thermodynamic conditions in all streams of the CLC cycle (Fig. 2). The governing one is the turbine inlet temperature (TIT), *i.e.* the temperature at which the oxidation reaction takes place. As it is well known, TIT should be as high as possible as long as the technical requirements in gas turbines and reactors are satisfied and the oxygen carrier particles are found to be physically stable, in order to increase the cycle's thermal efficiency. The other key parameters to be determined by analysis and optimization are temperature at the reduction reactor T_{red} and pressure at both reactors p_r.

It is supposed that chemical equilibrium is achieved at the reduction reactor, implicitly assuming that the reactants residence time is high enough in comparison with the characteristic chemical kinetics times. Catalysis may be needed. Equilibrium calculations lead to conversion ratios (CR) of H_2 and CO in reactions (4) as a function of temperature T_{red}. Conversion ratios are plotted in Figure 4[*]. Since the first of transformations (4) is endothermic, we expected from Le Châtelier's principle H_2's CR to increase with reduction temperature. It can be seen that for a temperature of around 800 K, H_2's CR reaches 99% and CO's CR is located at about 99.7%. For this reason, approximately 0.55% of the fuel's LHV is lost due to incomplete combustion. However, this effect is more than offset by the lower exergy destruction in the combustion process, as shown in a following section.

However, temperature at the reduction reactor is limited because this reactor must take the required heat from the flue gas streams at the gas turbines outlets. In other words, the temperature of the reactor reduction must be high enough for the reaction to occur in a high percentage so that almost no fuel is lost, but at the same time it should be sufficiently low compared with the gas temperature at the outlet of the turbine in order to allow the recovery of the necessary heat. Also the compression ratio of the air compressor has an influence on the optimal reduction temperature. For a higher compression ratio, the air temperature after the compression stage would increase, affecting the energy balances at reactors too, as we will discuss in a following section.

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[*] Since H_2O content for both syngas compositions considered, both curves for H_2's CR result to be completely indistinguishable. Only one of them is printed.
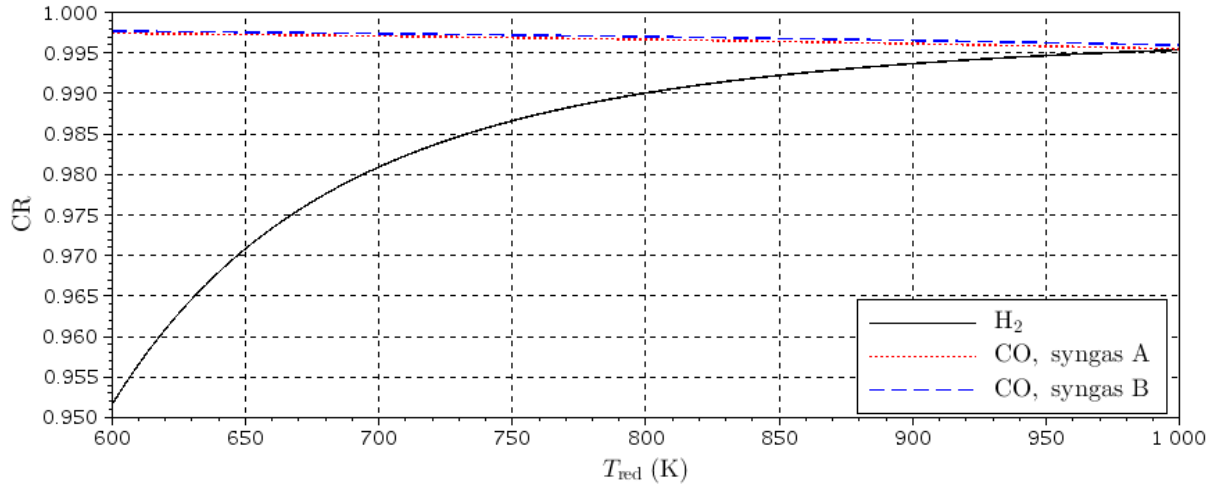
Figure 4. Conversion ratios of H_2 and CO as a function of T_{red}.

Simulations have been performed for a set of TIT in the range going from 1400 K to 1550 K and p_r in the range 15-30 bar. For each pair of TIT and p_r an optimization scheme is applied in order to find the optimal value of T_{red}, considering the overall IGCC power plant thermal efficiency as objective function.

Energy yields

The energy yields, together with the overall thermal efficiency of the CLC-IGCC have been calculated for a large/medium size power plant:

$$\eta_{th} = \frac{W_{net}}{m_{bm}LHV_{bm}} = \frac{W_{CLC-GT} + W_{ST} - W_{aux}}{m_{bm}LHV_{bm}} \tag{6}$$

Here, W_{CLC-GT} stands for the gross power generated by CLC gas turbines, W_{ST} is the steam turbines gross power output, W_{aux} represents the auxiliary power consumption (gasifier, acid gas removal, CO_2 compression, etc.), W_{net} is the net power porduction and m_{bm} is the biomass flow rate to gasifier.

RESULTS

Gasification

The final compositions and LHV of the resulting syngases (after gas cleaning, COS hydrolysis and H_2S removal) are presented in Tab. 2.

Table 2. Composition and LHV of resulting synthesis gases

Substrate	Label	Composition (volume %)						LHV (kJ/kg)
		CO	CO₂	H₂	H₂O	N₂	Ar	
wood waste	*syngas A*	46.90	18.45	26.02	0.02	8.09	0.53	8 069
rice straw	*syngas B*	46.47	8.23	37.19	0.03	7.84	0.24	11 257

While CO content of both syngases is quite similar, significant difference in hydrogen and CO_2 amount is found. Since hydrogen oxidation to water is the endothermic reaction at the fuel reactor, a higher efficiency of the CLC system is expectable when hydrogen content

increases. In addition, for a same mass flow of syngas, power should increase due to its higher LHV. This will be quantified in a following section.

Cycle optimization

The overall power plant thermal efficiency dependence with the CLC reactors pressure is shown in Fig. 5 and 6 for each of the several TIT values tried. Similar curves for the optimal temperature at the reduction reactor can be seen in Fig. 7 and 8.

The figures reveal a somewhat capricious behaviour that may be worth to discuss. As it was mentioned before, the optimal reduction temperature is the highest temperature that makes possible to satisfy the energy balance in the reactor, considering that the hydrogen oxidation needs to take some heat from its surroundings. In principle, to increase the pressure ratio makes the gas turbines outlet temperature after expansion go down. For this reason, at low pressure ratios the optimal T_{red} is reduced with a pressure ratio increase so that the reactor is able to take sufficient heat from the exhaust gas streams outgoing the turbines. However, there is another opposite effect. The outlet temperature of the compressors will also increase with pressure ratio. These temperatures are the fuel inlet temperature to the reduction reactor and the air compressor outlet temperature. Thus, after a heat exchange in the preheater (see Fig. 3) the solids are found hotter when entering into the reactor as well. In summary, there are two opposite effects happening when the reactors pressure is increased:
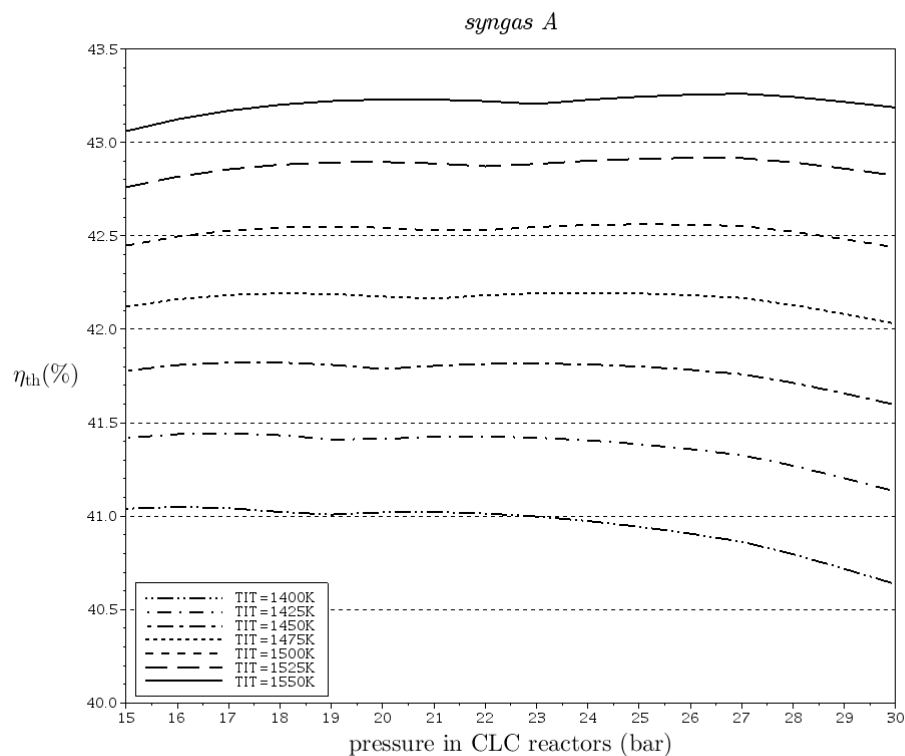

Figure 5. Thermal efficiency of the CLC-IGCC power plant for wood waste as fuel (*syngas A*)
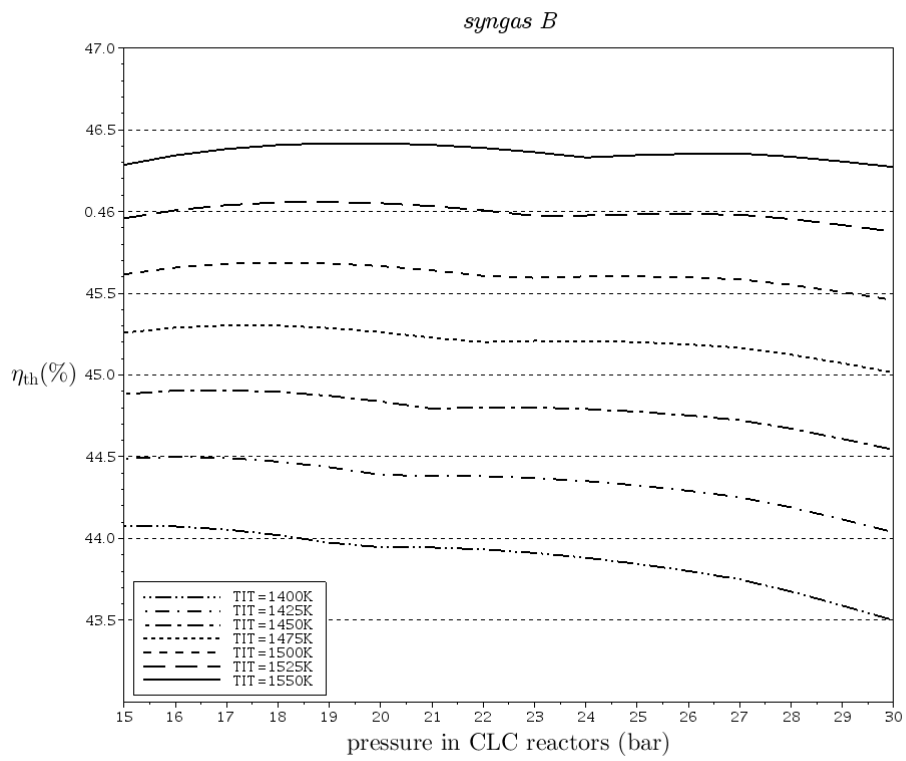
Figure 6. Thermal efficiency of the CLC-IGCC power plant for rice straw as fuel (*syngas B*)
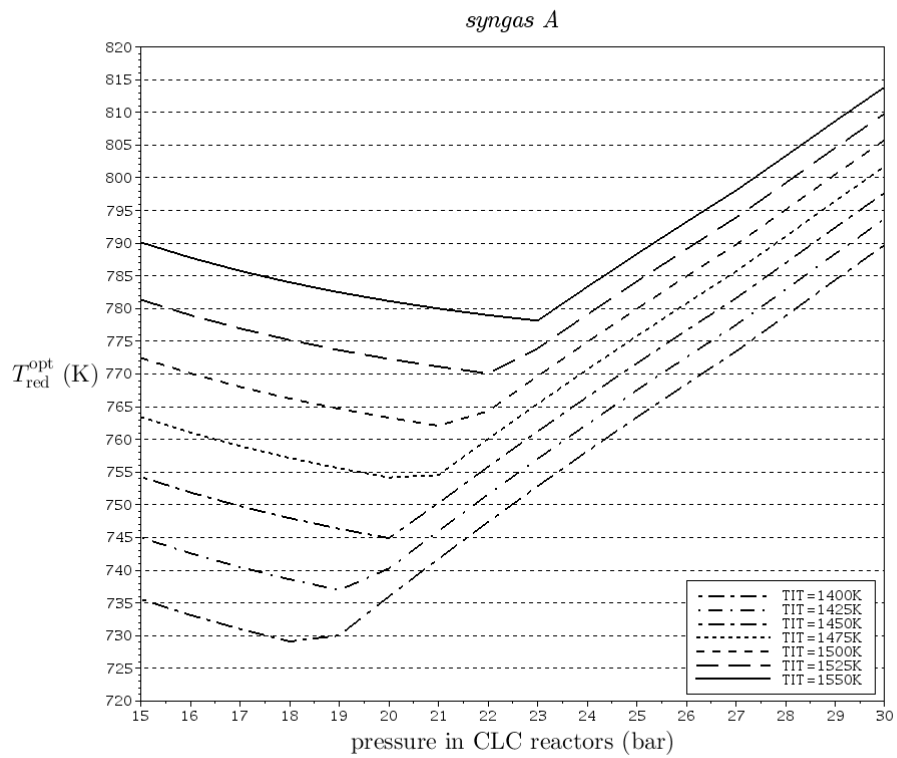

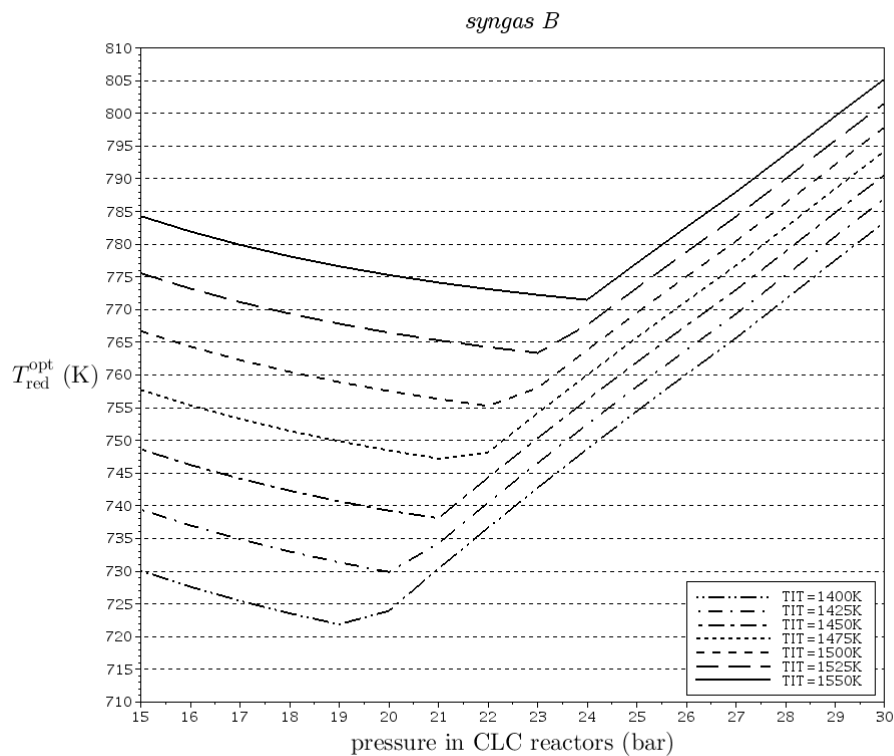Figure 7. Optimal temperature at reduction reactor for s*yngas A*

Figure 8. Optimal temperature at reduction reactor for s*yngas B*

   a) Lower temperature of gas streams at the outlet of the gas turbines
   b) Reduced need for heat in the reactor, since input streams enter with a higher temperature

As a consequence, if pressure ratio continues to increase b)-effect begins to dominate against a)-effect, and at some particular pressure the heat needed by the reactor is decreased to a point that it can be provided only by the $CO_2 + H_2O$ stream, leaving intact the flow of air. This can be seen as a typical "heat pump effect", *i.e.*, the energy introduced in the cycle as mechanical power in the air compressor comes back somehow as heat provided to reduction reactor. Due to the complex heat coupling of streams and reactors in the CLC cycle, this allows T_{red} to reverse its trend and begin to increase with pressure ratio.

This phenomenon is also revealed in the thermal efficiency plots. Instead of curves with a maximum, which would be found for a conventional combined cycle, for this CLC system we get curves with two local maxima, of similar values. Consequently a good thermal efficiency almost constant is achieved along a quite wide range of pressure ratios.

Overall energy yields

In this section we present the power plant energy yields for a reference case defined by TIT = 1500 K (1226.85 °C) and p_r = 20 bar for both fuels under study. Tab. 3 shows a summary of data:

Table 3. Energy yields of CLC-IGCC power plant. Reference case TIT = 1500 K; p_r = 20 bar.

Substrate	Label	m_{bm} (kg/s)	W_{CLC-GT} (MW)	W_{ST} (MW)	W_{aux} (MW)	W_{net} (MW)	η_{th} (%)
wood waste	*syngas A*	33.23	143.29	123.19	44.02	**221.46**	**42.54**
rice straw	*syngas B*	33.23	152.83	132.01	44.95	**239.90**	**45.67**

A detailed breakdown of the auxiliaries energy consumption for the particular case of rice straw with TIT and p_r of the reference case is given in Fig. 9. It can be seen that the major contributions come from the air separation unit (ASU), 40.4 %, and the CO_2 compressors, 24,06 % and 9.29 %.
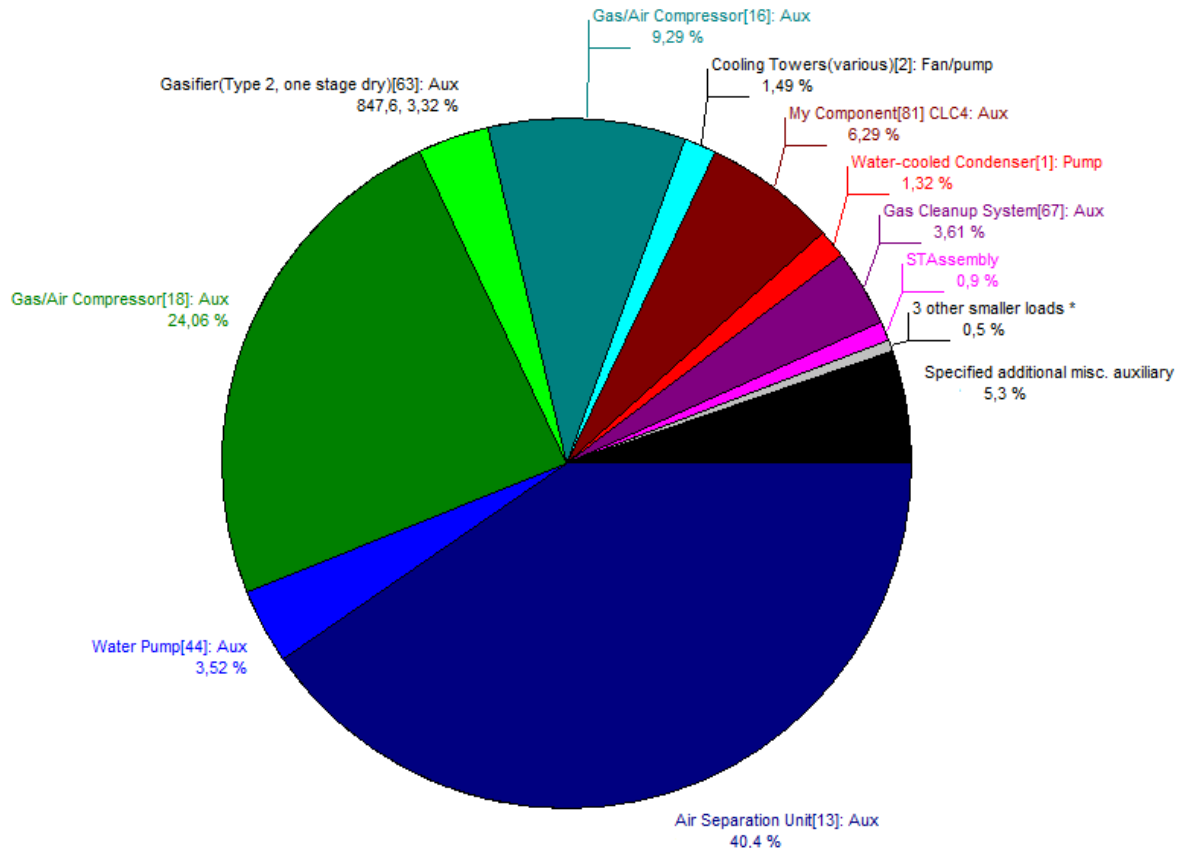

Figure 9. Breakdown of auxiliaries' energy consumption for a CLC-IGCC fuelled rice straw. Reference case TIT = 1500 K; p_r = 20 bar.

Comparison with conventional CLC

The main interest of this study lies on a comparison between the CLC-IGCC system modelled here and a conventional IGCC power plant with precombustion CO_2 capture, in order to quantify the possible energetic savings that could be attained by the alternative CLC technique.

Previous works [18,19] provide an extensive work in simulations on IGCC fuelled by biomass substrates. In particular, results for simulations on the gasification of the same substrates as here are given. Obviously, that is why we chose them for this study. The comparison of the IGCC power plant overall thermal efficiency is summarized in Tab. 4. We include in the table the working conditions of the gas turbines as well.

Even in the first case, where TIT is significantly higher than in the calculations carried out here, it is found a notable increase of the overall thermal efficiency of the process (~ 4%), considering all the stages of biomass-to-power conversion: gasification, gas turbines, steam cycle and auxiliaries and CO_2 separation and compression up to the storage pressure consumption. In the case with a lower amount of C present in fuel, the efficiency increase was expected to be somewhat higher, due to the fact that CO_2 compression power is almost the same in conventional and CLC cases, so in relative terms the energy savings on CO_2 separation would be increased. The result for this case is quite spectacular: ~ 7%), although it is true that the TIT used in the CLC model is about 80 degrees hotter. Nevertheless, the energetic performance of the IGCC ensemble is found to be significantly improved in all cases under study.

Table 4. Comparison of thermal efficiency and operating conditions between CLC and conventional IGCC

Substrate	IGCC type	Gas turbine	TIT (K)	p_r (bar)	η_{th} (%)
wood waste	*conventional*[†]	GE 9371 FB	1700	18.2	**38.4**
	CLC	*modelled*	1500	20.0	**42.54**
rice straw	*conventional*[‡]	Siemens SGT5-2000E	1422	11.8	**38.5**
	CLC	*modelled*	1500	20.0	**45.67**

CONCLUSIONS AND FINAL REMARKS

In this paper, an analysis on the energetic performance of an IGCC power plant with chemical-looping combustion with CO_2 sequestration and storage is presented. Two different substrates for gasification are analyzed in order to quantify the influence of syngas composition on results and iron oxides are used as oxygen carrier. The overall thermal efficiency of the power plant has been calculated, after determining the governing working conditions of the CLC cycle through an optimization procedure. A final comparison with similar systems with conventional combustion is given.

The main conclusions of this work can be summarized as follows:
- According to chemical equilibrium calculations, a temperature in the reduction reactor in a range 720-815 K ensures a high degree of conversion ratio in the chemical transformation, but also allowing the possibility of taking the required heat from the flue gases at the gas turbines' outlets. The more hydrogen content in the fuel, the more the reduction temperature should be lowered in order to achieve the energy balance.
- The optimal pressure in CLC reactors has found to be around 20 bar, which is more or less constant within a wide range of variation due to the particular behaviour of the heat flows and balances in the reactors.
- The increase of thermal efficiency has found to be very important. When the energy cost of CO_2 through chemical processes is taken into account, the overall thermal efficiency increase reaches significant values: 4 and 7 percentage points, depending on the total amount of carbon contained in the synthesis gas. The energy savings are higher for the fuel with larger amount of hydrogen.

[†] reference [19]
[‡] reference [18]

Although an important effort on research and significant technical development are needed in order to achieve the required maturity level for an industrial use, results show that the CLC cycle offers a great potential for efficient power generation and nearly zero or even negative CO_2 emissions. In a context of real urgency to reduce green house gas emissions, this work is intended to contribute to the conceptual development of efficient alternative power generation systems.

NOMENCLATURE

ASU: air separation unit
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
ΔH_{298}°: standard enthalpy of formation (kJ/mol)

T_{red}:   temperature at reduction reactor
p_r: pressure at CLC reactors
η_{th}:   power plant thermal efficiency
$W_{CLC\text{-}GT}$: gross gas turbines power
W_{ST}:   gross power by the steam turbines
W_{aux}:   auxiliaries power consumption
m_{bm}:   biomass flow rate (kg/s)

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