Optimization of a syngas purification line and CO₂ capture systems integrated with a small-scale up-draft gasifier

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Abstract:
With the aim of using small-size up-draft gasifiers for producing both clean energy and hydrogen, the gasification process has to be integrated with syngas purification and CO₂ capture systems. In this context, dedicated simulation models for performance evaluation of these systems have been developed using the Aspen-Plus software. In particular, the models have been adapted to evaluate the performance of a pilot gasification and gas treatment plant for hydrogen production, built at the Sotacarbo Research Centre in Sardinia. The syngas purification and CO₂ capture systems of the Sotacarbo plant include: a wet scrubber, two desulfurization systems, an electrostatic precipitator, an electric heater, a shift conversion section, a CO₂ removal system and a hydrogen production section. The primary aim of the study is to optimize the integration between the purification line, the CO₂ capture system and the gasifier as a whole, in order to increase the performance of the overall system. The molar ratio steam/CO in the shift reactor has been identified as a key parameter, both for shift conversion section performance and for the overall system energy balance.

Keywords:
Syngas from coal gasification, purification line, CO₂ capture, system integration.

1. Introduction

Coal gasification represents a competitive solution for both power generation [1] and production of chemicals or clean fuels, as, for example, hydrogen [2]. In fact, coal gasification processes integrated with high efficiency energy conversion systems, such as combined cycle based on new generation gas turbine [3] or high temperature fuel cells [4], will allow, in the future, to achieve efficiencies as high as 50-55%. Currently, gasification processes are mainly used in large-scale power plants, however, R&D activities are now also focussing on small- and medium-scale applications, based on fixed- and fluidized-bed gasifiers, using air as oxidant instead of pure oxygen [5-7]. As a consequence, in a short-term future, the use of syngas to co-produce electrical energy, hydrogen, other sustainable fuels and chemicals, will assume a large interest, even for medium- and small-scale industrial applications.

In this framework, a flexible and fully equipped pilot gasification platform was built at the Sotacarbo Research Center in Carbonia (South West Sardinia, Italy). The platform includes a 1.3 m diameter demonstration gasifier (5 MWth) and a 0.3 m diameter pilot unit (200 kWth). Both gasifiers are based on an air-blown, up-draft and fixed-bed technology, suitable to be fed with both coal (including high sulphur low rank coals) and biomass [8]. Among fixed-bed gasification processes, up-draft has the advantages of high reliability, high efficiency, low specific emissions and feedstock flexibility, but typically produces a syngas with a high tar content [9]. In addition, process handling is easier [10], the counter-current fluid dynamics ensures a higher efficiency than other gasification technologies [11] and the air-blown gasification is more available and simple than the oxygen-blown one [12]. As a consequence, this gasification process assumes particular commercial interest in the field of medium and small scale industrial applications. A detailed description of the
Sotacarbo pilot platform can be found in [13]. The Sotacarbo pilot platform was built with a simple and flexible layout to test different plant solutions at different operating conditions, performing more than 1700 h of experimental tests since June 2008.

In this framework, the Department of Mechanical, Chemical and Materials Engineering (DIMCM) of the University of Cagliari and Sotacarbo are engaged in a long time cooperation concerning small- and medium-scale coal gasification systems. Within this cooperation, DIMCM has developed a zero-dimensional, steady-state, fixed-bed gasification model with the aims to investigate the gasification process in different operating conditions and to support the design of experiments in pilot gasification units [14]. In particular, it was applied to design the recent experimental campaigns performed in the Sotacarbo gasification pilot plant.

The pilot unit gasifier was integrated with a syngas treatment line composed by a purification system and a CO₂ capture section, to allow hydrogen and electrical energy production. In particular, the syngas treatment line includes a hot-gas desulfurization process, a water gas shift (WGS) section, a CO₂ removal system and a hydrogen separation section (PSA). During experimental campaigns the syngas treatment line has allowed to test and characterize some gas treatment process and materials (solvents, sorbents, catalysts,...) for syngas desulphuration, water gas shift, CO₂ removal and hydrogen production [13]. As a matter of fact, the syngas treatment line is characterized by a simple and flexible layout, but, in the future, with the aim of integrating with commercial small-size gasification units, the energy balance of the overall system should be optimized.

In this context, the DIMCM has developed several simulation models for evaluating the performance of the syngas purification and of CO₂ capture systems installed downstream of the Sotacarbo pilot platform, with the aim of optimizing the integration between the purification line, the CO₂ capture system and the plant as a whole. The simulation models were developed through the Aspen Plus commercial software.

2. Gas purification and CO₂ capture system

The following is a brief description of the complex gas treatment and conditioning system installed downstream of the Sotacarbo pilot gasifier, composed by two main sections for syngas purification and CO₂ capture.

2.1. Description of the system

The syngas produced by the gasification section is sent to a first purification section which includes:

• a wet scrubber;
• a cold-gas desulfurization process;
• an electrostatic precipitator (ESP).

In the wet scrubber, a water flow reduces the syngas temperature from the gasifier exit temperature (between 150 to 300 °C, depending on the operating conditions and coal rank) to 50 °C. The wet scrubber also operates a first separation of dust and tar, that are almost completely removed in the following ESP. The purification line also includes a cold desulphurisation section, based on sodium hydroxide as a solvent, that reduces the concentration of sulfur compounds before entering the ESP. Downstream of the ESP the syngas purification process is divided into two lines: a power line that, through a further cold-gas desulphurisation section, directly feeds an internal combustion engine (ICE), and a hydrogen production line. The latter is the object of the present study and includes:

• a fan;
• an electric heater;
a hot-gas desulfurization process;
• two CO shift conversion reactors in series;
• two coolers before CO₂ removal;
• a two-stage CO₂ removal system;
• a compressor;
• a hydrogen separation section (PSA).

Figure 3.1 shows a simplified scheme of the gas purification and CO₂ capture system. Due the relative simplicity of the entire system lay-out the opportunity of resorting to a pinch-technology analysis was not considered.

Currently only a small portion (about 20-25 Nm³/h) of the overall syngas flow (70-80 Nm³/h) can be sent to the hydrogen production line. The fan allows to overcome the pressure drop along the first purification line. Downstream of the fan, an electric heater increases syngas temperature from the fan exit temperature (slightly higher than 50 °C, due to the low compression ratio) to the hot-gas desulphurization temperature (typically between 300-500 °C). The hot-gas desulphurization reactor is composed by three sections. In the first one COS is converted into H₂S through a catalytic filter, based on Ni-MoO₃/Al₂O₃. The latter sections are composed by two absorbers, equipped with zinc oxide (ZnO) sorbents, that react with the H₂S forming zinc sulfide (ZnS) and water vapour. These systems allow to reduce H₂S concentration in the syngas to less than 10 ppm, before the Water Gas Shift (WGS) section.

The shift conversion of CO takes place in two reactors: the high temperature (HT) one operating typically between 350 and 450 °C and the low temperature (LT) one operating at about 250 °C. Upstream of each reactor a steam injection is provided to increases the H₂O/CO molar ratio in the syngas up to a suitable value. Downstream of the HT reactor the syngas flow is splitted in two streams. A first one directly feeds the LT shift reactor, whereas the second one feeds the intermediate CO₂ removal reactor, which operates at a nearly ambient temperature (about 35 °C). The decarbonized syngas feeds the LT shift reactor after being mixed with the other syngas stream. The two flow rates are determined imposing the desired temperature at the inlet of the LT shift reactor. Downstream of the WGS section the syngas enters into the final CO₂ removal system, still operating at a nearly ambient temperature, in order to reach the desired CO₂ removal efficiency. As known, the WGS process is favoured by increasing the H₂O/CO molar ratio and by decreasing the operating temperature, being the CO shift an exothermic reaction, while the operating pressure is irrelevant since moles number remains practically unchanged during reaction. The operating
temperature of the HT reactor does not allow a high CO conversion, but it allows to accelerate the shift reaction. The required process steam is produced through a boiler fuelled by LPG.

The CO\textsubscript{2} capture process takes place in two bubbling reactors in which the syngas is injected through 40 diffusers based on ceramic membranes. The system operates at a pressure slightly higher than the atmospheric one and at a temperature of about 35 °C. The CO\textsubscript{2} absorption takes place through a solvent based on a mixture of water and monoethanolamine (MEA). The CO\textsubscript{2} capture system also performs the amine regeneration, through a column equipped with an electric reboiler (which operates in the range 120-150 °C), a condenser and a mist separator to split the CO\textsubscript{2} from residual water and solvent vapours. The CO\textsubscript{2} removal section is also equipped with some heat exchangers for preheating the CO\textsubscript{2}-rich solvent from the absorber and for cooling the CO\textsubscript{2}-lean solvent from the regenerator. Finally, downstream of the decarbonization section the syngas enters the hydrogen separation section, based on the PSA (Pressure Swing Adsorption) technology, which is widespread in industrial processes thanks to its low cost. In particular, the installed system is a PSA molecular sieves based on the simple two-stage. The PSA system operates at about 5-6 bar and therefore a compressor is installed downstream of the CO\textsubscript{2} removal section.

The main purpose of this study is to check the feasibility of the integration between different sections of the syngas purification and decarbonization line, in order to optimize the energy balance. In fact, the current layout is simple and flexible, with heating and cooling flows not integrated, in order to reduce equipment cost and to provide a greater flexibility to the plant experimental campaigns.

### 2.2. Performance analysis of the system

The process simulation models developed at the DIMCM were used to evaluate main performance of syngas purification and CO\textsubscript{2} removal systems integrated with the Sotacarbo pilot plant. The investigation was carried out considering a syngas obtained by the gasification process of the Usibelli coal. The main characteristics of Usibelli coal and syngas, determined during experimental campaigns, are reported in Table 1.

<table>
<thead>
<tr>
<th>Proximate Analysis (%wt)</th>
<th>Syngas characteristics</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fixed carbon</td>
<td>31.33</td>
</tr>
<tr>
<td>Volatile matter</td>
<td>41.00</td>
</tr>
<tr>
<td>Ash</td>
<td>10.02</td>
</tr>
<tr>
<td>Moisture</td>
<td>17.64</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Ultimate Analysis (%wt)</th>
<th>Syngas composition (%vol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total carbon</td>
<td>CO</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>CO\textsubscript{2}</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>H\textsubscript{2}</td>
</tr>
<tr>
<td>Sulphur</td>
<td>N\textsubscript{2}</td>
</tr>
<tr>
<td>Oxygen</td>
<td>CH\textsubscript{4}</td>
</tr>
<tr>
<td>Ash</td>
<td>H\textsubscript{2}S</td>
</tr>
<tr>
<td>Moisture</td>
<td>COS</td>
</tr>
<tr>
<td>LHV</td>
<td>CH\textsubscript{4}</td>
</tr>
<tr>
<td>LHV</td>
<td>H\textsubscript{2}O</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>LHV analysis (MJ/kg)</th>
<th>Syngas composition (%vol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>O\textsubscript{2}</td>
<td>48.56</td>
</tr>
<tr>
<td>H\textsubscript{2}O</td>
<td>5.96</td>
</tr>
</tbody>
</table>

Table 1 shows proximate, ultimate and thermal analysis of Usibelli coal and mass flow, temperature, pressure, molar composition and lower heating value of the syngas on dry basis. During experimental campaigns, syngas composition was measured on a dry basis and, as a consequence, mole fraction of steam was not known. Therefore starting from syngas composition,
coal rank and gasification parameters, a mole fraction of steam in the syngas equal to 0.07 was imposed. However, the steam concentration influences only partially the performance of the system, since, in the wet scrubber, the syngas is cooled to 50 °C and saturated with water. Consequently, the steam molar fraction in the syngas just influences the water flow rate feeding the wet scrubber, also influenced by the syngas temperature.

The model for simulating the purification line does not analyze in detail the cold-gas desulphurisation section and the electrostatic precipitator, since their energy consumption is slight. Therefore the model assumes the purified syngas free of ash and with a concentration of sulfur compounds amounting to some tens of ppm. Experimental data showed that the pressure drop of this first purification line is about 10 kPa. However, the simulation procedure considers that the overall syngas produced by the gasification section (80 kg/h) can be sent to the hydrogen line, whereas in the Sotacarbo plant the line is designed to treat not more than 20-25 kg/h of syngas.

The fan overcomes the pressure drop along the first purification line providing a slight syngas pressure increase from 0.13 to 0.14 MPa with a power requirement of about 300 W. An electric heater is installed downstream of the fan to increase the syngas temperature up to 350 °C, suitable for hot-gas desulfurization process. An electric power of 8.5 kW is required to heat the syngas from about 55 °C up to 350 °C. Downstream of the desulfurizer the syngas enters into the HT-WGS reactor, where a suitable amount of steam is required to enhance the CO shift conversion reaction.

The stoichiometric value of the steam/CO molar ratio is equal to 1, but values between 1.5 and 3 are commonly adopted for enhancing the reaction. In the purification line, downstream of the desulfurizer the syngas is characterized by a steam/CO molar ratio equal to about 0.45, therefore a steam integration is required. Due to the primary importance of this parameter, both on the shift reactors performance and on the energy balance of the overall system, a parametric analysis of the steam/CO molar ratio n, in the syngas entering the HT-WGS section, was carried out, varying its value in the range 1.5-2.5.

Figure 2 shows steam mass flow and thermal power required for water preheating, vaporization and steam superheating as a function of the steam/CO molar ratio n. Figure 3 shows CO conversion and syngas temperature downstream of the HT-WGS reactor as a function of n.

Figure 2 shows that, in order to obtain a molar ratio n=1.5, a steam mass flow equal to 14 kg/h must be injected in the HT-WGS reactor. Assuming water preheating from ambient temperature and steam superheating up to 350 °C, a thermal power of 12 kW is required. A molar ratio n=2.5 requires a higher steam production (27.2 kg/h) and a corresponding higher thermal power of 23.2 kW.
kW. Thermal power requirements are reported in Figure 2 without considering boiler losses; anyhow, actual thermal power consumption was calculated assuming a boiler efficiency of 0.90.

A CO conversion between about 70-82% is obtained in the HT-WGS reactor varying the value of \( n \) in the range 1.5-2.5. No data were available on thermal losses, so the reactor was considered adiabatic and the outlet temperature calculated by the model is greater than the expected values. The syngas temperature is around 485 °C, with a maximum for \( n=1.9 \), despite the increase of CO conversion with \( n \) enhances the WGS exothermic reaction. In fact, thermal energy from shift reaction is partially used to superheat unreacted steam up to the reaction temperature.

Downstream of the HT-WGS reactor the syngas is split in two streams. A first one feeds the bubbling reactor for \( \text{CO}_2 \) removal, where is cooled up to about 35 °C, and a second one is directly sent, at high temperature, to the LT-WGS reactor. The two streams are mixed before entering the LT-WGS reactor and the final temperature is a function of their relative proportions. Imposing a syngas temperature of 200 °C at the entry of the LT-WGS reactor, the syngas fraction flowing to the decarbonisation section ranges between 70-75%, depending on the steam/CO molar ratio \( n \); by increasing \( n \) the syngas fraction flowing to the bubbling reactor increases.

As described before, the \( \text{CO}_2 \) capture occurs in two bubbling reactors, where the \( \text{CO}_2 \) is absorbed by a solvent based on a mixture of water and MEA (30% by weight). In this analysis, a \( \text{CO}_2 \) capture efficiency equal to 90% was assumed for each reactor, requiring a solvent mass flow rate 3.5 times greater than the syngas mass flow. The solvent regeneration takes place in the electrical reboiler and requires a thermal energy equal to 3.5 MJ for kg of captured \( \text{CO}_2 \).

Figure 4 shows the thermal power obtained by syngas cooling (total and latent) before entering the first bubbling reactor and the thermal power required for solvent regeneration as a function of \( n \). Figure 5 shows the steam/CO molar ratio \( n' \) of the syngas entering the LT-WGS reactor as a function of \( n \).

![Graph](https://via.placeholder.com/150)

**Fig.4 Thermal and electrical power as a function of steam/CO molar ratio \( n \)**

**Fig.5 Steam/CO molar ratio \( n' \) as a function of steam/CO molar ratio \( n \)**

Figure 4 shows that the thermal power obtained by cooling the syngas from about 485 °C to 35 °C is in the range 16-25 kW, growing with \( n \), but it is partially due to latent heat associated to steam condensation (4-9 kW). Also thermal power required by the reboiler for solvent regeneration increases with \( n \), because the higher CO conversion in the HT-WGS reactor gives a higher \( \text{CO}_2 \) flow rate in the capture section. The solvent regeneration process requires a thermal power ranging between 19-23 kW.

The current purification line integrated with the Sotacarbo pilot plant includes a further steam supply before the LT-WGS reactor, significantly lower than the previous one. Nevertheless
numerical results reported in Figure 5 show that, for each value of n considered, the steam/CO molar ratio $n^*$ of the syngas entering the LT-WGS reactor is high enough to ensure an almost complete CO conversion. As a consequence, numerical simulation does not consider any additional steam supply. Figure 5 shows that a value of n equal to 1.5 leads to a steam/CO molar ratio $n^*$ equal to 1.35, while for n=2.5 the ratio $n^*$ increases up to about 3.5.

Figure 6 shows the CO conversion and the syngas temperature downstream of the LT-WGS reactor as a function of the steam/CO molar ratio n in the case of adiabatic process. Figure 7 shows the thermal power obtained by syngas cooling (total and latent) before entering the second CO$_2$ capture reactor and the thermal power required for solvent regeneration as a function of steam/CO molar ratio n.

Figure 6 shows that the CO conversion rate of the overall WGS section is considerably higher than 90% for each value of n considered in the range 1.5-2.5. In particular, for n=1.5 the CO conversion rate results 92.7%, rising to 98.2% for n=2.5. The syngas temperature at the LT reactor outlet decreases with n (it is about 260 °C for n=1.5 and about 240 °C for n=2.5), due to either the greater dilution with steam and the lower thermal energy developed by the shift reaction. In fact, for lower values of n, despite a lower overall CO conversion rate, the amount of CO involved in the shift reaction in the LT-WGS is significantly higher, due to the lower CO conversion in the HT-WGS reactor.

Downstream of the LT-WGS reactor the syngas is cooled again to 35 °C before entering the second bubbling reactor. Due to a greater amount of CO$_2$ removed in the first reactor increasing n, the syngas mass flow slightly decreases with n, varying in the range 65-68 kg/h. Also the CO$_2$ molar fraction slightly decreases with n (from 0.123 to 0.112 for n=1.5 to n=2.5).

Figure 7 shows that thermal power produced by syngas cooling before the second bubbling reactor is considerably lower than that obtained by the first reactor (Figure 4), mainly due to the lower syngas temperature (Figures 3 and 6). Thermal power obtained by syngas cooling increases with n, despite the syngas is characterized by a lower temperature (Figure 6) and mass flow. As a matter of fact, a substantial part of the thermal power is obtained by the condensation of steam, whose amount in the syngas increases with n. Latent heat of condensation is equal to 0 for n=1.5, due to the low value of steam partial pressure. Conversely, the thermal power required by the reboiler for solvent regeneration decreases with n. In fact, for minor values of n a lower amount of CO$_2$ was captured, due to either the lower syngas flow rate and the CO$_2$ concentration.
At the end of the purification line the syngas enters the PSA section for hydrogen production. A compressor is installed downstream of the CO₂ capture section to increase the syngas pressure up to about 5 bar. The compressor power requirement is just slightly influenced by purification line operating parameters, ranging between 4.2-4.3 kW. Two separate streams leave the PSA section: a hydrogen-rich gas and the outlet syngas, mainly composed of nitrogen. The hydrogen-rich gas is characterized by a mass flow rate (about 4 kg/h) one order of magnitude lower than the outlet syngas flow rate (47-48 kg/h).

The hydrogen-rich gas is almost completely composed by hydrogen (95-96%), with small amounts of nitrogen (3.5%), CO₂ (0.35%) and CO (from 0.2 to 0.9%). Increasing the ratio n increases the hydrogen purity. For n=1.5 the hydrogen concentration is 95.1%, while for n=2.5 the concentration increases up to 95.9%. The hydrogen-rich gas does not contain methane, oxygen and steam. The outlet syngas is mainly composed by nitrogen (about 78-79%). Hydrogen concentration is around 4.5%, while methane and oxygen show a concentration of about 3% and CO₂ is around 2%. These concentrations are not appreciably influenced by n; on the contrary, CO molar fraction decreases with n. For n=1.5 CO concentration is 2.4%, while for n=2.5 is reduced to 0.6%. Steam molar fraction is around 7-8%; for n greater than 1.6, steam concentration remains almost unchanged as n increases. In fact, the greater amount of steam introduced for WGS process condensates during the cooling process.

2.3. CO₂ emissions

The CO₂ removal for reducing CO₂ emissions is the most important task of the syngas purification line. To better evaluate CO₂ emissions the mass flow of CO₂ equivalent was calculated adding to the actual CO₂ emissions the amount of CO₂ that would be formed by combustion of residual CH₄ and CO. The mass flow of CO₂ equivalent in the syngas produced by the gasifier and entering the purification line is equal to 45.1 kg/h.

Figure 8 shows the mass flow rate of CO₂ equivalent of both the flows exiting the syngas treatment section: decarbonized syngas and hydrogen rich gas, as a function of the steam/CO molar ratio.

CO₂ equivalent emissions in both gas flows are reduced significantly from n=1.5 to n=2.5, mainly due to the lower contribution of residual CO. CO₂ equivalent emissions are in the range 4-6 kg/h for the decarbonized syngas and much lower than 1 kg/h for the hydrogen-rich gas.

Two different CO₂ removal efficiency parameters have been introduced. A first gross CO₂ removal efficiency, \( \eta_{CO₂,G} \), is defined by eq. 1 as the ratio between the CO₂ captured \( (m_{capt}) \) and the CO₂ equivalent in the syngas produced by the gasifier \( (m_{eq.in}) \). As a matter of fact, the difference between \( m_{eq.in} \) and \( m_{capt} \) is equal to the CO₂ equivalent globally emitted by both the outlet syngas and the hydrogen rich gas \( (m_{eq.out}) \), shown in Figure 8.

\[
\eta_{CO₂,G} = \frac{m_{capt}}{m_{eq.in}} = 1 - \frac{m_{eq.out}}{m_{eq.in}} \tag{1}
\]

As mentioned before, further thermal and electrical energy supplies are required for operating purification line and CO₂ capture section and contribute to increase the actual CO₂ emissions from the overall system. In particular, thermal energy for steam production in WGS section provided by the boiler (Figure 2) and electrical energy required to power the electrical heater (8.5 kW) and the reboiler (Figures 4 and 7) should be taken into account. On the contrary, electrical energy required by fan and solvent recirculation pumps are negligible while large energy consumption of compressor is not considered when calculating CO₂ equivalent emissions. As a matter of fact, the pressure increase provided by the compressor is a requirement of the PSA system and not of the CO₂ removal process.
For this reason, a second net CO₂ removal efficiency, \( \eta_{\text{CO}_2, \text{N}} \) can be defined by eq. 2 considering also the CO₂ produced by the boiler (\( m_{\text{boil}} \)) and CO₂ produced to power the electrical heater and the reboiler, (\( m_{\text{el}} \)):

\[
\eta_{\text{CO}_2, \text{N}} = \frac{m_{\text{capt}}}{m_{\text{eq.in}}+m_{\text{boil}}+m_{\text{el}}} = 1 - \frac{m_{\text{eq.out}}+m_{\text{boil}}+m_{\text{el}}}{m_{\text{eq.in}}+m_{\text{boil}}+m_{\text{el}}}
\]  

The CO₂ mass flow produced by the boiler is easily calculated from the LPG consumption, while to evaluate the CO₂ produced by both electrical heater and reboiler an average value of CO₂ emissions equal to 406.3 g/kWh for the Italian electricity generation system was assumed [15].

Figure 9 shows both the CO₂ removal efficiencies \( \eta_{\text{CO}_2, \text{G}} \) and \( \eta_{\text{CO}_2, \text{N}} \) as a function of the steam/CO molar ratio. The gross CO₂ removal efficiency \( \eta_{\text{CO}_2, \text{G}} \) obviously increases with \( n \), since the CO₂ captured in the two bubbling reactors increases and furthermore the CO₂ equivalent from unreacted CO is greatly reduced. For \( n=1.5 \) \( \eta_{\text{CO}_2, \text{G}} \) is equal to 0.854, while for \( n=2.5 \) it reaches 0.898. Counting the CO₂ produced by the boiler and the CO₂ produced to power the electrical heater and the reboiler, the net efficiency \( \eta_{\text{CO}_2, \text{N}} \) is notably lower than the gross efficiency \( \eta_{\text{CO}_2, \text{G}} \). In particular, an efficiency reduction of 10–12 percentage points, increasing with \( n \), is caused by boiler and electrical heater, whereas largest reductions of \( \eta_{\text{CO}_2, \text{N}} \) up to 20 percentage points, are due to major CO₂ emissions associated to the electrical reboiler. Globally the efficiency \( \eta_{\text{CO}_2, \text{N}} \) is slight lower than 0.6, with a maximum of 0.586 for \( n=1.9 \).

\[\text{Fig. 8} \quad \text{Equivalent CO}_2 \text{ mass flows as a function of steam/CO molar ratio} \ n\]

\[\text{Fig. 9} \quad \text{CO}_2 \text{ capture efficiencies as a function of steam/CO molar ratio} \ n\]

3. Integration of the system

The main aim of the study is to evaluate a possible integration between the syngas purification line and the CO₂ capture system. In the overall system the main thermal energy requirements are devoted to:

• syngas heating upstream of the hot-gas desulfurization process;
• solvent regeneration in the reboiler;
• steam production for the WGS processes.
As previously specified, currently the thermal energy for syngas heating and for solvent regeneration are provided through two different electric heaters, while a boiler fuelled by LPG allows to produce steam for WGS reactions.

This study evaluates the opportunity of recovering thermal energy from syngas cooling processes (Figures 4 and 7) upstream of both the CO₂ capture reactors, that operate at a temperature slightly higher than the ambient one. Currently, this thermal energy is wasted. It is noteworthy to point out that a substantial amount of the wasted thermal energy derives by steam condensation (Figures 4 and 7), and as a consequence, it can not be fully recovered. To perform this thermal energy recovery, a partial syngas cooling can be achieved in specific heat exchangers, to a temperature which depends on steam molar fraction. As a matter of fact, the syngas must be further cooled to nearly ambient temperature before entering the decarbonisation reactors.

A first simple solution (configuration A) requires two heat exchangers, as specified below. A first heat exchanger S1 allows to heat the cold syngas for the hot-gas desulfurization process through the thermal energy recovered by the syngas cooling upstream of the first bubbling reactor. A second heat exchanger S2 allows to recover thermal energy from the syngas cooling before the second bubbling reactor through the preheating of water for WGS process. Not enough thermal energy can be recovered inside the heat exchanger S2 to perform a complete preheating, vaporization and superheating of the steam.

A second possible solution (configuration B) still requires the installation of two heat exchangers, namely S3 and S4. The heat exchanger S3 allows the preheating and the partial vaporization of the water for WGS process through the thermal energy recovered by the syngas cooling upstream of the first bubbling reactor. Therefore, in configuration B the heat exchanger S4 preheats the cold syngas entering the desulfurizer through the syngas cooling upstream of the second bubbling reactor. Due to the low amount of thermal power recovered in S4, the configuration B still requires the electric heater, but reduces its power consumption.

Both the solutions do not consider the integration with the electrical reboiler, due to the very large amount of thermal energy required for solvent regeneration (Figures 4 and 7), considerably higher than the thermal energy recoverable through the syngas cooling.

Figure 10 shows a simplified scheme of the configurations proposed.

Fig. 10. Block diagrams of system integration: a) Configuration A; b) Configuration B.
With reference to the heat exchanger S1, Figure 11 shows the thermal power recovered through the syngas cooling (equal to the electrical heater consumption) and the hot syngas outlet temperature as a function of the steam/CO molar ratio n.

Referring to the exchanger S2, Figure 12 shows the thermal power recovered through syngas cooling (equal to the thermal power for water preheating) and the thermal power to be still provided by the boiler for vaporization and superheating as a function of the steam/CO molar ratio n.

As shown by Figure 11, with the configuration A the thermal energy recovered from the syngas cooling process in S1 allows to fully satisfy the power required for cold syngas heating (8.5 kW), for any value of n. Therefore the configuration A allows to eliminate the electric heater. A large amount of thermal power is still wasted, as it is clear from the hot syngas temperature reported in the figure. Figure 12 shows that the thermal power recovered from syngas cooling in S2 ranges between 1.5-3.5 kW, increasing with n. Moreover, it is considerably lower than the thermal power required for the WGS, reported in Figure 2, and therefore heat recovery can not assure the steam production necessary for WGS process. As a consequence, the heat recovered in S2 can allow to preheat the water up to saturation conditions, just reducing the thermal energy to be provided by the boiler.

A more effective heat recovery can be obtained through the configuration B.

Despite the thermal energy obtained by the syngas cooling before the first bubbling reactor (Figure 7) is greater than the thermal energy required for the steam production (Figure 2), as already mentioned, a substantial amount of heat is obtained by the condensation of the steam contained in the syngas. As a consequence, the cooling thermal energy can not be fully recovered. Therefore, inside the heat exchanger S3, assuming a minimum ΔT at the pinch point equal to 10 °C, it is possible to fulfil just the water preheating and a partial water vaporization. On the contrary, condensation of water vapour does not take place in heat exchanger S4. In fact, imposing a minimum ΔT of 30 °C between hot (Figure 6) and cold syngas, the hot syngas exiting temperature is equal to about 90 °C, higher than steam condensing temperature.

With reference to the heat exchanger S3, Figure 13 reports the fraction of vaporized water and the fraction of steam superheating achieved inside S3 as a function of the steam/CO molar ratio n. Figure 14 shows the cold syngas temperature at the outlet of the heat exchanger S4 and the thermal power to be supplied by electric heater as a function of the steam/CO molar ratio n.
Figure 13 shows that for n<1.8, a complete water vaporization is possible together with a partial superheating of the steam (up to 70% for n=1.5), while the increase of n reduces the fraction of water vaporized to a value of 0.77 for n=2.5. Figure 14 shows that a large portion of syngas preheating takes place in the heat exchanger S4, with a cold syngas temperature at the exit between 210 and 230 °C. Overall, syngas heating requires 8.4 kW, and just 3.5-4.1 kW must be provided by the electric heater. Higher recovery is obtained at low values of the molar ratio n, since an increase of n causes a reduction of mass flow rate and temperature (Figure 6) of the hot syngas entering the heat exchanger S4. Figure 15 shows the CO₂ removal efficiencies η_COG and η_CO₂,N as a function of the steam/CO molar ratio n for the configuration A. Finally, Figure 16 shows the CO₂ removal efficiencies η_COG and η_CO₂,N as a function of the steam/CO molar ratio n for the configuration B.

As expected, for the configuration A the efficiency η_COG reported in Figure 15 shows the same values reported in Figure 9, because η_COG is not influenced by the thermal recovery. Conversely, the efficiency η_CO₂,N increases of about 3-4 percentage points with respect to values reported in
Figure 9, due to both the absence of the electrical heater and the lower LPG consumption in the boiler. The maximum value of $\eta_{CO2,N}$ is equal to 0.624 and it is still obtained for $n=1.9$. For high values of $n$ the increase of efficiency $\eta_{CO2,N}$ with respect to the current situation is slightly more marked, thanks to the greater heat recovery with $n$. In addition to a substantial reduction of the CO$_2$ emissions, this solution allows to eliminate the electric heater, which in the current configuration provides a power of 8.4 kW, and to reduce the LPG consumption in the boiler.

Obviously, also for configuration B efficiency $\eta_{CO2,A}$ shows the same values reported in Figure 9. On the other hand, Figure 16 shows that this configuration ensures values of the efficiency $\eta_{CO2,N}$ 1-1.5 percentage points higher than those of configuration A, leading to an efficiency increase of about 5 percentage points compared with the case without recovery. The efficiency $\eta_{CO2,N}$ increases up to a maximum value of 0.637 for $n=2.0$. On the other hand, configuration B entails complications in the design of the heat exchangers greater than the configuration A, due to the partial water vaporization required in the heat exchanger S3.

4. Conclusions

In this paper several simulation models developed at the University of Cagliari for predicting performance of gasification and syngas treatment processes were revised and adapted to evaluate the performance of the syngas purification and CO$_2$ capture system installed downstream of the Sotacarbo pilot gasification platform. The Sotacarbo syngas treatment line includes a hot-gas desulfurization process, a WGS section, a CO$_2$ removal system and a hydrogen separation section (PSA).

Due to the fundamental role of the steam/CO molar ratio in the syngas entering the WGS, on both shift reactors performance and overall system energy balance, a parametric analysis of this key parameter was carried out. Results show that an increase of $n$ allows to obtain a purer hydrogen-rich gas and to perform a better CO$_2$ removal, as shown by the gross CO$_2$ removal efficiency, $\eta_{CO2,G}$. Nevertheless, several thermal and electrical energy supplies are required for operating purification line and CO$_2$ capture section and contribute to increase the actual CO$_2$ emissions from the overall system. Counting the CO$_2$ produced by the boiler for the WGS steam production and to power the electrical heater and the reboiler, a net CO$_2$ removal efficiency, $\eta_{CO2,N}$ was calculated, resulting notably lower than the gross efficiency $\eta_{CO2,G}$.

The main aim of the study was to evaluate a possible integration between the syngas purification line and the CO$_2$ capture system. The overall system requires thermal energy for syngas heating, solvent regeneration and steam production for the WGS processes. Currently the thermal energy is provided by electric heaters and a boiler fuelled by LPG. The opportunity of recovering thermal energy from syngas cooling upstream of both the CO$_2$ capture reactors was evaluated in this study. To perform thermal energy recovery, a partial syngas cooling can be carried out in specific heat exchangers. Two different configurations were studied coupling differently the syngas cooling processes with syngas heating and WGS steam production. Due to the very large amount of thermal energy required for solvent regeneration, the integration with the electrical reboiler was not considered. Both the configurations allow to increase $\eta_{CO2,N}$: the first one (configuration A) of about 3-4 percentage points, the second one (configuration B) of about 5 percentage points.

The integration between the syngas purification line and the CO$_2$ capture systems is part of an ongoing research. A future application of this thermal energy recovery to the overall system will allow to test the plant and to validate the results discussed above.

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