Pre-combustion CO$_2$ removal in IGCC plant by MDEA scrubbing: modifications to the process flowsheet for energy saving

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Abstract

Amine scrubbing can be successfully applied to pre-combustion CO$_2$ removal from syngas before its combustion for power production. However, the amine-based CO$_2$ capture process is highly energy intensive, due to the heat duty at the reboiler of the regeneration column of the plant.

This paper focuses on configurations of the acid gas removal section that could promote energy saving in an air-blown integrated gasification combined cycle plant. Some modifications of the base scheme, composed of the absorption and distillation columns, have been considered. A sensitivity assessment has been carried out in order to choose the optimal operating parameters, with regard to the energy requirements. Among the configurations presented in this paper, the scheme that meets the constraints on the CO$_2$ recovery and on the composition of the acid gas rich stream, with an improvement of the energy performance of the overall power plant, has been determined.

1. Introduction

   Global climate change leads to high interest in technologies for carbon capture and storage, generally considered the most suitable solution to significantly reduce CO$_2$ emissions from power plants to the atmosphere. This can be

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achieved with chemical absorption by means of aqueous amines, which is one of the most mature capture technology used for the purification of gaseous mixtures such as natural gas and flue gases [1,2]. Amine scrubbing can be successfully applied also to CO_2 removal from syngas, prior to its combustion for power production.

The CO_2 capture process is highly energy intensive, due to the heat duty at the reboiler of the regeneration column where energy, supplied by steam, is required to remove the absorbed CO_2 and to regenerate the solvent to be recycled. Thus, CO_2 capture in power plants is a step causing a reduction of the overall efficiency [3,4].

The current paper focuses on configurations of the acid gas removal section that could promote energy saving in an air-blown IGCC plant, which has been studied in detail in a previous work [5]. In particular, some modifications of the base scheme of the CO_2 removal section, with absorption and distillation columns, have been studied by modeling in ASPEN Plus® environment. A sensitivity assessment has been carried out in order to choose the optimal process parameters, mainly with regard to the energy requirement. Ultimately, a scheme which could improve the energy performance of the overall IGCC plant has been determined.

Nomenclature

AGR	Acid Gas Removal
EBTF	European Best Practice Guidelines for CO_2 Capture Technologies
ER	Emission Rate, kg_{CO2}/MWh
IGCC	Integrated Gasification Combined Cycle
MDEA	MethylDiEthanolAmine
SPECCA	Specific Primary Energy Consumption for CO_2 Avoided, MJ/kg_{CO2}
TIT	Turbine Inlet Temperature, °C
VLE	Vapor-Liquid Equilibrium
η	Efficiency

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2. The power plant with CO_2 capture

2.1. The IGCC plant

The power plant including CO_2 capture is an advanced IGCC, based on air-blown technology [6]. The syngas resulting from low-sulfur bituminous coal gasification, after cleaning and conditioning, feeds a state-of-the-art combustion turbine (TIT equal to 1305°C) as H_2-rich stream. The exhaust heat from the combustion turbine is ultimately recovered in a two pressure level steam cycle with reheat.

A thorough description of the IGCC, along with all the calculation assumptions, is here omitted but it is duly reported in a recent work [5]. As a matter of fact, the modifications detailed in the current paper refer just to the acid gas removal plant, as better reported in the following.

2.2. The acid gas removal plant

The gaseous stream obtained from coal gasification, mainly composed of H_2 and CO, is converted into CO_2 by water-gas shift. It is rich in CO_2, and H_2S may be present depending on the primary feedstock used. The gaseous stream sent to the AGR plant considered in this paper is composed of H_2 (27.837%), CO_2 (25.225%), H_2S (0.070%), CO (0.775%), CH_4 (0.416%), H_2O (0.192%), Ar (0.533%) and N_2 (44.954%), with a mass flowrate of 232.86 kg/s, and is available at 29.32 bar and 35°C.

The AGR station, aiming at removing both H_2S and CO_2, has been designed in order to achieve all the desired specifications according to the EBTF guidelines [7]. In detail, the CO_2 rich gaseous stream obtained from the CO_2 capture plant should contain at maximum 200 ppm(v) of H_2S, with at least 90 vol.% of CO_2. In order to achieve this specification, two separated sections for the removal of H_2S and CO_2, respectively, must be considered, each one operating at different conditions [8]. The gas is fed to a first section where most of H_2S is removed, and then to a

second one, where most of the CO_2 is absorbed, to leave only less than 5% of the CO_2 content present at the entrance of the AGR plant (*i.e.*, a CO_2 removal of 95%). In order to guarantee a selective H_2S removal in the first section of the plant (needed to accomplish the specification on maximum content of H_2S in the final compressed CO_2), an aqueous solution containing MDEA has been used, being MDEA characterized by high selectivity towards H_2S in presence of CO_2. The same solvent, with a different composition, has been employed also for CO_2 removal, because of some advantages, *i.e.* lower vapor pressure and lower heat of absorption in comparison with other amine solutions.

The H_2S removal section is composed of two parallel identical absorption columns, each one treating half of the large syngas flowrate fed to the plant. The rich amine solution is recovered from the two absorbers and is sent to the regeneration section, consisting of one flash vessel and one distillation column. Though MDEA is able to selectively absorb H_2S, a given amount of CO_2 is co-absorbed. This may be undesirable because the H_2S stream must be fed to the downstream Claus plant for sulfur production, which requires a minimum amount of H_2S equal to 20% in the feed stream to avoid more expensive configuration based on oxygen-blown thermal stage [9]. Thus, in the regeneration section, a flash vessel is added in order to remove most of the CO_2 co-absorbed with the H_2S by lowering of the pressure. This unit also removes the H_2 co-absorbed with acid gases, though present in few amount. The gaseous stream leaving the flash vessel is recovered in order to be mixed again with the H_2S-free syngas before entering the CO_2 absorption column (Fig. 1). The semi-lean amine solvent exiting the bottom of the flash vessel is pre-heated in a process-process heat exchanger by the hot lean amine solution and is fed to the distillation column for regeneration with the desired purity. A more detailed description of the plant can be found elsewhere [5].

Two parallel and identical trains are considered in the CO_2 removal plant (only one of them is shown in Fig. 1), in order to remove the amount of CO_2 in columns with characteristics suitable for handling the operation [10]. Each train consists of one absorption column and one regeneration column. The absorber is fed at the bottom with the gaseous stream coming from the H_2S absorber (2) mixed with the vapor stream recovered from the flash vessels of the H_2S removal plant (1), and at the top with the lean MDEA aqueous solution (15) which flows counter-currently along the column. The amine solvent from the absorption column is rich in acid gases (6), mainly CO_2, and is sent to the regeneration section, composed of a distillation column at atmospheric pressure with a partial reboiler and a partial condenser (condenser temperature = 303 K), and recovered at the desired specification, *i.e.* with a mole fraction of CO_2 in the solvent equal to $2.7 \ 10^{-3}$, corresponding to a lean loading (mol_{CO2}/mol_{MDEA}) of 2%. The column is coupled to a recuperative heat exchanger (calculated assuming a $\Delta T_{approach} = 10$ K), which allows to recover part of the heat required at the reboiler.

Because of the large amount of CO_2 compared to H_2S, the operating requirements (circulating solvent, heat supplied to the reboiler, etc.) in the H_2S removal section are much lower than the ones of the CO_2 removal section [5,11]. Therefore, attention has been paid to the plant separating CO_2 and possible modifications with respect to the base scheme in Fig. 1 have been preliminarily evaluated.

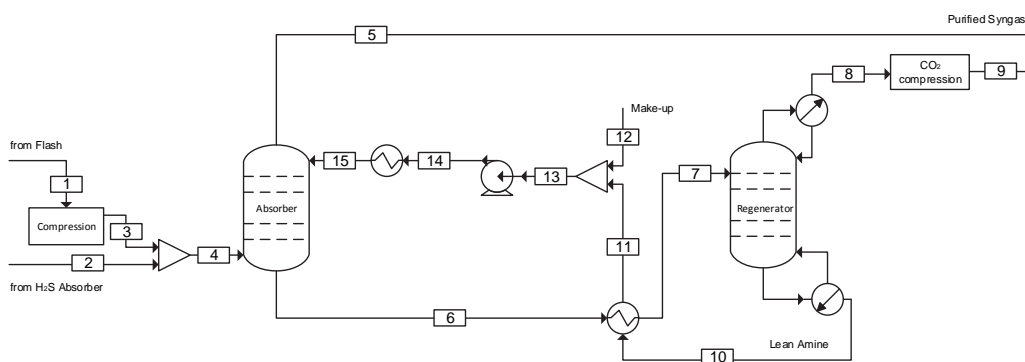Fig. 1. Process scheme of the baseline configuration for pre-combustion CO_2 removal from syngas.

3. The modeling environments

The mass and energy balances of the IGCC have been calculated by means of the in-house code GS [12], already used in past works oriented to simulate a variety of IGCC plant configurations [13-17].

The simulations of the AGR section have been performed by using the commercial software ASPEN Plus®, previously modified as for thermodynamics and mass transfer, with particular focus on the absorption of H_2S and CO_2 in the MDEA solution. The thermodynamic behaviour of the vapor phase is represented by means of a cubic equation of state, while the non-ideality of the liquid phase is reproduced by using an activity coefficient model (Electrolyte-NRTL) [18-23], where the excess Gibbs free energy is taken into account [24]. The solubility of gaseous compounds other than H_2S and CO_2 is simulated by considering the Henry's constant as default in ASPEN Plus®. Mass transfer limitations in the absorption section [25,26] have been described according to the Eddy Diffusivity theory combined with the Interfacial Pseudo First Order model, by linking an external subroutine [27]. Finally, the Peng-Robinson equation of state was used for simulating the CO_2 compression station in ASPEN Plus®.

4. Modifications of the base scheme

The high energy consumption is generally considered one of the major limitations in the application of amine scrubbing processes. Lowering the energy demand of the baseline process can help in reducing losses of power generation efficiency due to acid gas removal. Some modifications of the base case scheme of the AGR section may improve the absorber/regenerator performance, allowing for reductions in the energy consumption of the plant.

Several modifications are reported in the literature [28-30], which have been mainly studied for possible application to post-combustion CO_2 capture from flue gases of power plants, operated at near atmospheric pressure, often by employing an aqueous solution of monoethanolamine. In order to understand whether the same benefits proven in the cases of post-combustion CO_2 capture can be obtained also for pre-combustion mode from syngas, a preliminary evaluation of alternative schemes for the CO_2 removal section has been performed. In particular, the following modifications have been taken into account:

- *Scheme 1*: addition of one flash vessel in the regeneration section;
- *Scheme 2*: addition of two flash vessels in the regeneration section;
- *Scheme 3*: use of semi-lean solvent in the absorption section and addition of two flash vessels with use of CO_2-rich stream in the regeneration section.

The analysis of these alternative configurations has been carried out by considering the same column dimensions of the base case and the same amount of CO_2 to be absorbed (95% of the CO_2 entering the AGR plant). The operating conditions of the added equipment and the amount of solvent flowrates may have been modified in order to achieve the desired specifications according to the different schemes.

4.1. Scheme 1

The first modification is the addition of a vapor-liquid separation unit as reported in Fig. 2. By flashing the rich solvent, some CO_2 is separated from the amine solution with no energy supply. A different composition of the solvent fed to the distillation column (10), characterized by a lower mole fraction of acid gases, is obtained. The acid gas stream (11) exiting the partial condenser can be characterized by a H_2S content higher than the desired one, because part of the CO_2 diluting the H_2S is present in stream (9). However, the required purity specification is satisfied in the overall CO_2 stream sent to compression, obtained by mixing stream (11) with the vapor stream (9) released at the flash vessel.
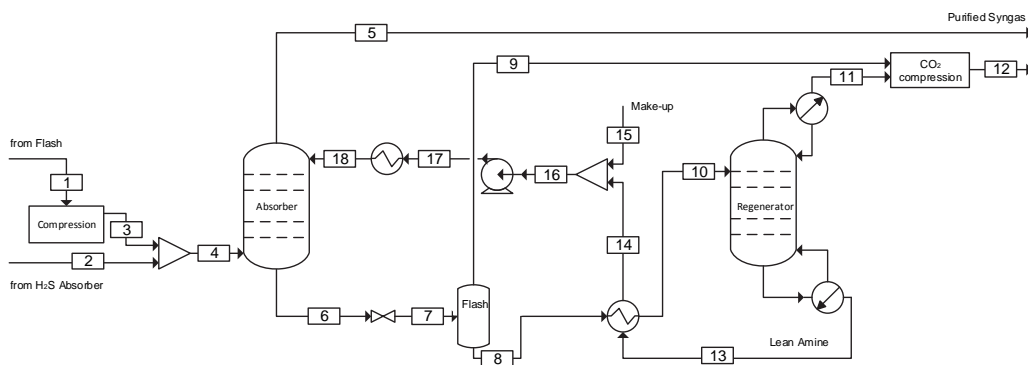
Fig. 2. Modification of the baseline configuration for pre-combustion CO_2 removal from syngas: *Scheme 1*.

4.2. Scheme 2

The configuration of *Scheme 2* is detailed in Fig. 3. It is similar to *Scheme 1*, but there are two flash vessels, operated at different pressures. Some CO_2 released at the first flash vessel (*High Pressure Flash*), operated at higher pressure, and present in stream (9) can be compressed from a higher pressure, therefore saving part of compression power. The second flash vessel (*Low Pressure Flash*), operated at lower pressure, can help in pre-removing additional CO_2 from the liquid solution (10) as in *Scheme 1*, therefore enhancing energy saving in terms of reboiler duty of the distillation column, operated to regenerate a low loaded solution (12). The pressures of the two flash vessels have been chosen based on a sensitivity analysis considering both the regeneration section and the CO_2 compression unit.
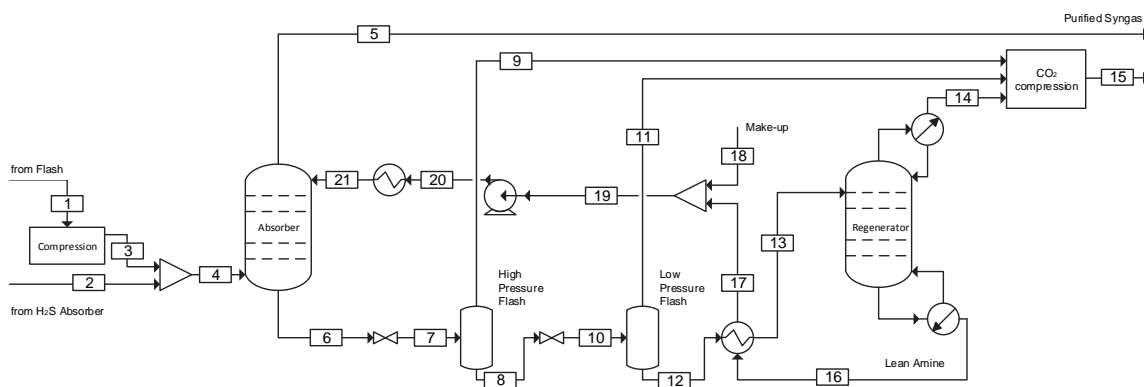

Fig. 3. Modification of the baseline configuration for pre-combustion CO_2 removal from syngas: *Scheme 2*.

4.3. Scheme 3

The configuration with two flash vessels can be further modified (Fig. 4) by recycling part of the partially regenerated solvent (14) to the absorption column without additional regeneration, resulting in a lower amount of amine flowrate to be treated in the regenerator. In particular, a stripping column has been employed instead of a distillation column, without a condenser, so a stream rich in CO_2 at temperature higher than 303 K can be obtained. This stream contains lots of moisture, which needs to be removed before compression. However it can help in further reducing the reboiler duty by increasing the temperature of the second flash equipment, with the

enhancement of the release of volatile species (CO_2) from the liquid phase. The CO_2-rich stream (18) leaving the regeneration column has been therefore split into two streams ((19) and (20)) with the same flowrate. A pump is present to slightly increase the pressure of stream (15) to the operating pressure of the column, which is 0.1 bar higher than the one of the *Low Pressure Flash* unit in order to overcome possible pressure drops and therefore to facilitate stream (19) inlet in the *Low Pressure Flash* without need of compression.
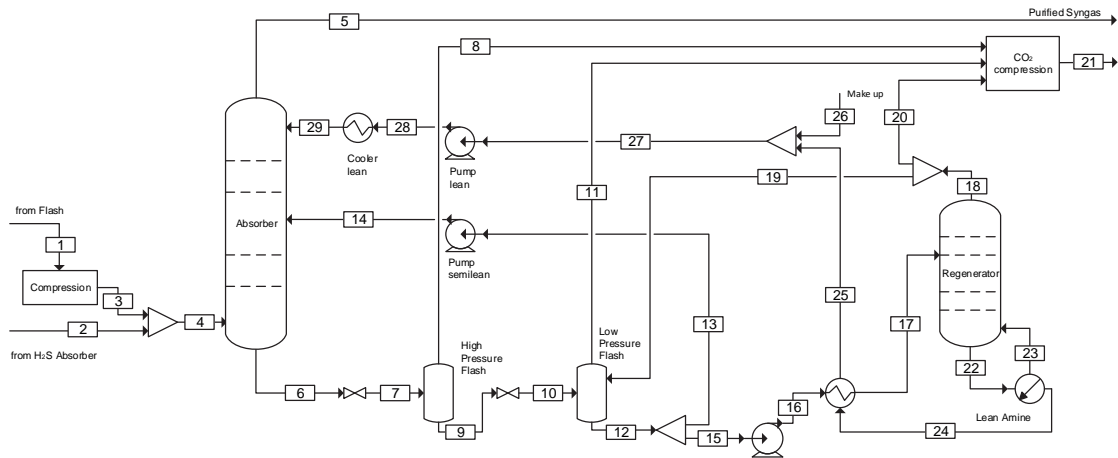

Fig. 4. Modification of the baseline configuration for pre-combustion CO_2 removal from syngas: *Scheme 3*.

The desired level of absorption is achieved by employing both a lean and a semi-lean solution, so the amount of the lean solvent flowrate can be reduced, though the overall amount of circulating solvent is greater than in the other configurations considered in this paper (because a lower absorption capacity is used for the semi-lean solvent).

This scheme reproduces the one proposed by Meissner and Wagner [31], who applied a configuration for activated MDEA scrubbing similar to the one in Fig. 4.

5. Results

The operating pressure of the flash vessels has a strong influence on the amount of vapor coming out of the vessels and, as a consequence, on the amount of CO_2 which can be removed from the liquid rich solvent without heat supply.

Fig. 5 reports results of sensitivity analyses performed on *Scheme 2* for different operating pressures of the *High Pressure Flash* and of the *Low Pressure Flash*. The analyses have been performed for all the studied configurations, with exception of the pressure of the *Low Pressure Flash* of *Scheme 3*, which has been chosen as reported later. For the sake of conciseness, the results shown for *Scheme 2* can be extended also to *Scheme 1* and to *Scheme 3*.

The *High Pressure Flash* operating pressure has been varied in the range from 3 to 7 bar. These limit values have been chosen after considering that the first inter-cooling in the CO_2 compression turbomachinery occurs at around 3 bar and a negligible amount of flowrate can be obtained for pressures higher than 7 bar. The pressure of the *Low Pressure Flash* has been selected between 3 bar, the lower value for the *High Pressure Flash*, and 1 bar, the same operating pressure of the regeneration column of *Scheme 1* and of *Scheme 2* (for *Scheme 3* an operating pressure of 1.1 bar has been assumed, in order to let stream (19) to flow to the *Low Pressure Flash* without need of a compressor, as explained in Section 4.3). As the pressure of the equipment decreases, a higher amount of vapor is recovered. Being the vapor flowrate mainly composed of CO_2, a lower loading in the solvent can be obtained, without energy supply. The operation of the *High Pressure Flash* at 7 bar allows less than half of the CO_2 to leave the aqueous solution if compared to the operation of the same equipment at 3 bar. The obtained CO_2 loading can be close to 0.45 for the lower operating pressure. A similar trend is observed also for the *Low Pressure Flash*, as

reported on the right in Fig. 5. As the CO_2 loading is determined by VLE, for a given pressure of the *Low Pressure Flash*, it results similar both for the case of *High Pressure Flash* operated at 7 bar and for the one of *High Pressure Flash* operated at 3 bar. The two cases differ mainly for the amount of vapor released in the *Low Pressure Flash*, which also depends on the amount of vapor released in the first flash vessel. This amount is lower for the one operated at 7 bar, therefore a higher vapor flowrate is obtained in this case, for a given pressure in the *Low Pressure Flash*.

The lower the pressure, the higher the amount of CO_2 released and the lower the heat duty in the distillation column of the AGR plant. Moreover, the lower the pressure, the higher the pressure ratio necessary in the CO_2 compression section, though a lower amount of CO_2 is recovered for higher pressures of the flash equipment. Pressures in the flash vessels have been determined in order to obtain low reboiler and CO_2 compression duties. For all the considered configurations, the *Low Pressure Flash* has been operated at a pressure near to 1 bar (considering where necessary a pressure of 1.1 bar to facilitate the rich stream inlet in the regenerator without need of an additional pump), in order to meet the lowest reboiler duty. The choice of the *High Pressure Flash* has been taken considering that operating the *High Pressure Flash* at 3 bar allows to recover a more than double vapor flowrate than operating the same unit at 7 bar. Thus, the amount of CO_2-rich stream to be compressed from near 1 bar (exiting the *Low Pressure Flash* and from the regeneration column) would be lower in the case of *High Pressure Flash* operated at 3 bar.
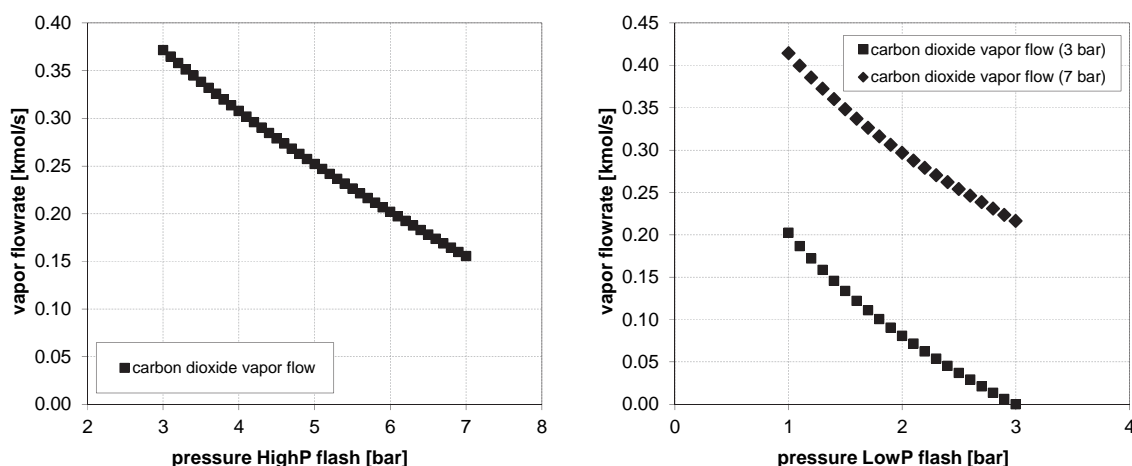

Fig. 5. Variation of CO_2 vapor flowrate of vapor stream from *High Pressure Flash* as a result of the change in pressure of *High Pressure Flash* (on the left) and variation of CO_2 vapor flowrate of vapor stream from *Low Pressure Flash* as a result of the change in pressure of *Low Pressure Flash* for *High Pressure Flash* operated at 3 bar and for *High Pressure Flash* operated at 7 bar (on the right).

The operating pressure of *Low Pressure Flash* of *Scheme 3* has been chosen considering the results shown on the right in Fig. 5 from the same unit in *Scheme 2*, taking into account that in the two units the behavior of the system at vapor-liquid equilibrium should be similar from a theoretical point of view: for any temperature in the range of considered temperatures, a decrease in the operating pressure allows an increase in the amount of CO_2 removed from the rich solution and therefore a decrease in the regeneration in the stripping column. This behavior is not modified by the recycle of stream (19), whose main effect is that the temperature in this flash unit is higher than the one in *Scheme 2*, therefore facilitating the release of a higher CO_2 amount.

Looking at the results in Table 1, operating at 7 bar does not imply significant advantages in terms of savings in the CO_2 compression duty. As a matter of fact, for a given amount of CO_2 available at 7 bar, a large amount of CO_2 needs to be compressed from near 1 bar, with high compression costs.

Table 1. Main results of the CO_2 removal section (referring to just one train), IGCC power balance and main overall performance.

	Base scheme	Scheme 1	Scheme 2	Scheme 2	Scheme 3
CO_2 removal, %			95		
Mass flowrate of lean solvent fed to the absorber, kg/s	404.50	404.50	404.50	404.50	385
MDEA concentration, wt.%			50		
Lean solvent loading, mol/mol			0.02		
Mass flowrate of semi-lean solvent fed to the absorber, kg/s	NA	NA	NA	NA	45.36
Mass flowrate of rich solvent exiting the absorber, kg/s	454.2	454.2	454.2	454.2	480.06
Pressure of the first flash vessel, bar	NA	1.1	3	7	3
Mass flowrate of vapor from the first flash vessel, kg/s	NA	26.32	16.77	6.95	17.48
Mole fraction of CO_2 in the stream from the first flash vessel, mol/mol	NA	0.8763	0.9414	0.9600	0.9407
Pressure of the second flash vessel, bar	NA	NA	1.1	1.1	1
Mass flowrate of vapor from the second flash vessel, kg/s	NA	NA	9.42	19.29	23.03
Mole fraction of CO_2 in the stream from the second flash vessel, mol/mol	NA	NA	0.8761	0.8775	0.8672
$\Delta T_{approach}$ in the cross heat exchanger, K			10		
Reboiler duty, MW	63.41	54.31	53.88	54.09	49.52
Reboiler duty/totally absorbed CO_2, MJ/kg$_{CO2}$	1.28	1.10	1.09	1.09	1.00
Combustion turbine power, MW$_{el}$			227.20		
Combustion turbine auxiliaries, MW$_{el}$			0.80		
Steam turbine, MW$_{el}$	241.63	245.22	245.39	245.31	247.20
Steam cycle pumps, MW$_{el}$	5.69	5.86	5.86	5.86	5.94
Air booster, MW$_{el}$			32.72		
Auxiliaries for H_2S removal, MW$_{el}$			0.65		
Auxiliaries for CO_2 capture, MW$_{el}$	3.31	3.31	3.33	3.32	3.56
Other IGCC auxiliaries, MW$_{el}$			5.11		
CO_2 compression, MW$_{el}$	34.02	34.02	30.72	31.49	30.59
IGCC overall net power, MW$_{el}$	386.53	389.96	393.41	392.57	395.02
Heat input, MW$_{LHV}$			1018.55		
IGCC net efficiency, %	37.95	38.29	38.62	38.54	38.78
Specific emissions, kg$_{CO2}$/MWh	96.73	95.88	95.04	95.25	94.66
SPECCA, MJ/kg$_{CO2}$	3.15	3.01	2.88	2.91	2.82

Compared to the baseline scheme, simulation results of *Scheme 1* show that only 1.10 MJ per kg of absorbed CO_2 is required, corresponding to a saving of around 14% compared to 1.28 MJ/kg$_{CO2}$ [5].

The main effect of the two flash vessels, operated at two different pressures, is the energy saving in the CO_2 compression section, up to 9.7% for *Scheme 2* (with *High Pressure Flash* operated at 3 bar). The calculated CO_2 compression power in *Scheme 2* (with *High Pressure Flash* operated at 7 bar) is slightly higher than the one of *Scheme 2* (with *High Pressure Flash* operated at 3 bar) because of the lower amount of CO_2 exiting the *High Pressure Flash*, though operated at 7 bar. Thus, *Scheme 2* (with *High Pressure Flash* operated at 7 bar) results less interesting than the same configuration with the *High Pressure Flash* operated at lower pressure (3 bar).

As regards the auxiliaries for CO_2 capture, including amine pumps and equipment for heat rejection, the higher demand calculated for *Scheme 3* is mainly due to the additional pump for the semi-lean solution. The increase in the circulation rate, resulting from recycling part of the semi-lean solution to the absorption column in *Scheme 3,* is

mitigated by the lower amount of heat required in the regenerator, equal to 1 MJ/kgCO$_2$, in agreement with the value reported by Meissner and Wagner in their work [31]. Despite the higher capital costs due to the additional components and the higher pump circulation requirement, the reduction in the heat duty at the reboiler of around 22% makes this configuration really interesting from an energy saving point of view.

As reported in Table 1, a reboiler duty lower than the one of the base scheme reflects on less steam from the bottoming power cycle for CO$_2$ stripping, *i.e.* on larger steam turbine power output. This result and the reduced CO$_2$ compression power lead to improvements in IGCC efficiency and reductions in both specific emissions and SPECCA, the latter defined as

$$SPECCA = \frac{3600 \cdot \left(\dfrac{1}{\eta} - \dfrac{1}{\eta_{REF}} \right)}{ER_{REF} - ER} \tag{1}$$

where the subscript REF stands for the reference IGCC without CO$_2$ capture, with a net power output of 415.98 MW$_{el}$, an efficiency of 47.96% and specific emissions of 725.19 kg$_{CO2}$/MWh.

Based on this figure of merit, referring to the specific primary energy consumption for CO$_2$ avoided, a reduction of around 0.30 MJ/kg$_{CO2}$ for the IGCC with CO$_2$ capture based on *Scheme 3* compared to the base case seems to be possible, resulting in a more than 10% lower SPECCA. Thus, CO$_2$ capture process modifications as those necessary for *Scheme 3* can significantly improve the energy performance of the CO$_2$ capture process and of the overall IGCC, though increasing the complexity and the costs of the plant.

6. Conclusions

This paper has been focused on the analysis of some possible alternative schemes of the pre-combustion CO$_2$ removal section in an advanced IGCC plant based on air-blown technology. All these schemes allow for energy saving as they are solutions oriented to limit the reductions of the overall IGCC efficiency.

A sensitivity assessment has been carried out in order to choose the optimal operating parameters, with regard to the energy requirements. The operating pressure of additional flash vessels and other variables specific for each analyzed configuration have been selected, considering the influence exerted by these variables on the energy requirements of the CO$_2$ capture plant.

Focusing on IGCC performance, all the investigated schemes for CO$_2$ capture require a reduced heat duty for CO$_2$ stripping and three of them even result in lower CO$_2$ compression power if compared to the base case. These results clearly reflect on higher IGCC efficiency and lower specific CO$_2$ emissions. On the other hand, if some process modifications can significantly improve the overall IGCC performance, they cause the complexity and the costs of the plant to increase.

Ultimately, a detailed analysis including also capital and operating costs of the possible configurations here investigated would be useful in order to define the most promising scheme for large-scale plants.

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