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Performance improvement of cooled ammonia-based CO₂ capture in combined cycles with gasification of high-sulfur coal

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Abstract

This paper focuses on carbon capture in an advanced combined cycle with air-blown gasification of high-sulfur coal. The technology selected for CO₂ capture is based on cooled ammonia scrubbing in post-combustion mode, as recently investigated by the authors in another work. Here, a stronger integration among the desulfurization unit, the CO₂ capture plant and the steam cycle is considered, in order to improve the ultimate performance. In detail, a specific primary energy consumption for CO₂ avoided (SPECCA) as low as 1.97 MJ/kg_{CO2} is calculated in case of 90% of CO₂ capture, with an ultimate power plant efficiency of 42.53%.

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Keywords: cooled ammonia; CO₂ capture; high-sulfur coal; IGCC; SPECCA

1. Introduction

Coal-based combined cycle is one of the most promising power generation systems to achieve a higher thermal efficiency using state-of-the-art technologies. Currently, the integrated gasification combined cycle technology is mainly based on oxygen-blown gasification, but a significant activity on air-blown gasifiers has been conducting during the last years by Mitsubishi Heavy Industries in Japan, where the 250 MW_{el} demonstration plant in Nakoso was started up in 2007 [1]. Shifting from oxygen- to air-blown gasification technology implies the economic

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advantage related to the much smaller air separation unit and to the potentially higher cycle efficiency [2]. In perspective, IGCC efficiency values as high as 53% may be obtained based on advanced technologies such as 1500°C-class combustion turbine and hot fuel gas clean-up [3]. However, the use of coal increases CO₂ emissions, so carbon capture and storage (CCS) technologies applied to coal gasification plants assume a significant role.

Pre-combustion CCS is the favored choice for IGCC compared to other CO₂ capture options [4], due to the possible removal of highly concentrated CO₂ in the syngas under high pressure. In this case, the size of a CO₂ removal plant can be smaller than in case of post-combustion mode at atmospheric pressure. In pre-combustion mode, coal is converted to syngas in the gasifier and the CO is then converted to CO₂ and H₂ through a water-gas shift process. In post-combustion mode, CO₂ is separated from the exhaust gas without altering the traditional combustion procedure. In this process, the syngas is directly burnt in the gas turbine combustor without first undergoing a water-gas shift reaction and the product stream contains CO₂ in the range of 9–15% [5]. Thus, the post-combustion capture is flexible in operation because the power plant can still operate even if the capture plant is shut down. Post combustion CO₂ capture solutions for IGCC plants have been investigated by the authors [6] and by other researchers [7,8]. However, independently of the specific technology, the energy cost related to CCS is not negligible and results in lower power plant performance.

Nomenclature

| | |
|--------|---|
| CCS | carbon capture and storage |
| conc | ammonia initial concentration in the aqueous solution, wt% |
| COP | coefficient of performance |
| HRSC | heat recovery steam cycle |
| HRSG | heat recovery steam generator |
| IGCC | integrated gasification combined cycle |
| K | NH ₃ -to-CO ₂ ratio in the absorber, mol/mol |
| ric | percentage of rich solution recycled to the absorber, wt% |
| SPECCA | specific primary energy consumption for CO ₂ avoided, MJ/kg _{CO2} |
| TIT | turbine inlet temperature, °C |
| TOT | turbine outlet temperature, °C |

1.1. Post-combustion CCS in IGCC plants

The authors have investigated post-combustion CCS solutions for IGCC plants in past works.

A former study revealed an IGCC efficiency of 38.8 % in case of post-combustion CO₂ capture by amine-based chemical absorption [6], with the specific primary energy consumption for 90% of CO₂ avoided (SPECCA) equal to 3.14 MJ/kg_{CO2}. Significant improvements in IGCC performance are possible if hot coal-derived gas clean-up is realized before the syngas fuels the combustion turbine: an IGCC net efficiency as high as 41.5% was calculated, though such an improvement is related to the gas clean-up technology [3] and not strictly to the CCS technology. Effects of exhaust gas recirculation were investigated as well, without significant performance improvement against a more complicated plant layout.

More recently, attention has been paid to post-combustion CO₂ capture by aqueous ammonia. Some preliminary results [9] have been revised and ammonia scrubbing in chilled and cooled modes has been investigated [10]. Since chilling down to 7°C both the exhaust gas and the ammonia solution results in significant power consumption of chillers [11-13], the cooled mode has been found to be promising as far as a reduction of the energy cost related to CO₂ capture is concerned. As a matter of fact, the energy saving, possible when adopting an air cooling system instead of a chilling plant, is significant with a SPECCA value decreasing from 2.79 to 2.54 MJ/kg_{CO2}, when switching from the chilled to the cooled mode. In order to better control ammonia slip in cooled mode operation, an absorption-desorption cycle has been adopted, just before a final acid wash, where use of the H₂S removed from the coal-derived gas at the desulfurization unit of the IGCC has been made. Even better performance has been

calculated in case of gasification of high-sulfur coal [14]. Once again, a cooled ammonia-based process has resulted to be more attractive than a conventional chilled ammonia-based solution: in case of 90% of CO₂ capture, the overall power plant efficiency is equal to 41.7%, with a SPECCA value as low as 2.3 MJ/kg_{CO₂}.

1.2. Objective of the paper

This paper aims at further investigating the air-blown IGCC with the CCS process based on cooled ammonia scrubbing recently studied by the authors [14]. The feedstock is always a high-sulfur coal, in order to improve the performance of the acid wash limiting the ammonia slip, and attention to a stronger integration among the desulfurization unit of the IGCC, the CCS plant and the steam cycle is made. In detail,

- the heat released with H₂S combustion in the sulfuric acid plant, which was too prudently neglected [14], is here used for medium-pressure steam production delivered to the steam cycle;
- a two-stage (instead of a three-stage [14]) intercooled CO₂ compression is considered and the heat from CO₂ intercooling is recovered for hot water production directed to the steam cycle and to an absorption chiller, replacing the previously considered vapor compression chilling system [14].

2. The power plant with CO₂ capture

2.1. The power plant

The power plant including CO₂ capture is an advanced IGCC, based on air-blown technology [2]. The syngas resulting from the gasification of high-sulfur bituminous coal (wt.%.: 61.27 C, 4.69 H, 8.83 O, 1.1 N, 3.41 S, 12 moisture, 8.7 ash), after cleaning and conditioning, fuels an advanced combustion turbine (compressor pressure ratio and TIT equal to 18.1 and 1360°C, respectively). The exhaust heat from the combustion turbine (with a mass flow rate at the outlet of 665 kg/s) 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 elsewhere [14]. As a matter of fact, the modifications considered in the current study refer just to the CCS plant, as better reported in the following.

2.2. The CCS plant

The CCS plant is schematized in Fig. 1, where the gas stream exiting the HRSG is cooled in a first section of the plant with two contact coolers in cascade (the systems operate with an ambient air-cooled water loop). As the gas flowing through these contact coolers and the next absorber experiences a pressure drop, a fan (BL) is present. The gas temperature rise due to the fan is promptly reduced by the second contact cooler, before the gas enters the CO₂ absorption section.

The CO₂ capture plant consists of an absorber and a regenerator with a recuperative heat exchanger (RCP1) as well as a high-pressure pump (PM2) for the rich solution to the regenerator and a heat exchanger (HX2) on the lean solution to the absorber. All the streams entering the absorber are cooled down to 20°C, selected based on 15°C of ambient temperature. Because of the exothermic reactions, the absorber temperature varies along the reactor from around 20°C (at the top) to 25°C (at the bottom). The operation temperature of the absorber results in increased solubility of the salts present in the system, as also shown by other researchers [15]. This condition, alone, is not enough to avoid the precipitation of salts a priori, so a hydro-cyclone (HC) is present in the layout of Fig. 1.

Ammonia slip in such a cooled mode operation is significant, so an absorption-desorption cycle just before the final acid wash (WT1) is necessary for a level of ammonia slip reducible by the H₂S available from the desulfurization unit of the IGCC, as described hereafter. In particular, part of the NH₃ in the gaseous stream exiting the absorber is firstly reduced in a wash tower (WT3), resulting in a NH₃-rich solution pumped from the wash tower to a stripper, where NH₃ in the gaseous phase is recovered and delivered to the absorber. The ultimate ammonia slip is reduced to traces by acid washing in a dedicated tower (WT1).

The H₂S-rich stream stripped after solvent regeneration at the desulfurization unit of the IGCC is used for producing sulfuric acid, which is ultimately used to drastically reduce ammonia slip in the gas stream to the stack, according to the following reaction:



The heat released with H₂S combustion, which was too prudently neglected in the previous work [14], is here used for producing medium-pressure (36 bar) steam directed to the bottoming cycle of the power plant. In detail, the H₂S-rich gas is incinerated to SO₂ in a burner followed by a waste heat boiler. The gas leaving this boiler is supposed to have a temperature of 400°C [16]. Thus, based on the amount of sulfur in the coal, around 16.5 MW can be recovered.

As regards the path of the CO₂ stream exiting the regenerator, a condenser for moisture separation, operating with a dedicated air cooler, and a water wash tower (WT2) for ammonia slip control, with an air-cooled closed loop, are present. Finally, there is the CO₂ compression station, which consists of two intercooled stages with the same pressure ratio (CM1 and CM2), heat exchangers, air coolers and a condensed water knockout to dehydrate the CO₂ stream. The CO₂ stream enters the compression station at 5 bar and exits the last stage at slightly supercritical pressure. After recycling the amount of CO₂ necessary for coal loading and a further cooling, the supercritical liquid CO₂ stream is ultimately pumped to storage. Differently from the previous work [14], where the heat from CO₂ intercooling was dissipated to the environment, now it is recovered for producing hot water directed to the steam cycle and to an absorption chiller (linked to the heat exchanger on the line from the NH₃ stripper to the wash tower WT3).

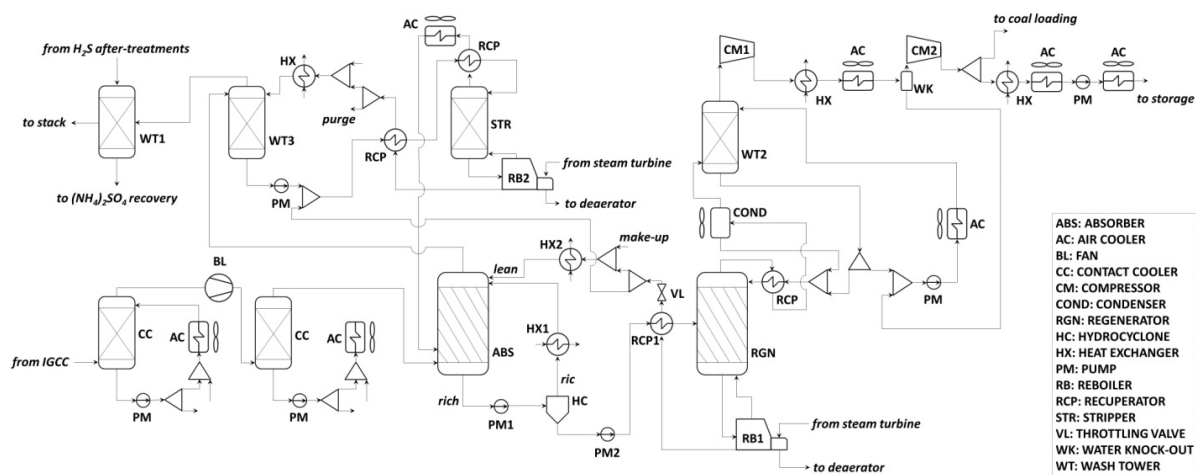


Fig. 1. The CCS plant based on cooled ammonia scrubbing.

3. Calculation environments

The thermodynamic models of the IGCC with CO₂ capture have been implemented in the modular simulation code GS, integrated with the commercial code Aspen Plus[®].

Mass and energy balances for the IGCC have been carried out with the code GS, which is a simulation tool originally designed for research purposes by the authors' research group to calculate gas-steam cycles and progressively improved to calculate more complex systems. It has proved to yield highly accurate results in estimating the performance of combustion turbines and combined cycles [17] and has been successfully used to calculate mass and energy balances of a variety of power plant configurations [18-24].

The commercial code Aspen Plus[®] has been used for the calculations of the CCS plant. In detail, the absorption and regeneration processes have been simulated by means of the Extended UNIQUAC model that is not built inside the code but defined by the user, as successfully done by Darde et al. [25]. An exhaustive validation of the thermodynamic model was previously carried out and the model results compared with experimental data available in literature [14,26]. Besides, the Soave-Redlich-Kwong equation of state has been chosen to simulate both the cooling of the gas entering the CCS plant and the CO₂ compression station.

The simulation of the CO₂ capture plants were run by changing four operation parameters for a specific amount of CO₂ captured [14]:

- the ammonia initial concentration in the aqueous solution:

$$\text{conc} = \frac{\dot{m}_{\text{NH}_3}}{\dot{m}_{\text{NH}_3} + \dot{m}_{\text{H}_2\text{O}}} \quad (2)$$

- the NH₃-to-CO₂ ratio in the absorber (K), defined as the ratio between the number of NH₃ moles in the lean solution line to the absorption reactor and the number of CO₂ moles in the line from the HRSG outlet:

$$K = \frac{\dot{n}_{\text{NH}_3, \text{lean}}}{\dot{n}_{\text{CO}_2, \text{in}}} \quad (3)$$

- the percentage of rich solution recycled (ric) to the absorber,
- the regeneration pressure.

As anticipated, due to NH₃ volatility, it is necessary for the absorption temperature to be as low as possible. Based on the ambient temperature (15°C), 20°C was selected as the minimum operation temperature of the streams entering the absorber. Air-cooler systems allow to reach this temperature. In particular, setting the temperature of the streams entering the absorber directly affects (i) the temperature for the exhaust gas treatments, (ii) the flow rate of the lean solution and (iii) the amount of rich solution recycled to the absorber (ric in Fig. 1).

Based on the experience achieved with former works [12,13], a significant number of cases were investigated as parametric simulation runs [14]. The final operation parameters were selected as their combination allowing for the lowest power consumption. Accordingly, Table 1 reports these values as the results of the parametric analysis: differently from the first case, with 80% of CO₂ capture, the same set of parameter values were found for the cases with 85 and 90% of CO₂ capture.

Table 1. Key-parameters set for CCS plant calculations

| Amount of CO ₂ captured, wt% | 80 | 85 | 90 |
|---|------|------|------|
| ric, wt% | 50 | 10 | 10 |
| conc, wt% | 10 | 5 | 5 |
| K, mol/mol | 5.25 | 4.75 | 4.75 |
| Regeneration pressure, bar | 5 | 5 | 5 |

4. Results

The results of the IGCC calculations are reported in Table 2, with details of all the main components of the CCS plant. Details of the power cycles are purposely limited as the modifications considered in this study focus just on the CCS plant and reflect on the steam turbine output, the HRSC pump demand and the heat rejected at the condenser. The results are reported with reference to one gasification train: C80, C85 and C90 refer to three cases with different level of CO₂ capture, i.e. 80, 85 and 90%. An asterisk is used for the cases previously investigated [14], in order to have a direct comparison of the possible performance improvements.

The results are discussed in the following with reference to a specific component of the IGCC.

Table 2. Power details for IGCC and the CCS plants (for just one gasification train) and overall power plant performance

| | C80* | C80 | C85* | C85 | C90* | C90 |
|---|-------|--------|--------|--------|--------|--------|
| Combustion turbine, MW _{el} | | | | 261.25 | | |
| IGCC auxiliaries, MW _{el} | | | | 30.91 | | |
| Steam turbine, MW _{el} | 223 | 228.36 | 219.4 | 225.09 | 216.57 | 225.05 |
| HRSC pumps, MW _{el} | 5.12 | 5.23 | 4.97 | 5.09 | 4.84 | 5.09 |
| Heat duty at the condenser, MW | 170.1 | 179.3 | 155.15 | 165.2 | 142.9 | 165.8 |
| Exhaust cooling | | | | | | |
| Air coolers, MW _{el} | | | | 0.81 | | |
| Fan, MW _{el} | | | | 5.48 | | |
| Pumps, MW _{el} | | | | 1.14 | | |
| CO ₂ capture plant | | | | | | |
| Heat duty at the regenerator, MW | | 177.3 | | 210.1 | | 221.7 |
| Regeneration temperature, °C | | 104.6 | | 105.3 | | 106.5 |
| Heat duty at NH ₃ stripper, MW | | 29.4 | | 13.6 | | 18.8 |
| Temperature for NH ₃ stripping, °C | | 97 | | 97.8 | | 97.9 |
| Chillers, MW _{el} | 0.92 | - | 0.5 | - | 0.71 | - |
| Air coolers, MW _{el} | | 2.94 | | 3.29 | | 3.53 |
| Pumps, MW _{el} | | 1.45 | | 1.95 | | 1.95 |
| CO ₂ compression | | | | | | |
| IC compressors, MW _{el} | 16.28 | 17.28 | 17.27 | 18.4 | 18.32 | 19.47 |
| Heat recovery from CO ₂ intercooling, MW | - | 17.4 | - | 18.5 | - | 19.6 |
| Air coolers, MW _{el} | 0.53 | 0.28 | 0.57 | 0.3 | 0.6 | 0.32 |
| Pumps, MW _{el} | 0.48 | 0.48 | 0.5 | 0.5 | 0.53 | 0.53 |
| Overall results (for two gasification trains) | | | | | | |
| Net electric power, MW _{el} | 836.4 | 847.2 | 826.3 | 836.9 | 817.7 | 834.1 |
| Net electric LHV efficiency, % | 42.64 | 43.20 | 42.13 | 42.67 | 41.69 | 42.53 |
| Specific emissions, kg _{CO2} /MWh | 170.3 | 168.1 | 127.9 | 126.3 | 86.4 | 84.7 |
| SPECCA, MJ/kg _{CO2} | 2.27 | 2.04 | 2.29 | 2.07 | 2.27 | 1.97 |

- The combustion turbine power output is always the same for the investigated cases. As a matter of fact, the power plant is not affected by modifications to the CCS plant, with the exception of the steam cycle. IGCC auxiliaries do not vary as well.
- Based on the better heat recovery from H₂S combustion and in the modified CCS plant, the steam cycle receives steam at 36 bar, resulting from H₂S combustion, as well as hot water from CO₂ intercooling. Thus, more steam expands in the turbine, resulting in greater power output. In detail, steam turbine power increases by 2.5% for cases C80 and C85 and by 4% for case C90.
- A slight increase in HRSC pump demand is justified by the higher condenser heat duty, reflecting in more auxiliary power demand for heat rejection.
- As regards the exhaust cooling at the exit of the HRSG, before the gas enters the CO₂ absorber, the overall duty is always the same, as this section of the CCS plant does not present layout modifications.
- At the CO₂ capture plant, the heat duty of the regenerator clearly reduces when less CO₂ is removed from the gas, with an almost constant operation temperature at the regenerator, as the regeneration pressure is maintained at 5 bar [14]. When operating the CCS plant in cooled instead of chilled mode, ammonia slip in the gas phase is significant [14], because of the absorber temperature (variable from 20 to 25°C). As schematized in Fig. 1, a NH₃

stripper is present to preliminarily limit the amount of ammonia slip that is ultimately controlled in the acid wash WT1. As a matter of fact, the gas exiting the CO₂ absorber cannot be directly delivered to the acid wash WT1 because the amount of H₂S removed at the desulfurization unit of the IGCC is not sufficient to completely reduce NH₃ to traces. In the current study, without altering the operation the CO₂ capture plant, the possibility of replacing the vapor compression chiller with an absorption machine is considered, in order to save the related power consumption. Table 2 details the electric power necessary for a vapor compression chiller, for the stream from the NH₃ stripper to the wash tower WT3, with a COP equal to 5. Here, the same chilling power comes from an absorption chiller, driven by a stream of hot water at 98°C resulting from partial CO₂ intercooling. In this case, a COP of 0.76 is considered, based on indications of manufacturers [27], with the hot water experiencing a temperature drop of 10°C inside the absorption machine. On the other hand, air cooling and pump demands cannot be limited, so they remain unchanged.

- CO₂ compression power increases compared to the previous cases [14], as the consequence of two instead of three intercooled stages. In detail, CO₂ compression power is greater to the extent of around 6.5%. However, if the CO₂ stream temperature was limited to no more than 110°C at the exit of the three-stage turbomachinery, such a temperature raises to around 155°C in case of a two-stage turbomachinery, reflecting on a sensible heat recovery during CO₂ intercooling. Here, CO₂ intercooling is divided into three stages: (i) down to 98°C, resulting in hot water for the absorption chiller and hot water to the steam cycle, (ii) down to 60°C, with former economization of such a water stream up to 88°C, (iii) down to 25°C, with heat rejection. As detailed in Table 2, the heat recovery, which is similar to the CO₂ compression power, reflects on lower power consumption for heat rejection. CO₂ pump demand obviously remains unchanged.

Focusing on the overall results in Table 2, the net power output increases as the results of the considered modifications. As a consequence, the net electric efficiency increases as well. In spite of slight reductions of the specific CO₂ emissions, it is possible to appreciate sensible reductions of the specific primary energy consumption for CO₂ avoided (SPECCA):

$$\text{SPECCA} = \frac{3600 \cdot \left(\frac{1}{\eta} - \frac{1}{\eta_{\text{REF}}} \right)}{\text{ER}_{\text{REF}} - \text{ER}} \quad (4)$$

where the subscript REF stands for the reference IGCC without CO₂ capture, with a net power output of 950.1 MW_{el}, an efficiency of 48.93% and specific emissions of 647.7 kg_{CO2}/MWh [24]. In detail, the modifications considered in this study reflect on a SPECCA of around 2 MJ/kg_{CO2}, with a reduction up to 0.3 MJ/kg_{CO2} compared to the cases studied in a previous work [14].

5. Conclusions

This paper continues a recent work of the authors focusing on carbon capture in advanced combined cycles with air-blown gasification. Here, a stronger integration among the desulfurization unit, the CO₂ capture plant and the steam cycle is considered, in order to improve the ultimate performance of the power plant.

The cooled ammonia scrubbing in post-combustion mode has proved to be an interesting technology for the investigated power plant in case of gasification of high-sulfur coal. The key to better performance for this CCS technology is strictly related to the high content of sulfur in the feedstock, ultimately resulting in sulfuric acid. This by-product is necessary to reduce to traces the ammonia slip in the gas exiting the CO₂ absorber, which is an issue as relevant as higher the operation temperature in the CO₂ absorber is.

In detail, the investigated integration results in higher IGCC performance, with a specific primary energy consumption for CO₂ avoided of around 2 MJ/kg_{CO2}, with a reduction up to 0.3 MJ/kg_{CO2} compared to the cases studied in a previous work [14].

Ultimately, these results, which require an economic assessment for properly considering the proposed technology, are strictly related to the case investigated here, i.e. when fueling high-sulfur coal in IGCC plants. As a

matter of fact, in case of gasification of a more common low-sulfur bituminous coal [10], IGCC performance with the same CCS technology is not so clear and interesting as the one presented and improved in this paper.

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