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Thermodynamic Assessment of Cooled and Chilled Ammonia-Based CO₂ Capture in Air-Blown IGCC Plants

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Abstract

The energy impact of different post-combustion CO₂ capture plants integrated in an advanced air-blown IGCC is simulated in this paper. Ammonia scrubbing is considered as the CO₂ capture technology and chilled and cooled modes are investigated with reference to operation temperatures at the absorber equal to 7°C and 20°C respectively. Ammonia slip is controlled by means of an absorption-desorption cycle just before a final acid wash, where use of the H₂S removed from the coal-derived gas at the desulphurization unit of the IGCC is made.

Focusing on three levels of CO₂ capture, from 80% to 90%, it is possible to appreciate that the cooled mode is 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 the specific primary energy consumption for 90% of CO₂ avoided which decreases from 2.79 MJ/kgCO₂ to 2.54 MJ/kgCO₂, when switching from the chilled to the cooled mode, with a difference equal to about 0.7 percentage point in IGCC efficiency.

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1. Introduction

The use of coal for power generation assumes an increasing role in the global energy scenario since security of supply and cost effectiveness of fuel and electricity make coal-fired power plants one of the most viable solutions for

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power generation. However, the use of coal increases emissions of pollutants and of CO₂. In this framework, Carbon Capture and Storage (CCS) technologies, applied to coal combustion and gasification plants, can assume a significant role in controlling global warming emissions.

The performance of coal-fired power generation plants equipped with CCS systems has been thoroughly studied by the authors. In particular, ultra-super-critical pulverized coal combustion [1-4] and integrated gasification combined cycle [5-8] technologies currently represent the most promising solutions for a CO₂-free power generation from coal.

Although pre-combustion CO₂ capture is a more investigated solution for IGCC power plants [7,8], this paper focuses on post-combustion CO₂ capture and considers the option of the Chilled Ammonia Process (CAP) proposed by Alstom [9], which seems to be a feasible and mature technology in the short term, along with the flue gas scrubbing with MEA. Nevertheless, the CCS energy cost is not negligible and results in lower power plant performance [10]. The CAP technology is suggested also for CO₂ capture in iron and steel industry [11] and applied with membranes [12].

Here, starting from preliminary investigations [6] focusing on possible reductions of such a cost, a more accurate analysis of a CO₂ capture process based on (i) a chilled and on (ii) a cooled aqueous ammonia scrubbing is presented and discussed. In particular, an air-blown IGCC is considered, based on its higher energy conversion efficiency [13]. A proper integration of the IGCC and the CO₂ capture process, with reference to the possibility of using the H₂S removed from the coal-derived gas at the desulphurization unit of the IGCC in the CO₂ capture plant, allows for a better control of the ammonia slip.

Nomenclature and acronyms

AGR	Acid gas removal	LHV	Lower heating value
CAP	Chilled ammonia process	LP, IP, HP	Low, intermediate, high pressure
CCS	Carbon capture and storage	LT, HT	Low, high temperature
CT	Combustion turbine	MEA	Monoethanolamine
ER	CO ₂ emission rate (kg _{CO2} /kWh)	SPECCA	Specific primary energy consumption for CO ₂ avoided
HR	Heat rate (kJ/MWh)	TIT	Turbine inlet temperature
HRSG	Heat recovery steam generator	η	Efficiency
IGCC	Integrated gasification combined cycle		

2. CO₂–NH₃–H₂O system for CO₂ capture by ammonia scrubbing

Understanding the phase behaviour and the thermodynamics of the CO₂–NH₃–H₂O system is important for engineers and researchers interested in post-combustion ammonia-based CO₂ capture. Such a ternary system forms an electrolyte solution, whose thermodynamic properties must be studied with an appropriate model. In particular, the Extended UNIQUAC thermodynamic model for gas solubility in salt solutions, developed by Thomsen and Rasmussen [14], has been used in this paper. An exhaustive validation of the thermodynamic model was preliminarily carried out and the model results compared with experimental data available in literature. Some representative trends about this validation are reported in [15].

Focusing on the chilled ammonia process, a slurry consisting of a liquid in equilibrium with solid ammonium bicarbonate (NH₄HCO₃) is produced in an absorber. The slurry releases CO₂ at a relatively high pressure, when heated in a desorber, therefore the liquid is cooled and delivered to the absorber for a new cycle. Besides, the CAP layout is equipped with a water wash at the top of both the absorber and the regenerator, because of ammonia slip (NH₃ volatility results in NH₃ vaporization to the flue gas [16]), which is more significant for higher CO₂ absorption temperature. The issue of the NH₃ escape can be solved with other options, as shown in [17] and proven in [18, 19].

In the following, an interesting solution to control ammonia slip is suggested, as an integration of the CCS plant with the desulphurization unit of the IGCC.

3. Calculation tools

The thermodynamic models of the IGCC plants with CO₂ capture have been implemented in the modular simulation code GS, integrated with the commercial code Aspen Plus®. In particular, mass and energy balances for the IGCC plant have been carried out by means of the code GS [20], which is a simulation tool originally designed for research purposes by the authors' research group. It has proved to yield highly accurate results in estimating the performance of combustion turbines and combined cycles [21] and has been successfully used to calculate mass and energy balances of a variety of power plant configurations, including gasification systems [22], desulfurization reactors [23], water-gas shift processes [24] as well as coal drying systems [25]. As regards the simulation of the CCS plant, use of the commercial code Aspen Plus® has been made. In detail, the absorption and regeneration processes, better outlined in the next section, have been simulated by means of the Extended UNIQUAC thermodynamic model, after its implementation in Aspen Plus®, as successfully reported by Darde et al. [3].

4. IGCC and CCS plant layouts

The IGCC lay-out is schematically shown in Fig. 1: each power plant consists of two gasification trains and two combustion turbines with two heat recovery steam generators, which share the same steam turbine. Air-blown technology has been considered for coal gasification due to the higher IGCC efficiency [13]. As regards the descriptions of both the two-stage air-blown gasification system and the IGCC plant, reference to [13] is made for the sake of brevity. However, some differences between the former plant and the current one have to be specified.

- A low-sulfur coal (66.52% C, 3.78% H, 5.45% O, 1.56% N, 0.52% S, 8.01% moisture, 14.16% ash - on a weight basis; 25.17 MJ/kg LHV) is considered as fuel input.
- As a consequence of the pressure drop (5 bar) for the coal-derived gas at the CT fuel valve, the gasification pressure is set at 28.1 bar.
- A heat exchanger is present before air boosting to the gasifier, with HP steam production.
- The H₂S removed (about 750 kg/h for each gasification train) from the raw coal-derived gas at the AGR station is not recovered in a Claus plant [26], but used as input in a wet sulphuric acid process, as better described hereafter.
- As recommended in [10], an advanced combustion turbine (pressure ratio and TIT equal to 18.1 and 1360°C respectively, with 665 kg/s of exhaust gas) is used as topping cycle.

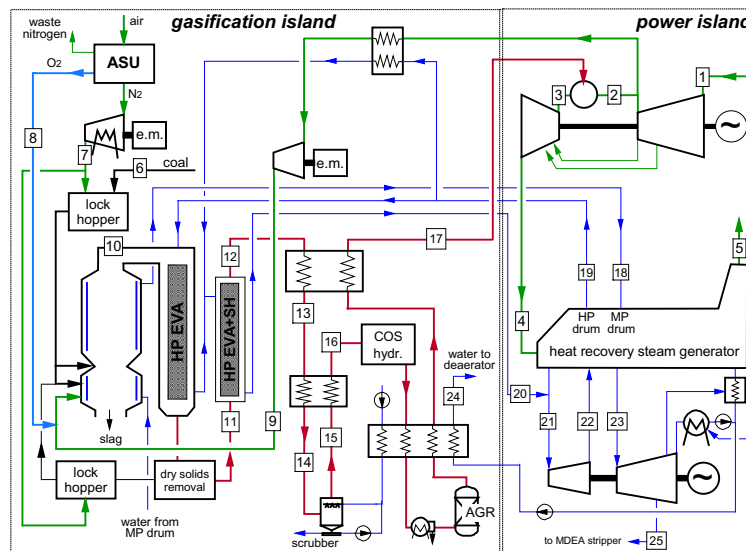


Fig. 1. Schematic of the IGCC plant without CO₂ capture.

- Further specific details concerning the streams numbered in Fig. 1, in terms of pressure, temperature and mass flow rate, are thoroughly reported in [13].

In case of CO₂ capture, the lay-out of the IGCC plant is strictly similar to the one shown in Fig. 1, even though there is no air separation unit. In detail, part of the CO₂ delivered by the CO₂ compressor at the CCS plant is recycled back to the gasification island for coal loading, so both the air separation unit and the N₂ compressor are not necessary. In this case, the air blown to the gasifier is not oxygen-enriched and using CO₂ instead of N₂ for coal loading causes slight variations in the cold gas efficiency and in the fuel gas composition [5]. In addition, when considering CO₂ capture, more steam is extracted from the bottoming cycle, so stream 25 in Fig. 1 has to supply both the heat necessary for H₂S stripping at the AGR unit and the heat required at the CCS plant for the regeneration of the CO₂-rich solution (with CO₂ release) and for NH₃ stripping, as detailed in the following.

Focusing on the chilled mode, Fig. 2 shows that the gas stream exiting the HRSG is chilled in a first section of the plant through three contact coolers in cascade at decreasing temperature levels: the first system operates with an ambient air-cooled water loop and the other two with chilled water loops (HX3 and HX4). The evaporation temperature of the chillers is set as low as possible, with prevention of ice formation (the gas from the HRSG releases moisture when passing through the contact coolers). As the gas flowing through the three contact coolers and the next absorber (ABS) experiences a pressure drop, a fan (BL) is present. The gas temperature rise due to the fan is promptly decreased by the third contact cooler, before the gas enters the CO₂ absorption section, whose layout is conventional and similar to the one proposed in [4].

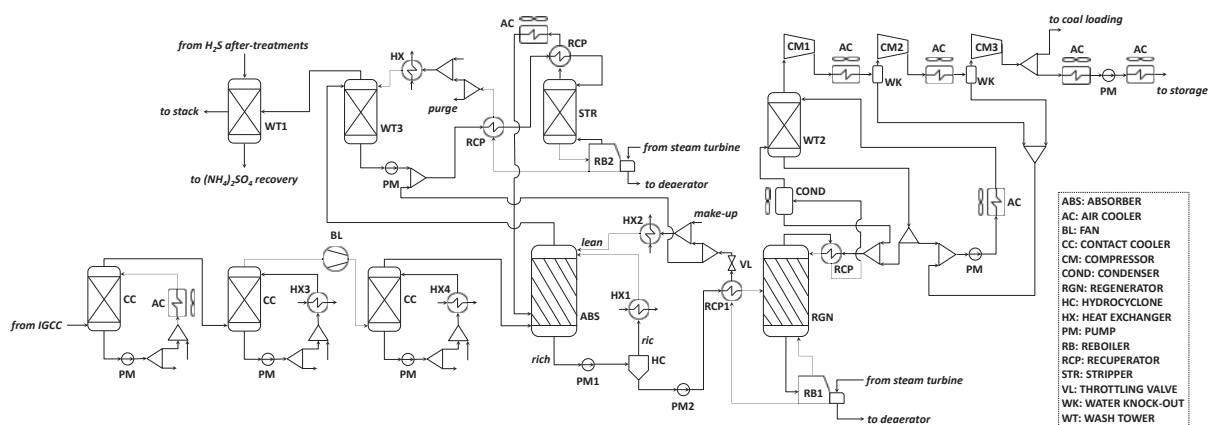


Fig. 2 Schematic of the CCS plant for the chilled case.

The CO₂ capture plant consists of an absorber (ABS) and a regenerator (RGN), along with a recuperative heat exchanger (RCP1) as well as a high-pressure pump (PM2) for the rich solution to the regenerator and a chilling exchanger (HX2) on the lean solution to the absorber. The rich solution from the absorber is pumped to a hydrocyclone (HC), which separates the solids, sent to regeneration with a small portion of liquid, from the bulk of the liquid, chilled in HX1 and recycled back to the absorber in order for the process temperature to be as cold as possible, since the absorption reaction is exothermal. The gas exiting the absorber is firstly treated in an absorption-desorption cycle, just before the final acid washing (WT1) in order to decrease the amount of ammonia slip reducible by the H₂S available from the desulphurization unit of the IGCC. In detail, 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 (STR), where NH₃ in the gaseous phase is recovered and delivered to the absorber (ABS). Later, the ammonia slipping from the absorber wash is reduced to traces by acid washing in a dedicated tower (WT1).

In detail, the H₂S-rich stream stripped after solvent regeneration at the desulphurization unit of the IGCC is supposed to react with air, according to the following reaction (the presence of N₂ and other inerts in air is neglected for the sake of simplicity):



After SO₂-to-SO₃ oxidation in presence of a vanadium oxide catalyst, hydration and condensation, H₂SO₄ is ready to be used in order to reduce ammonia slip to traces according to the following reaction:



The heat released with H₂S oxidation and the other exothermic reactions in the wet sulphuric acid process has been neglected in the next calculations.

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 air-cooled stages with the same pressure ratio (CM1 to CM3), intercoolers and condensed water knockouts to dehydrate the CO₂ stream. CO₂ enters the compression station at a pressure depending on regeneration conditions and exits the last stage at slightly supercritical pressure. After recycling the CO₂ necessary for coal loading and further cooling, the supercritical liquid CO₂ stream is ultimately pumped to storage.

The layout of the CCS plant in cooled mode, as shown in Fig. 3, is very similar to the one in Fig. 2, but two main differences have to be pointed out.

- When cooling the gas exiting the HRSG, two contact coolers are sufficient with no chiller.
- Ammonia slip in cooled mode is more significant than in chilled mode, so the load for the absorption-desorption cycle just before the final acid washing (WT1) is higher.

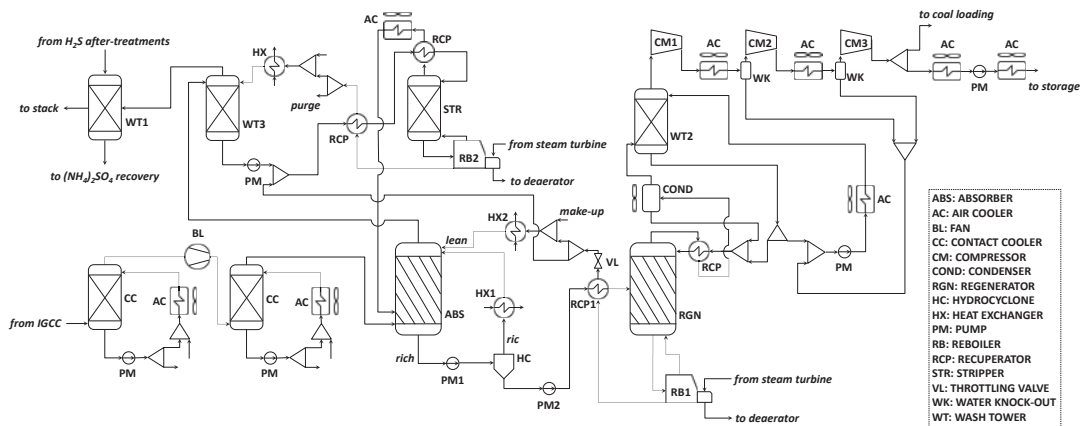


Fig. 3 Schematic of the CCS plant for the cooled case.

When running simulations of the CCS plants in both the chilled and cooled modes, the following operation parameters have been considered:

- the operation temperature at the absorber (chilled or cooled mode),
- the amount of CO₂ captured,

- the initial ammonia concentration (conc) in the aqueous solution, defined as

$$\text{conc} = \frac{\dot{m}_{\text{NH}_3}}{\dot{m}_{\text{NH}_3} + \dot{m}_{\text{H}_2\text{O}}}\bigg|_{\text{lean}} \quad (3)$$

- the NH_3 -to- CO_2 ratio in the absorber (K), as the ratio between the number of NH_3 moles in the lean solution line to the absorption reactor and the number of CO_2 moles in the line from the HRSG outlet:

$$K = \frac{\dot{n}_{\text{NH}_3, \text{lean}}}{\dot{n}_{\text{CO}_2, \text{in}}} \quad (4)$$

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

In particular, setting a regeneration pressure requires to operate at a specific temperature for a fixed level of CO_2 capture, whereas setting the operation temperature at 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.

Table 1. Common parameters for CCS plant calculations

<i>CO₂ capture plant</i>	
Fluid temperature at air-cooler outlet, °C	25
Pressure drop in air-coolers, %	1
Specific consumption for heat rejection by air-cooling, kW _{el} ·MW ⁻¹	15.9
Pinch point ΔT in heat exchangers, °C	5
Coefficient of performance for chilling, MW·MW _{el} ⁻¹	5
Fan polytropic efficiency, %	0.85
Fan pressure ratio	1.1
Pumps hydraulic/mechanical-electric efficiency, %	80/95
Pressure drop in contact coolers, bar	0.01
Pressure drop in columns, bar	0.03
Maximum ammonia in CO ₂ to storage, mg·Nm ⁻³	10
<i>CO₂ compression</i>	
Number of intercooled compression stages	3
Intercooled compressor isentropic efficiency, %	85
Pressure at the last intercooled compressor/pump outlet, bar	80/110
CO ₂ temperature at intercooler outlet, °C	25
Pump hydraulic efficiency, %	75

Details on common parameters assumed for CCS simulations are reported in Table 1, while Table 2 reports the values of specific operation parameters, fixed for the preliminary investigations of energy demand by the CCS plant, based on the experience achieved with former works [2, 4].

According to the preliminary investigations of energy demand by the CCS plant, in both chilled and cooled modes, the best set of operation parameters from an energy-saving point of view for each case has been chosen, as reported in Table 3. In detail, three levels of CO_2 capture (i.e. 80%, 85% and 90%) in both the modes have been considered for the cases presented and discussed in the next section. Cases with amounts of CO_2 captured less than

80% have not been investigated in this work, even though techno-economic considerations are always necessary to set a proper target of CO₂ capture.

Table 2. Preliminary assumptions for the investigated parameters of the CCS plant

Parameter	Values
Absorber operation temperature, °C	7, 20
Amount of CO ₂ captured, %	80, 85, 90
NH ₃ initial concentration (conc), kg/kg	0.05, 0.1, 0.15, 0.2
NH ₃ -to-CO ₂ ratio (K), mol/mol	4, 4.25, ... 5.75
Recycled rich solution (ric), %	10, 20, ... 90
Regeneration pressure, bar	5, 10, 15, 20

Table 3. Key-parameters assumed for CCS plant calculations

	CH-80	CH-85	CH-90	CO-80	CO-85	CO-90
ric, wt%	80	80	80	50	10	10
conc, wt%	20	20	20	10	5	5
K, mol/mol	4	4.25	5	5.25	4.75	4.75
regeneration pressure, bar	15	10	5	5	5	5

5. Results

The results of the IGCC calculations are reported in Table 4, with particular attention paid to power details of the bottoming cycle and to the after-treatment station (for one gasification train). Power balances and overall IGCC performance are reported as well. These results are strictly related to the size of the IGCC plant, which depends on the CT technology at the topping cycle, i.e. on the mass flow rate at the CT outlet (665 kg/s). *CH* and *CO* stand for chilled and cooled mode respectively, whereas numbers 80, 85 and 90 refer to the level of CO₂ capture.

Based on the calculations for the IGCC plants with CO₂ capture in chilled mode, the following results are noteworthy.

- The power output from the steam turbine decreases because of significant steam extraction for the heat duty of the regenerator. On the other hand the heat duty at the NH₃ stripper is very limited. Power requirements of bottoming cycle pumps decrease too, as less steam expands through the LP stages of the turbine. Thus, the heat rejected at the condenser is almost halved.
- The same power for exhaust cooling is required, before the gas enters the CO₂ absorber.
- At the CO₂ capture plant, the heat duty of the regenerator decreases when less CO₂ is removed from the flue gas, even though the operation temperature at the regenerator slightly increases for lower CO₂ capture levels, as a consequence of the regeneration pressure detailed in Table 3. In case of higher levels of CO₂ capture, the number of reacting moles increases, so power requirements for chilling and air cooling increase (the CO₂ chemical absorption is an exothermic process), whereas pumping power nearly halves from case *CH*-80 to case *CH*-90, due to the specific regeneration pressure.
- Different CO₂ compression energy costs in Table 4 depend both on the amount of CO₂ captured and on the CO₂ pressure at the inlet of the compression system.
- Focusing on ammonia slip, the lower absorber temperature prevents large NH₃ slipping in the gas phase and the final acid wash is sufficient to reduce NH₃ to traces after the NH₃ absorption-desorption cycle whose thermal load is really limited. In particular, the NH₃ amount in the gas exiting the absorber ranges from about 0.8 to 1.15 kg/s and increases with the CO₂ captured.
- Paying attention to the overall IGCC results and moving from case *CH*-90 to case *CH*-80, it is possible to appreciate an obvious increase in net power output and LHV efficiency. However, referring to the specific primary energy consumption for CO₂ avoided (SPECCA) defined as:

$$\text{SPECCA} = \frac{3600 \cdot \left(\frac{1}{\eta} - \frac{1}{\eta_{\text{ref}}} \right)}{\text{ER}_{\text{ref}} - \text{ER}} \quad (5)$$

the minimum value is calculated just for case *CH-85*, but no significant difference is appreciated for case *CH-80*.

Based on these results and referring to the performance of air-blown IGCC plants with pre-combustion CO₂ capture [7], where a SPECCA of 2.76 MJ/kgCO₂ was calculated for 87% of CO₂ avoided, chilled ammonia scrubbing does not seem to be a particularly attractive technology, especially if considering the larger size and cost of the post-combustion CO₂ capture plant.

On the other hand, looking at the results reported for the CCS cases in cooled mode, IGCC performance seems to be slightly different. Although similar considerations to the ones for the cases in chilled mode can be repeated, proper attention should now be paid to the absence of chillers, with a significant energy saving, and to the higher heat duty of the NH₃ stripper. As a matter of fact, ammonia slip in cooled mode is larger (up to about 3.8 kg/s for the *CO-80* case), due to the higher operation temperature at the absorber. However, the overall energy demand of the CCS plant decreases if compared to the cases in chilled mode, so some reductions in the SPECCA values are possible with particular reference to the cases with higher CO₂ capture (85% and 90%).

Table 4. Power details for the bottoming steam cycle and the CCS plant (the latter just for one gasification train) and overall IGCC performance

	<i>REF</i>	<i>CH-80</i>	<i>CH-85</i>	<i>CH-90</i>	<i>CO-80</i>	<i>CO-85</i>	<i>CO-90</i>
Steam turbine, MW _{el}	515.8	490.4	491.0	485.2	435.9	433.1	424.2
Steam cycle and condenser HRSC pumps, MW _{el}	13.5	10.8	10.5	10.2	9.8	9.7	9.3
Heat rejected at the condenser, MW	688.0	394.4	366.7	335.9	290.3	278.5	239.9
<i>Exhaust cooling</i>							
Air Cooler and chillers, MW _{el}	-	7.6	7.6	7.6	0.8	0.8	0.8
Fan, MW _{el}	-	5.9	5.9	5.9	5.5	5.5	5.5
Pumps, MW _{el}	-	0.2	0.2	0.2	1.1	1.1	1.1
<i>CO₂ capture plant</i>							
Heat duty at the regenerator, MW	-	179.0	191.1	204.3	183.3	219.1	230.2
Regeneration temperature, °C	-	73.7	70.1	63.9	103.9	104.7	105.9
Heat duty at NH ₃ stripper, MW	-	6.8	8.2	13.4	82.3	53.9	66.6
Temperature for NH ₃ stripping, °C	-	96.9	96.9	96.9	98.3	98.8	98.8
Chillers and air cooler, MW _{el}	-	34.1	36.4	38.5	7.4	6.45	7.4
Pumps, MW _{el}	-	1.9	1.4	1.0	1.5	2.1	2.1
<i>CO₂ compression</i>							
IC compressors and pump, MW _{el}	-	9.8	13.4	19.9	17.5	18.6	19.7
Air coolers, MW _{el}	-	0.5	0.6	0.7	0.6	0.6	0.7
<i>Overall results</i>							
Gross electric power output, MW _{el}	1059.4	1010.3	1010.8	1005.1	955.7	952.9	944.0
IGCC plant auxiliaries, MW _{el}	101.1	75.0	74.7	74.4	73.9	73.8	73.4
CO ₂ C&S plant auxiliaries, MW _{el}	-	119.9	130.8	147.5	68.7	70.4	74.5
Net electric power, MW _{el}	958.3	815.4	805.2	783.1	813.1	808.8	796.1
Net electric LHV efficiency, %	50.03	42.27	41.74	40.59	42.15	41.92	41.27
Specific emissions, kgCO ₂ /MWh	694.8	181.2	137.6	95.0	181.9	136.7	93.0
SPECCA, MJ/kgCO ₂	-	2.57	2.56	2.79	2.62	2.49	2.54

6. Conclusions

The integration between an air-blown IGCC and a post-combustion ammonia-based CCS plant has been assessed. In detail, three levels of CO₂ capture (from 80% to 90%) have been investigated based on operation conditions at the CCS plant set to limit the energy cost of CO₂ capture. In particular, two absorber temperatures have been fixed in order to evaluate a first chilled and a second cooled case. The ammonia slip, which is the main drawback related to ammonia-based CCS technology, is here controlled and reduced to traces by acid wash with the H₂S recovered in the desulphurization unit of the IGCC, resulting in ammonium sulphate.

Compared to the case without CO₂ capture, IGCC efficiency decreases from 7.8 to 9.4 and from 7.9 to 8.8 percentage points when increasing CO₂ capture, in chilled and cooled mode, respectively. Despite of variations in the specific primary energy consumptions for CO₂ avoided calculated for the investigated plants, the cooled ammonia-based CCS technology seems to be attractive, even though more significant potential of this technology is reasonably expected for cases when high-sulphur coal is used as primary feedstock in the power system.

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