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A study of CO_2 capture in advanced IGCC systems by ammonia scrubbing

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Abstract

This paper deals with post-combustion CO_2 capture by aqueous ammonia in air-blown gasification-based combined cycles and follows previous authors' investigations of CO_2 capture by MEA scrubbing. Based on the calculations, CO_2 capture seems to be more penalizing when realized by chilled ammonia instead of MEA. As a matter of fact, chilling down to 7°C both the exhaust gas and the ammonia solution results in significant power consumption of chillers, which is only partly balanced by the lower consumption for CO_2 compression and lower steam extraction from the bottoming cycle compared to the MEA case. Cases with cooled instead of chilled ammonia are investigated as well. In particular, raising the process temperature up to 20°C seems to be an interesting solution, since temperature control in the absorber can be realized by passing the aqueous ammonia solution through an heat exchanger, using ambient-temperature water as refrigerant medium and removing the chillers from the system.

Keywords: Air-blown; IGCC; CO_2 capture; Chilled ammonia; SPECCA

1. Introduction

Coal is a natural resource available in many countries worldwide, whose use for power generation assumes a significant role in the global energy scenario. However, environmental issues require a sustainable use of coal and great efforts for greenhouse gas reduction, so several technologies that can significantly reduce emissions have

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Nomenclature and acronyms

AGR	Acid gas removal	LHV	Lower heating value
ASU	Air separation unit	LP, IP, HP	Low, intermediate, high pressure
CAP	Chilled ammonia process	LT, HT	Low, high temperature
CCS	Carbon capture and storage	MDEA	Methyldiethanolamine
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)		
HRSG	Heat recovery steam generator	TIT	Turbine inlet temperature
IGCC	Integrated gasification combined cycle	η	Efficiency

been developed and are commercially available. Among these clean coal technologies, there is the integrated gasification combined cycle (IGCC), considered as a valid alternative to conventional pulverized coal-fired plants for future power generation systems [1].

An IGCC consists of a coal gasification system, heat exchangers for syngas cooling, a station for fuel gas clean-up and two power blocks with gas and steam turbines. Despite most of the R&D and demonstration projects on large-scale IGCCs are today based on oxygen-blown gasifiers [2], air-blown gasification should also be considered as an option, because of (i) the potentially higher IGCC efficiency, (ii) the economic advantage related to the much smaller ASU required and (iii) the possibility of using low-grade feedstocks [3]. A significant activity on air-blown coal gasification has been conducting during the last years by Mitsubishi Heavy Industries (MHI) in Japan, where the 1700 tpd-250 MW_{el} demonstration plant in Nakoso was started up in 2007, after preliminary research activities on lab-scale gasifiers and pilot plants [4]. According to the first results and considering the use of state-of-the-art G-class combustion turbines for the topping cycle, MHI declares really interesting performance: 600 MW_{el} as gross power and 53% as gross LHV efficiency, for short-term commercial power plants [5].

Nevertheless, carbon capture and storage (CCS) are essential to reduce CO_2 emissions considerably, even though their impact on plant performance is anything but negligible [6]. In the short term, post-combustion CO_2 capture seems to be the most favorable technology, since suitable for both new and existing power plants. An alternative to the more mature flue gas scrubbing with MEA is the Chilled Ammonia Process (CAP), currently commercialized by Alstom. The first conceptual scheme of CO_2 chemical absorption with aqueous ammonia is likely that proposed by Bai et Yeh [7], which is envisioned to have a water wash at the top of both the absorber and the regenerator because ammonia slip is already recognized as a possible drawback. Resnik et al. [8] are probably the first investigators suggesting the use of ammonia solution for the multi-pollutant (CO_2, SO_2, NO_x, HCl and HF) flue gas from fossil fuel-fired plants, an idea that is being pursued by Powerspan Corp. [9]. In 2005 Gal patented the concept of conducting the absorption in chilled conditions (0-20°C), in order to both favor CO_2 capture and limit the ammonia slip [10]. Nowadays, Alstom has licensed the exclusive world-wide rights to market and sell the process patented by Gal [11]. Until about 2009, Alstom designs and operates a pilot plant based on the conventional absorption-regeneration scheme where the regeneration pressure is fairly high (20-40 bar). Subsequently, Alstom redesigns the layout, modifying the way ammonia is recovered from the flue gas and implements it in a few test sites.

The authors' research group has gained significant experience in the field of CO_2 capture technologies with CAP, as detailed in a number of papers: integrations with the power plants are proposed in [12], economic perspectives in [13], whereas different layouts are investigated in [14]. Following a recent authors' study [15], this paper deals with post-combustion CO_2 capture in an air-blown gasification-based combined cycle by ammonia scrubbing, adopting a scheme recently proposed in [14].

2. The IGCC system and the CO_2 capture plant

The air-blown gasification-based combined cycle considered in this work is the one thoroughly investigated in [16], consisting of two gasification trains and two combustion turbines with two heat recovery steam generators (HRSGs), which share the same steam turbine.

Referring to the layout schematically drawn in Fig. 1, a low-sulfur South African coal (64.44% C, 3.95% H, 7.40% O, 1.49% N, 0.85% S, 9.20% H_2O, 12.67% ash; 24.62 MJ/kg LHV) is used as primary feedstock without pre-drying, according to [3]. Coal (stream 6) is loaded with nitrogen (7) produced in a small-size stand-alone ASU based on a pumped liquid oxygen process. The oxygen produced is used to enrich the gasification air and the size of the ASU is determined to obtain 25 vol.% oxygen in the gasifying stream. Air for gasification (9) is extracted from CT compressor outlet and further compressed by a booster. IP steam is produced in the gasifier waterwalls. The syngas (10) exits the gasifier at 1200°C and is cooled down to 400°C (12) by producing HP steam and to 237°C by pre-heating the clean syngas (17) up to 250°C. Syngas scrubbing is carried out for the removal of entrained ash and soluble contaminants. Use of a regenerative heat exchanger is made to pre-heat the syngas exiting the scrubber (15) up to the hydrolysis catalytic reactor temperature, where COS is converted into H_2S. The hydrolyzed syngas is finally cooled down to near-ambient temperature by pre-heating (i) scrubbing water, (ii) LP water for the bottoming steam cycle directed to the deaerator and (iii) the desulfurized syngas exiting the AGR station, where H_2S is removed by means of a MDEA-based process and sent to the sulfur recovery unit. H_2S-free syngas (0.65% Ar, 0.56% CH_4, 27.62% CO, 3.19% CO_2, 10.66% H_2, 0.26% H_2O, 57.06% N_2) is finally heated before fuelling the combustion turbine (17).

Focusing on the power cycles, a state-of-the-art air-cooled combustion turbine is used for the topping cycle, with the same TIT and pressure ratio of the turbomachinery fuelled with natural gas (1335°C and 17). As a matter of fact, according to these assumptions and considering a choked CT expander, syngas would require less air for combustion than natural gas. However, taking air for gasification into account, CT compressor load is not significantly reduced and compressor stall conditions are avoided by means of a little closure of the inlet guide vanes. Heat from gas turbine exhaust and raw syngas cooling is exploited in a two pressure level steam cycle with reheat. LP steam is extracted from the steam turbine to provide the heat necessary at sour water and MDEA regeneration strippers, in the AGR station, and to pre-heat the water directed to the deaerator in a regenerative heat exchanger. Gas exits the HRSG at 115°C, so possible acid condensation should be avoided.

In IGCC systems with post-combustion CO_2 capture, the gas exiting the HRSG (0.9% Ar, 10.27% CO_2, 3.82% H_2O, 75.24% N_2, 9.78% O_2) enters an after-treatment station, before directing to the chimney. This station is subdivided into islands: (i) exhaust chilling, (ii) absorption-regeneration-gas wash, (iii) CO_2 compression, (iv) chilling plant and (v) ammonia removal. The layout schematized in Fig. 2 is an evolution of the one proposed by Alstom and previously investigated for USC plants in [14].

The gas exiting the HRSG passes through a direct contact cooler, so it is delivered by a fan into the exhaust chilling/cooling island. For cases that require lower gas temperature than ambient, an additional cooler with chilled water is present. Later the cooled/chilled gas enters the CO_2 capture station with the absorber AB and the CO_2-rich

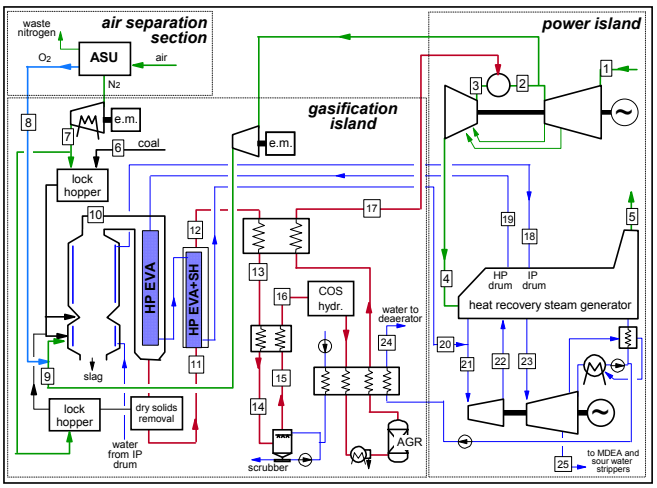

Fig. 1. Layout of the air-blown gasification-based combined cycle

solution is firstly delivered by pump PM21 from the absorber into a hydrocyclone (HC) and secondly, by pump PM22, through the recuperative heat exchanger RC21 to the regenerator (RG). The CO_2-lean solution from the regenerator, after passing through heat exchangers RC21 and HX22, returns to the absorber. The heat of reaction and the temperature in the absorber are controlled by recirculating part of the rich solution from the absorber, cooled in the exchanger HX21. The heat duty for CO_2 stripping is released in reboiler RB21 by steam bled from the bottoming cycle. The gas exiting the absorber contains ammonia that is separated with a chilled water wash in a sub-station with an absorption column (WT21) a recuperative heat exchanger (RC22) and a stripper (ST) with the same operating pressure of regenerator RG, using heat from reboiler RB22. The water-ammonia vapor phase is delivered to regenerator RG. Ultimately, the treated gas passes through an acid wash column, where the residual ammonia is eliminated with H_2SO_4, useful to obtain valuable fertilizer, with negligible impact in terms of energy demand. The stripped CO_2 stream exiting the top of the regeneration column passes through an air-cooled exchanger in order to condensate and separate the water content, before entering the CO_2 compression station. The CO_2 compressor consists of two air-intercooled stages (CM31), followed by an air-aftercooler (AC31) and a condensate knockout (WK) to dehydrate the CO_2 stream, and other two air-intercooled stages (CM32), followed by a final air-aftercooler (AC32), to deliver the carbon dioxide at a slightly supercritical liquid state through a pump (PM31) up to the CO_2 delivery pressure.

In IGCC systems with post-combustion CO_2 capture, part of the carbon dioxide delivered by the CO_2 compressor is recycled back to the gasification island for coal loading, as previously considered in [15] and [17]. Thus, no ASU is necessary and the air blown to the gasifier is not oxygen-enriched. The amount of inert gas used for coal loading is reduced with respect to the case without CO_2 capture [18]. However, using carbon dioxide instead of nitrogen for coal loading results in slight variations in the cold gas efficiency (74.4% vs. 74.88%), as well as final syngas composition (0.67% Ar, 0.53% CH_4, 27.41% CO, 5.84% CO_2, 8.69% H_2, 0.26% H_2O, 56.6% N_2).

3. Calculation methodology

The thermodynamic models of the power cycles were carried out with the simulation code GS, developed since several years by the authors' research group [19], integrated with the commercial code Aspen Plus®.

The GS code was originally designed 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 [20] and has been successfully used to calculate mass and energy balances of a variety of power plant configurations, including gasification processes and other chemical reactors [21],[22]. Table A1 reports the main assumptions for the calculations of the gasification and power islands in the IGCC system with no CO_2 capture. In particular, the cooled gas turbine model presented in [20] was used to simulate and reproduce the performance of a Siemens SGT5-4000F combustion turbine, as state-of-the-art turbomachinery: some data in Table A1 come from model tuning [23].

On the other hand, the commercial code Aspen Plus® was used for calculations related to CO_2 absorption and compression, employing a model that is not built inside the code but defined by the user, as successfully done in

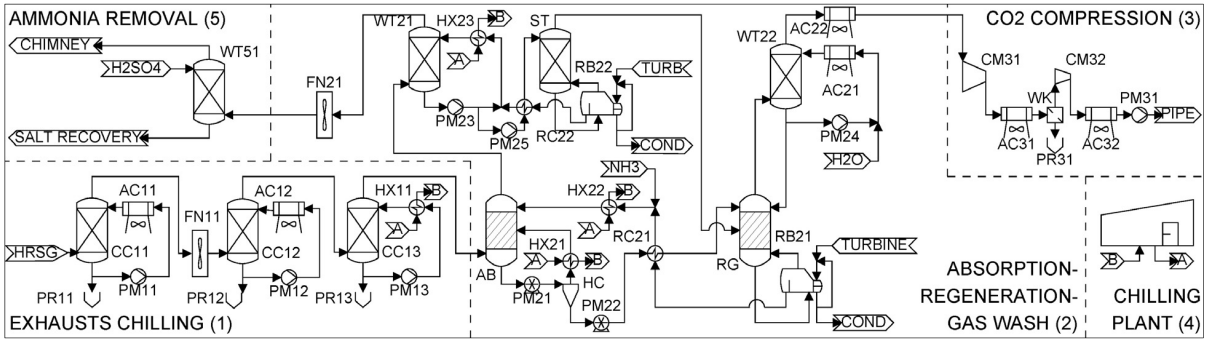

Fig. 2. Layout of the after-treatment station for CO_2 capture

[24]. In particular, the Extended UNIQUAC thermodynamic model for gas solubility in salt solutions developed by Thomsen and Rasmussen [25] was used. It is derived from the original UNIQUAC expression by Abrams and Prausnitz [26], by adding a Debye-Hückel term to account additional excess Gibbs energy from the electrostatic interactions between ionic species. The model requires UNIQUAC volume and surface area parameters for each species, along with temperature-dependent binary interaction energy parameters for each pair of species. Phase equilibrium calculations are performed with the $\gamma - \varphi$ approach coupled with equilibrium speciation reactions with potential solid phase precipitation. The liquid phase activity coefficients are calculated from the Extended UNIQUAC model, while the gas phase fugacity coefficients from the Soave-Redlich-Kwong equation of state. Besides phase relations, the model reproduces also thermal properties, such as enthalpy and entropy, within the experimental accuracy [25]. Table A2 details the main assumptions for CO_2 capture and compression calculations.

In the following, the specific primary energy consumption for CO_2 avoided (SPECCA) is considered as a figure of the energy cost related to CO_2 capture:

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

where the subscript REF stands for the power plant with no CO_2 capture.

4. Results and discussion

In all the systems considered in this study, the expander of the combustion turbine defines the actual size of the IGCC, so different values are calculated for each gasification station, which delivers enough syngas to the choked CT expander.

Table 1 reports general results for the cases without and with CO_2 capture, namely case N and case C. In particular, when considering CO_2 capture, part of the captured and compressed carbon dioxide is used for coal loading, so neither ASU nor lock-hopper compressors are present. On the other hand, air entering the gasifier is not oxygen-enriched and the air booster is unavoidably bigger to blow the amount of oxygen necessary for coal gasification. Thus, CT compressor power increases, reflecting on less power produced by the topping cycle.

Main stream details reported in Table 1 suggest slightly larger size for gasifier, syngas coolers and heat exchangers of the system with CO_2 capture.

Power details of the bottoming cycle and after-treatment station are reported in Table 2, as concerns one gasification train. In particular, cases C1, C2 and C3 refer to CO_2 capture by aqueous ammonia and are characterized by absorption process temperatures equal to 7, 15 and 20°C. When considering CO_2 capture, the following results are worth of attention.

- Steam turbine power reduces, owing to steam extraction for heat duties at the two reboilers (RB21 and RB22).
- Power requirement of bottoming cycle pumps reduces too, owing to less steam expanding in the LP stages of the turbine and delivered to the condenser.
- At the exhaust chilling/cooling station, the same power is required by the fan, with less significant demands by pumps and systems for heat rejection, depending on the selected CO_2 capture process temperature. Before next

Table 1. Power (on the left) and main stream details (on the right) for the IGCC systems without and with CO_2 capture (one gasification train)

	Case N	Case C		Case N	Case C
CT power, MW_{el}	287.8	278.1	Coal to gasifier, kg/s	40.2	40.8
CT auxiliaries, MW_{el}	1	1	Air to ASU, kg/s	32.9	-
Air Separation Unit, MW_{el}	11.4	-	Air from CT compressor, kg/s	115.9	152.8
Lock hopper compressors, MW_{el}	10.7	-	Air at CT inlet, kg/s	648.8	672.4
Air booster, MW_{el}	16.7	22	Syngas to combustor, kg/s	175.9	194
Pulverizers, coal and slag handling, MW_{el}	2.5	2.5	Gas at CT outlet, kg/s	708.7	713.6
Auxiliaries for acid gas removal, MW_{el}	0.7	0.7	Heat recovered in HRSGs, MW	729.9	740.5
Balance of plant, MW_{el}	1.5	1.5	HT heat from gasification islands, MW	443.9	463.2
Thermal input, MW	989.7	1005	LT heat from gasification islands, MW	68.3	85.1

Table 2. Power details for the bottoming cycle and after-treatment station (one gasification train)

	Case N	Case C1	Case C2	Case C3	Case CM
Steam turbine power, MW_{el}	247.1	210.8	210.6	201.1	186.4
Condenser, feedwater and other pumps, MW_{el}	6.4	4.7	5.1	4.7	4.1
Exhaust cooling					
Fan, MW_{el}	-	7.07	7.07	7.07	7.07
Systems for heat rejection, MW_{el}	-	0.93	0.16	0.41	0.72
Pumps, MW_{el}	-	0.21	0.35	0.10	0.03
Absorption-Regeneration Wash					
RB21 heat duty, MW	-	258.12	145.08	169.04	353
RB22 heat duty, MW	-	10.43	82.29	101.13	-
Systems for heat rejection, MW_{el}	-	0.20	0.09	0.47	1.42
Pumps, MW_{el}	-	1.59	0.86	1.71	2.22
CO_2 compression					
Systems for heat rejection, MW_{el}	-	0.56	0.56	0.56	0.83
Intercooled compression, MW_{el}	-	21.03	21.03	21.03	29.84
Chilling plant					
HX11, MW_{el}	-	5.17	4.91	-	-
HX12, MW_{el}	-	2.45	-	-	-
HX21, MW_{el}	-	35.24	27.65	2.45	-
HX22, MW_{el}	-	15.51	15.82	0.96	-
HX23, MW_{el}	-	0.12	4.16	3.42	-
Overall results					
Gross electric power, MW_{el}	1069.8	977.8	977.4	958.4	929.1
Auxiliaries, MW_{el}	101.8	245.1	232.4	142.8	148.4
Net electric power, MW_{el}	968.1	732.7	745	815.6	780.7
Net electric LHV efficiency, %	48.91	36.45	37.06	40.58	38.84
Specific CO_2 emissions, g/kWh_{el}	707.7	106.38	104.64	95.57	99.85
SPECCA, MJ/kg_{CO2}	-	4.18	3.90	2.47	3.14

after-treatments, gas composition is: 0.9% Ar, 10.34% CO_2, 3.13% H_2O, 75.78% N_2, 9.85% O_2.

- At the absorption-regeneration-gas wash station, the heat duty for reboiler RB21 does not present a clear trend, whereas the heat duty for reboiler RB22 increases according to the CO_2 capture process temperature, since high process temperature causes larger amount of ammonia slipped from the absorber. Fixing regeneration pressure at 5 bar, it is possible to strip ammonia conveniently at 148°C. The vapor phase from the stripper ST to the regenerator RG favors a partial recovery of the heat duty initially addressed to the ammonia removal station. In order to respect the target of 90% as CO_2 capture efficiency, the regeneration temperature in reboiler RB21 is calculated equal to 100, 85 and 103°C for cases C1 to C3, respectively, always less than 120°C as for the case with MEA [15]. However, the total amount of heat required by the two reboilers could be optimized, by tuning some process parameters in Table A2, just to limit the high quality steam extraction from the bottoming cycle.
- CO_2 compression cost is obviously the same, since quantity and quality of the gas exiting the HRSG do not vary.
- Cases C1 and C2 are really affected by significant power consumption of chillers. Differences can be appreciated with case C3, where temperature control in the absorber is realized by passing the aqueous ammonia solution through an heat exchanger, where water at ambient temperature is used as refrigerant medium.

According to these results, energy cost of CO_2 capture for systems C1 and C2 is higher than in similar power plants where CO_2 capture is realized with MEA [15]: case CM is reported as well in the extreme right column in Table 2. Referring to IGCC efficiency and SPECCA as figures of merit for the sake of simplicity, the authors calculated values respectively greater and lower than the ones characteristic of cases C1 and C2. However, the different cooling solution adopted for system C3 suggests ammonia scrubbing as an attractive technology for post-combustion CO_2 capture in the investigated energy system. This result is mainly due to (i) lower heat duty for CO_2 and ammonia stripping in reactors RG and ST, respectively, i.e. higher steam turbine power output and (ii) lower CO_2 compression power, due to the higher pressure (5 vs. 1.5 bar) at the inlet of the CO_2 compressor.

5. Conclusions

This study has highlighted that CO_2 capture in air-blown gasification-based combined cycles is more penalizing when realized by chilled ammonia instead of MEA scrubbing, as previously investigated by the authors [15]. As a

matter of fact, chilling down to 7°C both the exhaust gas and the aqueous ammonia results in significant power consumption of auxiliary chillers, which is only partly balanced by the lower demand for CO_2 compression and lower steam extraction from the bottoming cycle compared to the MEA case. Realizing a cooled instead of a chilled process, i.e. raising the process temperature from 7°C to 15°C, does not seem to be convenient, since power consumption of chillers is always significant. However, if temperature control in the absorber is realized by passing the aqueous ammonia solution through an heat exchanger where water at ambient temperature is used as refrigerant medium, an interesting reduction of the auxiliary power demand, compared to the case with the chillers, is possible. In this case, the results are better than the ones achieved when dealing with CO_2 capture by MEA scrubbing [15].

Considering these preliminary results, further investigations will be oriented to better study the effects of ammonia concentration in the aqueous solution, regeneration pressure, exhaust gas recirculation rate, etc., in order to propose the layout of the after-treatment station with an impact as little as possible on IGCC performance.

Appendix

Table A1. Main assumptions for calculations of gasification and power islands

Gasifier		***Combustion turbine***	
Gasification pressure, bar	25.3	Compressor pressure ratio	17
Combustor/reductor temperature, °C	1900/1200	Compressor polytropic efficiency, %	92.25
Heat to membrane walls, % of input coal LHV	2	Turbine cooled/uncooled stage efficiency, %	93.3/91.3
Carbon conversion, %	99.9	Turbine inlet temperature, °C	1335
Pressure/temperature of gasifying air, bar/°C	29.8/530	Heat loss at combustor, % of fuel LHV	0.9
N_2 pressure at lock hoppers, bar	50.6	Air pressure loss, %	3
N_2 to coal mass ratio at lock hoppers	0.53	Compressor leakage, % of the inlet flow	0.75
ASU		CT auxiliaries, % of gross power	0.35
O_2 purity, % mol.	95	Turbine/compressor mechanical efficiency, %	99.865
O_2 pressure/temperature, bar/°C	48/15	Electric generator efficiency, %	98.7
N_2 pressure/temperature, bar/°C	1.2/15	***Recovery Steam Cycle***	
ASU electric consumption, kWh/kgO_2	0.423	HRSG gas side pressure loss, kPa	3
CGCU station		Heat loss, % of transferred heat	0.7
COS hydrolizer temperature, °C	180	Pressure levels, bar	130/36
Temperature of absorption tower, °C	35	Maximum live steam temperature, °C	565
CO_2 removed along with H_2S, molar ratio	1.1	Minimum pinch point ΔT, °C	10
MDEA regeneration stripper (net of Claus plant), MJ of steam at 6 bar per kg of removed H_2S	20	Subcooling ΔT, °C	5
		Minimum stack temperature, °C	115
Auxiliaries for sulfur removal and recovery, MJ_{el}/kgH_2S	2	Pressure losses in HP/IP economizers, bar	16/25
Sour water stripper, kJ of steam at 6 bar per MJ of input coal LHV	15	Pressure loss in super-heaters, %	8
		Condensing pressure, kPa	4
Other auxiliaries		Power for heat rejection, MW_{el}/MW	0.01
Coal milling and handling, kJ_{el}/kg_{coal}	50	Pumps hydraulic efficiency, %	80
Slag handling, kJ_{el}/kg_{slag}	100	Turbine mechanical efficiency, %	99.5
BOP, % of input coal LHV	0.15	Electric generator efficiency, %	98.7

Table A2. Main assumptions for calculations of the after-treatment station

Air coolers		***CO_2 compression***	
Ambient temperature, °C	15	Compressor isentropic efficiency, %	85
Fluid end temperature, °C	25	Last compressor end pressure, bar	80
Relative pressure drop, %	1	Final delivery pressure, bar	110
Specific electric consumption, $kW_e \cdot MW_{th}^{-1}$	15.9	***Common capture condition***	
Heat exchangers and chilling plant		Initial ammonia concentration, wt/wt	0.1
Minimum temperature difference, °C	5	Regeneration pressure, bar	5
Coefficient of performance, $MW_{th} \cdot MW_e^{-1}$	5	Pressure drop in contact coolers, bar	0.01
Fans, pumps and motors		Pressure drop in other columns, bar	0.03
Fan polytropic efficiency, %	0.85	CO_2 capture efficiency target, %	90
Fan pressure ratio	1.1	Maximum ammonia in final gas, $mg \cdot Nm^{-3}{}_{6\%O2}$	100
Pumps hydraulic/mech.-electric efficiency, %	80/95	Maximum ammonia in CO_2 to storage, $mg \cdot Nm^{-3}$	10

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