Pre-combustion CO_2 capture by MDEA process in IGCC based on air-blown gasification

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1. Introduction

Current systems for large power generation are based on the combustion of fossil fuels that makes them the largest source of anthropogenic CO_2 emissions. Integrated gasification combined cycles (IGCCs) have the potential to meet high efficiency and low emission targets and are hence considered as valid alternative to conventional pulverized coal plants for future power generation systems, especially when CO_2 capture processes are included.

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Coal gasification is the core of an integrated gasification combined cycle (IGCC). Although most of the R&D and demonstration projects on large-scale IGCCs are based on oxygen-blown gasifiers, air-blown gasification should also be considered as an option, because of the potentially higher efficiency and the economic advantage related to the much smaller ASU [1,2]. 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 [3], after preliminary research activities on lab-scale gasifiers and pilot plants. According to the first results and considering further technological improvements, such as the use of a state-of-the-art G-class combustion turbine for the topping cycle, MHI declares outstanding net plant efficiencies of $48\%_{LHV}$ [4].

On the basis of public information from MHI, a thermodynamic assessment of an air-blown gasification-based combined cycle was carried out by Giuffrida et al. [5]. After reproducing the mass and energy balance of a large-scale MHI-type air-blown gasifier, a complete IGCC power plant was proposed and its performance compared to the one of an IGCC with an oxygen-blown Shell-type gasifier, calculated with coherent assumptions and using the same state-of-the-art combustion turbine as topping cycle. A gross efficiency of 54%, very close to the value indicated by MHI, was calculated, along with a net efficiency resulting 1.5 percentage points higher than the one characterizing the reference oxygen-blown IGCC [5]. Further studies on air-blown IGCC systems were carried out, focusing on (i) pre- and post-combustion CO_2 capture [6,7], (ii) solutions based on hot fuel gas clean-up technology [8] as well as (iii) the exploitation of low rank coals as primary feedstock [9].

This paper deals with pre-combustion CO_2 capture in air-blown IGCC systems, paying particular attention to the acid gas removal station of the power plant and revising former calculations based on a preliminary estimation of electric consumptions and heat duties [7].

Nomenclature

ASU	Air separation unit
CT	Combustion turbine
HP/MP/LP	High pressure / medium pressure / low pressure
HRSG	heat recovery steam generator
HT/LT	High temperature / low temperature
MDEA	Methyl di-ethanol amine
SPECCA	Specific primary energy consumption for CO_2 avoided [MJ_{LHV}/kg_{CO2}]
WGS	Water gas shift

2. Power plant description

The basic air-blown IGCC system considered in this work is similar to the one formerly investigated by the authors [7] and consists of two gasification trains and two combustion turbines (CTs) with two heat recovery steam generators (HRSGs), which share the same steam turbine. A schematic plant configuration is reported in Fig. 1. For the description of the two-stage air-blown gasification technology, reference to previous works [5-9] is here made, for the sake of brevity.

Coal loading is realized by means of a fraction of the CO_2 captured in the AGR station and recycled back (stream 7), so that the ASU adopted in the power system with no CO_2 capture to produce the pure N_2 for coal feeding [5] is not employed here. The syngas exiting the gasifier at 1200°C (9) is cooled down to about 350°C (11) by producing HP super-heated steam. Syngas is then further cooled down to 150°C (12) before scrubbing, with economization of HP water. From the scrubber outlet, syngas enters the sour WGS station, whose main components are two reactors and two heat exchangers, necessary to both pre-heat the syngas and recover the heat of the exothermic water-gas shift reaction. The steam necessary for the WGS reaction is extracted from the steam turbine and calculated to obtain a steam to carbon (as CO) ratio equal to 1.5 at the HT-WGS reactor inlet. The relevance of this assumption was thoroughly discussed in [7]. The syngas exiting the scrubber is firstly pre-heated in a recuperative gas-gas heat exchanger and then mixed with MP steam (13), extracted from the bottoming cycle, before entering the first WGS reactor. The shifted syngas exiting the HT WGS reactor at temperature of 480-490°C is firstly cooled down to about 350°C by producing HP super-heated steam and used as the hot stream in the regenerative gas-gas heat exchanger to pre-heat the syngas exiting the scrubber. Then, the shifted syngas at 210°C (14) enters the LT-WGS reactor to complete the conversion of CO into CO_2. An overall CO conversion as high as 97.4% is obtained in the WGS section and more than 95% of the total carbon in the shifted syngas is finally present as CO_2.

The shifted syngas exiting the LT-WGS reactor is then cooled down to 150°C (16) by heating the H_2-rich stream fuelling the combustion turbine. It is further cooled down to near-ambient temperature for acid gas removal, releasing heat for pre-heating the clean syngas from the AGR station and water for the steam cycle and for syngas scrubbing. Acid gases removal is carried out by means of a MDEA-based process. Here, H_2S and CO_2 are removed as separate streams and sent to a Claus unit for sulfur production and to permanent storage, respectively.

Finally, the clean syngas heated up to about 230°C (17) fuels the combustion turbine. Two CT technologies, representative of a state of the art machine and an advanced one, have been considered. The two technologies differ in the TIT, respectively equal to 1305°C and 1360°C, and the required fuel overpressure, respectively 5 and 10 bar overpressure respect to the air pressure. Therefore, the CT technology also affects the pressure at which the gasifier and the syngas cooling and treating section have to operate and the related electric consumption of the air booster. The CT exhaust heat is recovered in a two pressure level steam cycle with reheat, where the HP level is always fixed to 144 bar and the MP level is fixed to 36/40 bar, depending on the combustion turbine technology. This difference in the MP level is necessary in order to match the pressure required at the WGS station, which depends on the CT technology. The steam for the water-gas shift process is in fact always extracted from the HP cylinder outlet.

The contribution of the syngas coolers to the total steam cycle heat input is really significant for this IGCC with CO_2 capture, owing to (i) the huge presence of nitrogen in the syngas exiting the gasifier, (ii) the increased syngas flow rate with respect to the case with no CO_2 capture [5], (iii) the exothermic water-gas shift reaction and (iv) the increased syngas flow rate due to the injection of warm steam upstream of the WGS station. Another slight contribution to the steam cycle comes from the gasification air cooling before its boosting.

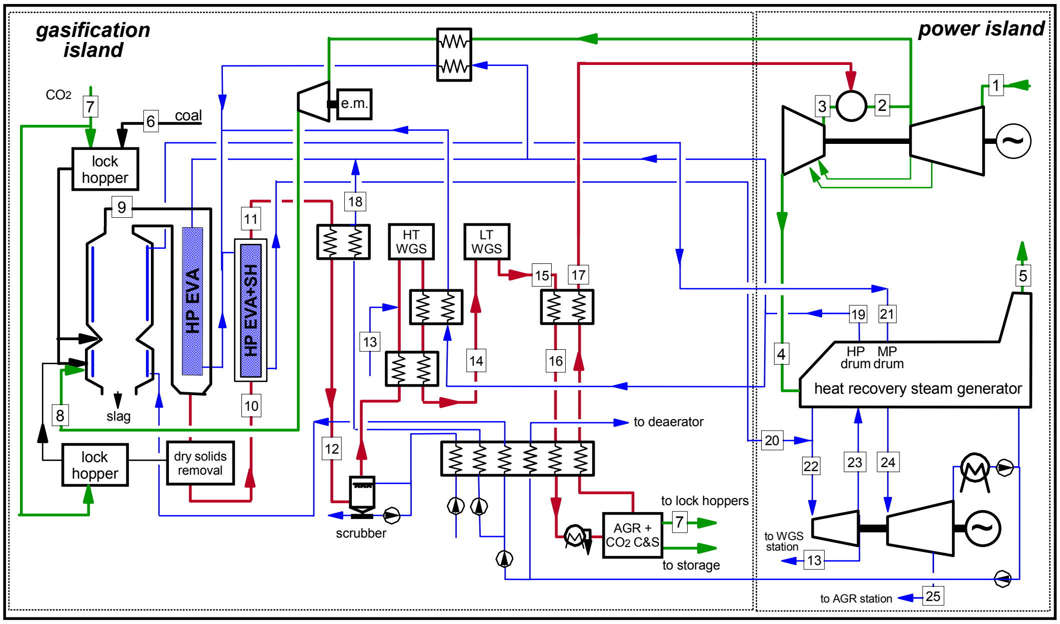

Fig. 1. Configuration of the IGCC power plant with pre-combustion CO_2 capture.

3. The acid gas purification section

The sour gas to be treated is rich in CO_2, with a certain amount of H_2S. The main characteristics (mass flow, molar flow, temperature, pressure, composition) of the stream entering the acid gas removal section are reported in Table 1, for the two cases involving the two CT technologies.

Table 1. Characteristics of the stream entering the acid gas removal section for the two analyzed cases.

characteristic	Case 1 (state of the art CT)	Case 2 (advanced CT)
Temperature, K	308.15	308.15
Pressure, bar	29.32	24.11
molar flow, kmol/sec	9.3855	10.4821
mass flow, kg/sec	232.86	261.48
Composition (mole fraction)		
H_2S	0.000700	0.000700
CO_2	0.252245	0.251270
H_2O	0.001920	0.002330
CH_4	0.004160	0.004120
CO	0.007750	0.007430
H_2	0.278364	0.272470
N_2	0.449531	0.456270
Ar	0.005330	0.005410

The aim is to remove carbon dioxide, whose amount is high (about 25%) before sending the H_2-rich syngas to the combustion turbine. Industrially, the most commonly used process for CO_2 capture is absorption by alkanolamines, widely applied also in natural gas treating plants and in exhaust gas purification plants.

According to the EBTF [10], the obtained CO_2-rich gaseous stream to sequestration should contain at least 90% mol. of carbon dioxide, and at most 200 ppm(v) of H_2S. Despite the H_2S/CO_2 ratio is relatively low (about 0.0028), it is not low enough to allow a content of $H_2S < 200$ ppm(v) in the CO_2-rich stream without a selective H_2S-CO_2 separation. As a consequence, hydrogen sulfide and carbon dioxide should be removed separately. In order to achieve this goal, the absorption plant has been divided into two sections: the gas is fed to a first absorber where most of H_2S is removed, and then it is treated in a subsequent section, where most of CO_2 is absorbed. The H_2S concentration in the H_2S rich gas, moreover, should be at least the one required for feeding a Claus plant. Because of its high selectivity towards H_2S, fundamental to run this plant configuration, a MDEA solution in water has been selected: by exploiting the different rates of reactions of the two species with the amine solvent, if proper residence times are chosen, most of CO_2 can remain in the purified gas.

Though being the process widely industrially used, the design of the absorber is still difficult, due to the different phenomena involved. Commercially, several process simulators can be found, based on different assumptions, both for thermodynamics and for diffusion with reaction, but they are not always as reliable as needed [11].

3.1. Modeling

An accurate description of the phenomenon involved in amine scrubbing systems, in terms of thermodynamics, kinetics and mass transfer, is fundamental for a reliable representation of the absorption process. Amine scrubbing involves the presence of chemical reactions occurring in the liquid phase. The thermodynamic description should take into account chemical equilibrium reactions, while the phenomenon of diffusion with reaction should be modeled considering also kinetic-controlled reactions, because reactions involving carbon dioxide, though being quite fast at the conditions of the system, do not attain chemical equilibrium. Acid gases and amines are weak electrolytes and they partially dissociate in the liquid phase, whose main component is water. In the liquid phase, then, water and amine are in mixture with very volatile molecular species (carbon dioxide and hydrogen sulfide) and non-volatile ionic species, not present in the vapor phase. Generally, when dealing with process simulations, a proper thermodynamic model is fundamental to model vapor-liquid equilibrium [12,13].

To describe the vapor-liquid equilibrium of this strongly non-ideal system, a γ/ϕ approach has been used. The Electrolyte-NRTL model (already implemented in ASPEN Plus®), developed by Chen and co-workers [14-17] and with *ad hoc* parameters previously obtained by regression of experimental data [18,19], has been used for the representation of the thermodynamic behavior of the system. Kinetics and mass transfer can be described by means of the "equilibrium based stage efficiency" approach or of the "rate based" one [20]. The former corrects the performance of a theoretical stage by using a stage efficiency, while the latter takes into account the real mass and heat transfer phenomena that occur on a real tray or on an actual packing height. The "equilibrium-based stage" approach is widely used also in amine scrubbing modeling, because it is not demanding in terms of computational time, however, in reactive systems, reactions affect both the vapor-liquid equilibrium relationship and the rate of absorption, so the stage efficiency cannot be kept constant along the column. The "rate-based stage" approach is more CPU consuming and convergence is not always easily achievable, because of the large number of simultaneous equations to be solved, but it is considered the most reliable method for amine scrubbing modeling [11].

In this work, ASPEN Plus® has been used as process simulator. It is provided by default with the "rate-based" approach based on the film theory [21], however it has been properly modified in order to take into account a different mass transfer theory. Indeed, a deep insight into the mass transfer phenomenon has shown some inaccuracies of the film theory in describing the mass transfer coefficient, when applied to amine scrubbing processes [22]. A different model, based on the Eddy Diffusivity theory [23] and on the Interfacial Pseudo First Order assumption, able to give the correct dependence on diffusivity of carbon dioxide for the mass transfer coefficient, had been previously developed [24-27] and implemented in a subroutine linked to ASPEN Plus®, used as a framework for simulation.

3.2. Simulations of the acid gas removal section

A possible scheme for the purification section, able to meet the specifications for the purified gas and for the concentrated acid gas streams, while allowing a reasonable energy requirement, has been studied. Two absorption sections, with two different amine concentrations and two different regeneration sections have been designed, each one with particular features aimed at meeting all the required specifications, considering an overall CO_2 removal of 95%. Fig. 2 and Fig. 3 show, respectively, the H_2S and CO_2 removal sections for Case 1 as simulated in ASPEN Plus®. For both the cases, two parallel trains are taken into account, each one fed with half the flow rate reported in Table 1.

The gaseous stream coming from cooling after the water gas shift stages is mixed with a recycle stream coming from the Claus plant (not simulated in this work) and fed to the absorption column (*ABSH2S*), with the aim of selectively removing most of all the contained H_2S. The lean amine solution (*LEANHP*) enters the top of the column and absorbs almost all hydrogen sulfide and a low amount of carbon dioxide, and exits the absorber rich in acid gases. The absorption column has been designed so that most of CO_2 is left in the purified gas.

The regeneration section is the most energy demanding section of the acid gas removal plant: it has been found that, for coal-fired power plants, CO_2 removal from flue gases can consume up to 30% of the produced power in the plant [28]. A careful design of the section is then fundamental in order to avoid uneconomic energy demands [29]. The regeneration unit is composed of a distillation column, preceded by a separator (*FLASH*), where the rich solution is flashed in order to recover a portion of the H_2S and the co-absorbed CO_2, before sending the amine to the column. Moreover, a cross heat exchanger *(CROSS*), with the aim of transferring heat from the hot regenerated lean solution exiting the reboiler of the distillation column to the cold rich solution flowing to the regeneration section, is added. Heating the rich solution to be regenerated can be of great help in reducing the amount of heat required at the reboiler.

The regenerated amine solution is mixed with fresh MDEA or fresh water, if a make-up is required, and recycled to the absorption column, while the gaseous stream *H2S* fulfills the characteristics for being fed to a Claus plant [20]. It has been supposed that the gases leaving the Claus plant still contain 4% of the fed H_2S and 100% of CO_2

(unreacted) and are treated by recycling them to the absorption column, after compression to the operating pressure of the absorber (*CLAUSRHP*).

The scheme shown in Fig. 2 can be run both for Case 1 and for Case 2, by simply changing the calculated required duty at the reboiler of the two trains is 9.1932 MW for Case 1 and 10.0137 MW for Case 2, much lower than the one required in the CO_2 removal section.

The free amine solution is composed of 10% wt. MDEA and 90% wt. H_2O. This composition is not similar to the one usually found for MDEA solutions [20], but it has been determined in order to satisfy the imposed specifications and to guarantee an acceptable G/L ratio. This choice, indeed, both for Case 1 and for Case 2, can make the column suitable to treat the gas flow rate, which is very high if compared to one of the amine solution circulating in the H_2S removal section.

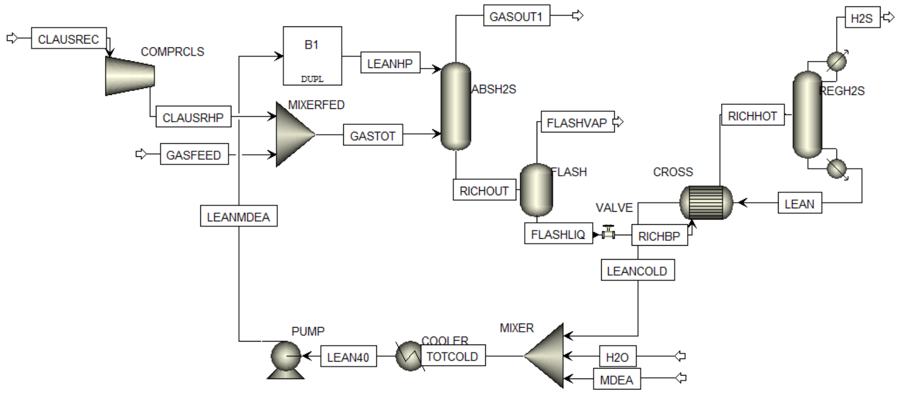

Fig. 2. Scheme of the H_2S removal section as simulated for Case 1.

The gaseous streams *GASOUT1* and *FLASHVAP* produced in the H_2S removal section are sent to the CO_2 removal section, mixed (after the stream *FLASHVAP* has been compressed to the same pressure of stream *GASOUT1*) and fed to the absorption column with the aim of removing CO_2.

A scheme has been studied in order to achieve a 95% removal (of CO_2 entering the acid gas removal section) without exceeding in the energy requirement of the regeneration section. As can be seen in Table 1, Case 1 and Case 2 differ mainly for pressure and for the total flow rate, while the composition is almost the same. In particular, the amount of carbon dioxide to be removed is higher, but the partial pressure, which influences the driving force for mass transfer, is lower in Case 2 than in Case 1. This causes a difference in the plant regarding CO_2 removal, in particular as for the absorption section.

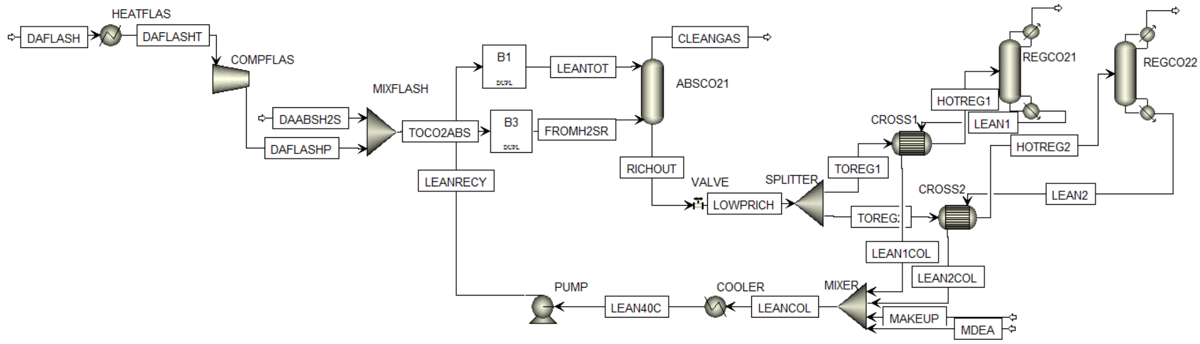

Fig. 3. Scheme of the CO_2 removal section as simulated for Case 1.

Fig. 3 shows the scheme for Case 1: a lean typical MDEA solution (50% wt. MDEA, 50% wt. H_2O) is fed to one absorption column and removes 95% of the CO_2 contained in the gas fed to the acid gas purification section. It is then regenerated in two parallel distillation columns (*REGCO21* and *REGCO22*) operating at atmospheric pressure, with two parallel economizer heat exchangers (*CROSS1* and *CROSS2*) with $\Delta T_{approach} = 10$ K.

An absorption column with a diameter of 6 m and 51 real trays, fed with 404.5 kg/s of lean amine solution (total lean loading = 0.0205) can guarantee the desired conditions and a consumption of 1.28 MJ per kg of absorbed CO_2. For lower amine flow rates a CO_2 removal of 95% would not be achieved (Fig. 4a), while for higher flow rates a higher reboiler duty would be required (Fig. 4b).

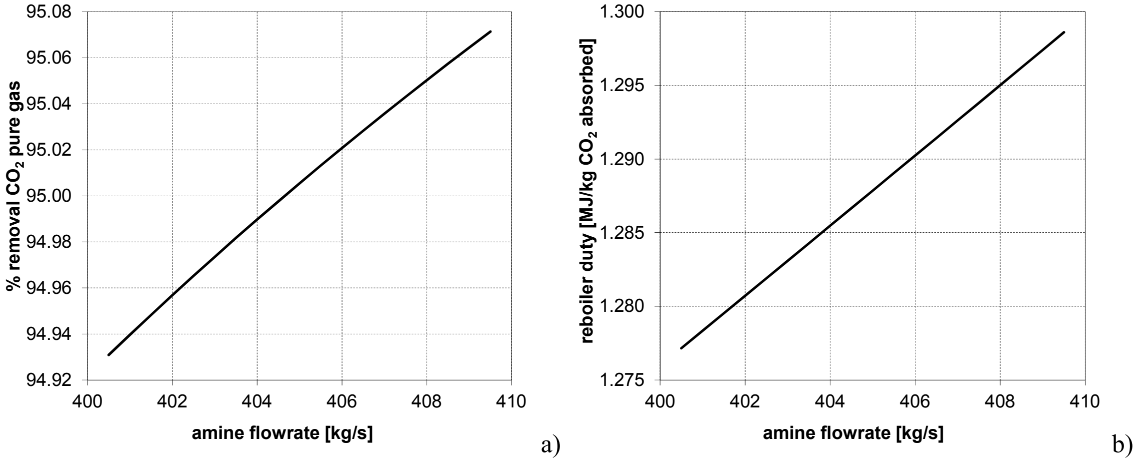

Fig. 4. a) % removal of CO_2 and b) reboiler duty vs. amine flow rate obtained for open loop simulations of Case 1.

For Case 2, a higher amount of amine solution is not sufficient to overcome the lower driving force. Therefore, a different scheme should be taken into account, with two parallel absorption columns (with the same height and diameter as the one of Case 1) each one treating half the flow rate fed to the CO_2 removal section. The amine solution (lean loading = 0.0206) is fed to each column with a flow rate of 235.5 kg/s, exits with a rich loading equal to 0.6525 and is regenerated in one regeneration column, requiring 1.32 MJ per kg of absorbed CO_2, slightly higher than the one required in Case 1.

4. Results

The overall results of the plants assessed are reported in Table 2. For the base case with the current CT technology, a net efficiency approaching 48% has been calculated, as declared by MHI [4]. In the CO_2 capture case, net efficiency reduces to 38.4%, with a penalty of 9.5% points. Such efficiency loss is largely due to the reduction of the gross efficiency (-7.5% points), consequence of the lower cold gas efficiency (i.e. to the portion of the LHV input converted into LHV of the CT fuel) and the high penalties for the steam cycle. The CGE reduction is mainly due to the exothermic WGS reactions, which convert part of the LHV of the CO into heat. Penalties for the steam cycle are instead due to the steam extractions to provide the necessary steam for the WGS and for the MDEA stripper reboiler. To these penalties, consumptions for CO_2 compression have to be added, responsible for a further efficiency decay of 3.4% points. Such penalty takes into account the compression power for the CO_2 used in the lock-hopper system for coal feeding. On the other hand, the lack of the ASU partly balances such consumption.

When considering the advanced CT technology, the net plant efficiency rise by about 2% points for both the non-capture and the capture cases. Net efficiencies approaching 50% without capture and achieving 40.5% with capture have been obtained. As a result of such efficiency decays and of the selected CO_2 capture efficiency, CO_2 avoided of about 87% are calculated. If the efficiency decay and the specific emission reduction are considered in the SPECCA index (specific primary energy consumption for CO_2 avoided), values of 2.95 and 2.76 MJ_{LHV}/kg_{CO2} can be calculated, representing an interesting result when compared to benchmark oxygen-blown IGCC with CO_2 capture by physical absorption [10].

Table 2. Power balance of the IGCCs assessed and main global performance indicators.

	Current CT technology		Advanced CT technology	
	w/o capture	with capture	w/o capture	with capture
Power balance, MW_e				
Combustion turbine	227.2	226.4	270.9	275.4
Steam turbine	238.4	243.9	256.8	261.1
Steam cycle pumps	-1.66	-2.35	-1.76	-2.38
Auxiliaries for heat rejection	-4.70	-5.09	-5.01	-5.38
ASU and N_2 lock-hoppers compression	-20.68		-22.52	
Air booster	-19.67	-32.71	-16.14	-27.85
MDEA pump		-1.28		-1.22
CO_2 compression		-34.04		-37.72
Other auxiliaries	-3.51	-4.13	-3.88	-4.57
Gross power, MW	465.6	470.3	527.7	536.5
Net power, MW	415.4	390.7	478.4	457.4
Heat input, MW_{LHV}	867.4	1018.2	957.7	1128.2
Gross efficiency, $\%_{LHV}$	53.68	46.18	55.10	47.55
Net efficiency, $\%_{LHV}$	47.89	38.37	49.95	40.54
Cold gas efficiency, $\%_{LHV}$	74.96	67.13	74.18	66.20
Specific emissions, kg_{CO2}/MWh	727.0	94.90	696.1	89.37
CO_2 avoided, %		86.95		87.16
SPECCA, MJ/kg_{CO2}		2.95		2.76

5. Conclusions

Four IGCC power plants based on air-blown gasification have been studied in this work through process modelling and simulation. Plants without CO_2 capture and with CO_2 capture by MDEA process have been assessed. MDEA absorption process was defined and accurately modelled, based on a two-stage process to selectively remove H_2S and CO_2. Net efficiency penalties of about 9.5% points have been obtained for the plants with capture for CO_2 avoidance of about 87%, in line with values reported in the literature for more conventional IGCCs based on oxygen-blown gasification and benchmark CO_2 absorption processes.

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