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Offgas Treatment downstream the Gas Processing Unit of a Pulverised Coal-Fired Oxyfuel Power Plant with Polymeric Membranes and Pressure Swing Adsorption

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Abstract

Due to climate change it is necessary to reduce anthropogenic climate gas emissions. The application of carbon capture and storage (CCS) technologies could be a suitable approach to lower the specific CO_2 emissions from coal-fired power plants. One of these CCS technologies is the Oxyfuel process. In the Oxyfuel process the coal is burned in a mixed atmosphere of O_2 and recycled flue gas. The flue gas thus generated has a high CO_2 concentration, because of the missing air nitrogen. Still the dried flue gas consists of approximately 15 mol-% impurities (O_2, N_2, Ar, NO_x and SO_x). To increase the CO_2-purity the flue gas is treated in a gas processing unit (GPU). Two promising technologies to perform the gas processing are partial condensation and distillation. Both are well known and available at industrial scale. Using these technologies about 90 % of the CO_2 can be separated. The remaining part of the CO_2 leaves the GPU with the offgas.
To increase the overall capture rate of the CO_2 in the Oxyfuel process the offgas from the GPU can be treated in either a pressure swing adsorption (PSA) cycle or a polymeric membrane (PM) cycle. These cycles generate a CO_2-enriched GPU-recycle stream and an exhaust gas stream which consists of the residual impurities. The CO_2-enriched GPU-recycle can be fed back to the GPU or mixed with the CO_2 product stream of the GPU. The exhaust gas stream with the impurities has a high content of O_2 and could be refed to the air separation unit (ASU) to increase the efficiency of the overall process. The additional gas treatment in the PSA- or the PM-cycle has influences on the specific energy demand of the GPU, the CO_2 capture rate, the composition of the CO_2 product stream and the overall process efficiency.
In the work presented here the feed gas of the GPU is the flue gas of a large scale bituminous coal-fired Oxyfuel power plant. The plant model is based on the actual state-of-the-art power plant technology. For the GPU two different reference process cases are modelled. One case with a distillation of the CO_2 and one case with a partial condensation of the CO_2 are considered. For both cases the GPU process is externally cooled. These reference cases are compared then with a distillation and a partial condensation which have an additional offgas treatment by PSA or PM. For the offgas treatment with membranes, polymeric membranes are considered due to their high CO_2/O_2-selectivity and high permeability. For the offgas treatment with PSA a multiple bed cycle is modelled to assure continuous operation of the plant. The overall CO_2 capture rate, the specific energy demand and the composition of the CO_2 product stream are calculated for the reference cases, the distillation with PSA or PM and the partial condensation with PSA or PM. With these results the potential of these technologies for the GPU shall be compared with the reference cases. Furthermore a recycle of the O_2-containing gas stream to the ASU is modelled in the overall process model. This recycled gas stream can be used to reduce the energy demand of the ASU. The influence of the offgas treatment is evaluated by calculating the net efficiency of the overall process.

Keywords: CO_2 capture; Oxyfuel; Membrane; CO_2 adsorption

1. Introduction

Climate change makes it necessary to reduce anthropogenic climate gas emissions. The electrical power generation economy is a big climate gas emitter. Especially coal-fired power plants emit huge specific amounts of climate gases (750–1,000 g_{CO2}/kWh_{el}). In the long term fossil power plants could be replaced by less emitting technologies. In the midterm they are indispensable, however, to ensure a reliable supply with electrical power. Two possibilities can be pursued to reduce the specific emissions of fossil power plants. On the one hand an improvement of the overall process and the technologies applied lead to an increase in efficiency of the power

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plants and thus to a CO_2-reduction. To reduce the emissions further and to much lower levels it is possible to capture CO_2 from the flue gas and to inject it underground.

In this study the Oxyfuel process is used to capture the CO_2 in the power plant (see Fig. 1). Purpose of this process is to increase CO_2 concentration in the flue gas, so that less energy is needed to separate the CO_2 from it. The nitrogen content is reduced by burning the coal in an atmosphere of almost pure O_2 and recycled flue gas. The oxygen is supplied by a cryogenic air separation unit (ASU). To realise a manageable temperature on the flue gas side (furnace exit temperature, limited by ash softening temperature) and the water steam side (maximal steam temperature at furnace outlet in the membrane wall, limited by allowable material temperature), about 2/3 or more of the flue gas has to be recirculated [13], [14]. The Oxyfuel process leads to an efficiency decrease of 8-10 % pts., considering an overall CO_2 capture rate of 90 %, compared to modern coal-fired power plants. The main components which are responsible for the efficiency decrease are the ASU and the gas processing unit (GPU).

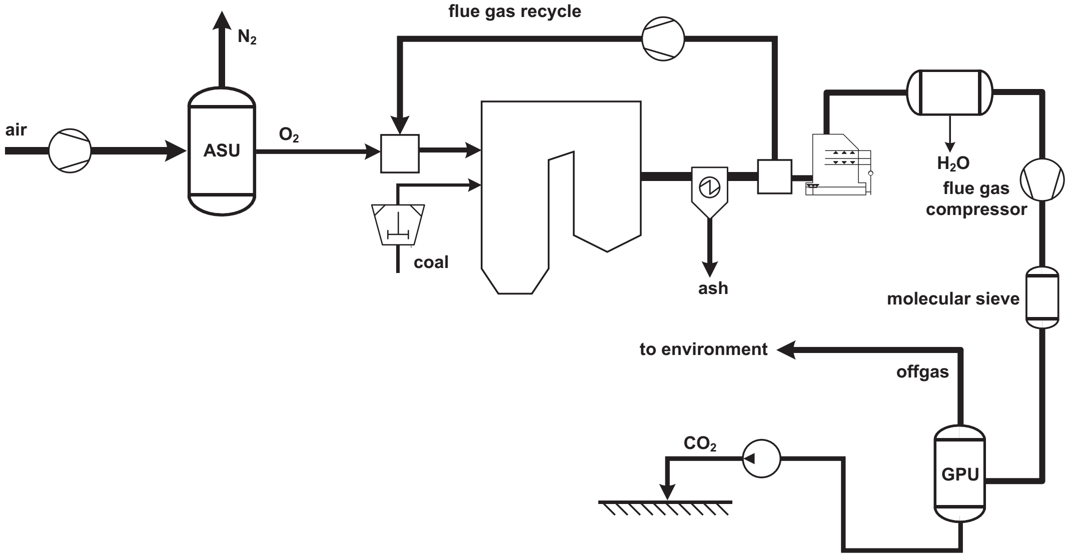

Figure 1: Process flow sheet of the combustion process with the main components of the Oxyfuel basic process. The flue gas that is recycled to transport the coal is ash- and sulphur-free. The flue gas recycled to control the combustion temperature is ash-free.

Current research aims to increase the overall process efficiency of the Oxyfuel process and to increase the CO_2 capture rate. One proposal is to apply an additional gas treatment to the offgas leaving the GPU [2], [5], [6]. The target is to capture most of the remaining CO_2 from this gas stream. Polymeric membranes (PM) and pressure swing adsorption (PSA) are promising technologies to capture the CO_2 from the offgas. Furthermore the remaining oxygen within the offgas can be used to decrease the energy demand of the ASU. The influence of this additional treatment on the capture rate and the overall process is evaluated in this work.

2. Approach followed

The overall process of the power plant is modelled with the commercial software tool *EbsilonProfessional*™. Due to the need for a more detailed physical and chemical data set, the GPU is modelled in *AspenPlus*™. To gain information about the overall process including the GPU, the two models are connected with a software link. Within one simulation time step of the overall process the GPU is calculated simultaneously with the rest of the power plant.

2.1 Overall process modelling

For the overall process a coal-fired power plant with an Oxyfuel adaption is used. The plant is fuelled with South African bituminous coal (see Table 1). The plant modelled in this study represents the current state-of-the-art for a new build plant. Therefore the base process is designed with parameters taken from [1]. The data of the water/steam cycle are shown in Table 2.

For the Oxyfuel adaption of the power plant process only mature technologies are taken into account. Therefore a cryogenic ASU is applied to deliver the oxygen for the combustion. The purity of the oxygen is kept at 95 vol.-% (dry) to minimise ASU energy demand [2]. The compression heat is integrated into the feed water preheating. The oxygen is preheated to a temperature of 183°C. The overall air ingress is estimated to 2 % of the flue gas mass flow upstream the recirculation. The pulverised fuel is transported into the combustion chamber with recycled, ash- and sulphur-free flue gas, while the mill classifier temperature is kept at 120°C. The flue gas recycle into the combustion chamber is ash-free. The remaining flue gas is dried and transported to the GPU. There the CO_2 concentration is increased from 81.2 vol.-% (dry) to at least 95 vol-% (dry).

Table 1: Ultimate analysis of the South African coal and the process parameters of the basic power plant used in this study.

Ultimate analysis		Basic power plant	
LHV	25100 kJ/kg	Power (gross)	600 MW
H_2O	0.078 kg/kg	Power (net)	555.5 MW
Ash	0.135 kg/kg	Steam parameters	600°C / 620°C / 285 bar / 60 bar
C	0.661 kg/kg	Condenser pressure	45 mbar
H	0.0383 kg/kg	Ambient temperature	15 °C
N	0.016 kg/kg	Efficiency (gross)	49.44 % pts.
O	0.066 kg/kg	Efficiency (net)	45.77 % pts.
S	0.0057 kg/kg	Spec. CO_2-emissions	750.7 g_{CO2}/kWh

2.2 GPU modelling

Two GPU processes are modelled, because the specifications concerning the purity of the injected CO_2 stream are still uncertain. For a required CO_2-purity of >95 vol.-% (dry) a partial condensation process is sufficient. For higher purities a distillation column has to be applied instead.

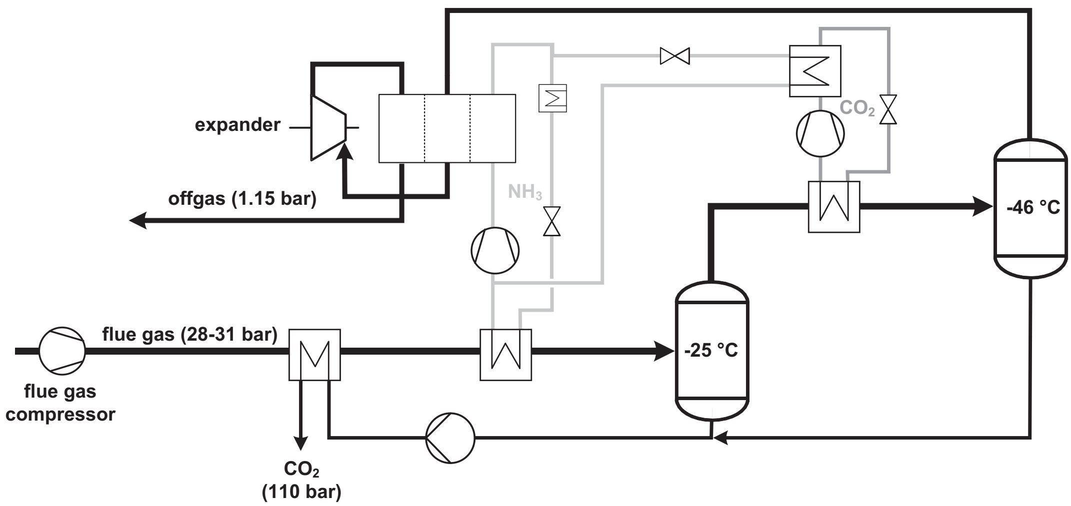

Figure 2: Process flow sheet model of the GPU with externally cooled partial condensation.

In Fig.2 the process flow sheet of the GPU model with partial condensation is shown. The CO_2 capture rate is kept constant at 90 %. The flue gas is compressed from 1 bar to 28-31 bar, depending on CO_2 concentration. The compression is implemented with 6 stages and interstage cooling after each compressor stage. The remaining water is absorbed by a molecular sieve. Downstream of the compression the dried flue gas is cooled down to a temperature of -25°C. Already liquefied CO_2 is separated then in a first flash vessel. The remaining flue gas is cooled down to -45°C and flashed again. The liquefied CO_2 has a purity of 97 mol % and is pumped at a pressure of 110 bar for transportation and storage.

The remaining offgas contains most of the flue gas impurities. In this study only CO_2, N_2, O_2 and Ar are considered, because residual SO_x and NO_x react with water during the compression to acid [3], [4]. The offgas is used to cool the NH_3 of the cooling cycle to the saturated vapour line. The offgas is expanded down to 1.15 bar. The actual condensation of the NH_3 is done with cooling water as coolant, because the heat capacity of the offgas is too small. In addition to the flue gas cooling, the NH_3 is used to condensate the CO_2 of the second cooling cycle. The expanded offgas is bled to the atmosphere.

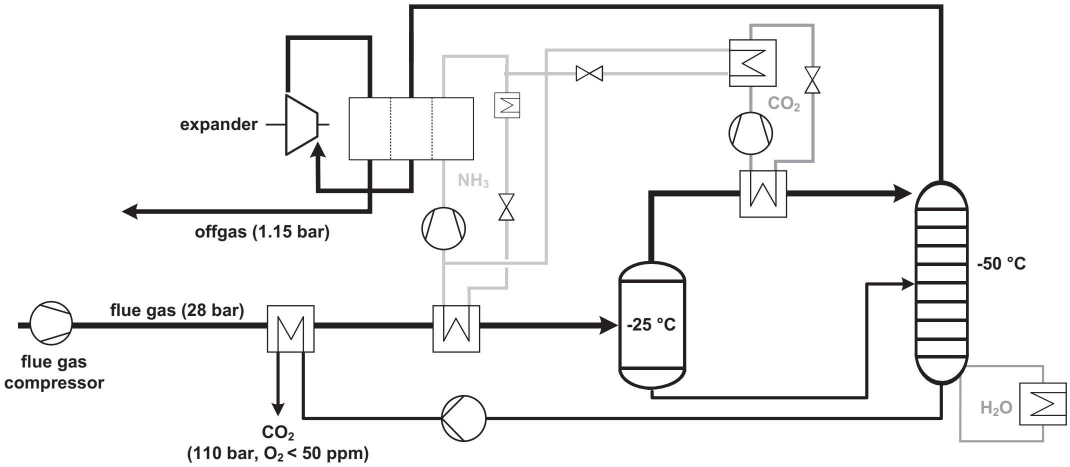

Figure 3: Process flow sheet model of the GPU with externally cooled distillation.

To achieve higher CO_2 purities the second flash in Fig.2 is replaced by a distillation column. This column has a reboiler and no externally cooled condenser. The flue gas is cooled to -50°C to keep the overall CO_2 capture rate at 90 %. The column has 15 stages and a reboiler at the bottom. The flue gas enters the column at the first stage. The gaseous part of the fluid leaves the column with the offgas. The liquid part of the fluid trickles down the column while heated up. The CO_2 condensate from the flash enters the column at stage 9 where the CO_2 concentration is similar to the concentration of the entering stream. At the bottom a maximum temperature of 7.7°C is reached. The purified liquid CO_2 is handled in the same way as in the partial condensation.

The distillation has a higher energy demand than the partial condensation. The reboiler duty leads to a higher cooling water demand. The liquefied CO_2 has a higher temperature what leads to a higher NH_3 coolant demand. Additionally the flue gas is cooled down to -50°C instead of -46°C. In Table 3 the calculated parameters for the two basic overall processes are listed. The higher CO_2 purity leads to a by 0.18 % pts. lower overall process efficiency. The results show that the distillation process can achieve O_2 concentrations below 50 ppm at high capture rates.

With the CO_2 capture rate of 90 % the specific power plants emissions are decreased down to 96 g_{CO2}/kWh. With the basic GPU processes studied here a further increase in the overall CO_2 capture rate would only be possible at the expense of additional high energy demand.

2.3 GPU with additional gas treatment

The offgas has a high pressure and low temperature. It contains 10 % of the total CO_2 and about 4-5 % of the O_2 produced in the ASU. By further treatment of the offgas it is possible to increase the CO_2 capture rate and to lower the energy demand of the ASU. In the literature [2], [5], [6] two major ways are discussed to realise this gas treatment: polymeric membranes and pressure swing adsorption. In this study the main aim of the gas treatment is to increase the overall CO_2 capture rate to 99 % with a CO_2 capture rate of the additional gas treatment of 90 % and a CO_2 capture rate of the basic GPU of 90 %. In addition to the overall capture rate the overall efficiency of the Oxyfuel process is maximised. Due to the last aim only a one stage process membrane or PSA process is considered, to spare an additional compressor or vacuum pump.

2.3.1 Polymeric membranes (PM)

PMs are suitable to separate CO_2 from the offgas stream because of their high CO_2 selectivity and permeability. The high selectivity is achieved by the ability to use the carrier transport mechanism [7] for CO_2. Membranes which use only diffusion transport have much lower selectivities and permeabilities concerning CO_2.

Table 2: Process parameters for the two base Oxyfuel processes studied here. The offgas parameters are calculated downstream of the second flash/distillation column. The CO_2 parameters are calculated downstram of the CO_2 pump.

Process parameters	Partial Condensation	Distillation
CO_2 purity in vol.-% (dry)	97	99.99
Capture rate in %	90	90
Cooling water demand in kg/s	2319	2489
Specific energy demand GPU in kWh/t_{CO2}	131	137.9
Overall process efficiency in % pts.	36.32	36.14
Offgas downstream separation in vol.-% (dry):		
CO_2	0.332	0.304
N_2	0.368	0.378
O_2	0.162	0.173
Ar	0.139	0.145
p in bar	30	29
T in °C	-46	-49.5
$\dot{m}$ in kg/s	27.7	29.9
CO_2 downstream of pump in vol.-% (dry):		
CO_2	0.974	99.99
N_2	0.015	16 ppm
O_2	0.009	50 ppm
Ar	0.006	16 ppm
p in bar	110	110
T in °C	-46	-0.7
$\dot{m}$ in kg/s	103.9	101.77

In this study the membrane is modelled on the basis of its separation characteristics [7]. With this modelling the concentration polarisation on the feed side and the pressure drop in the porous supporting layer are neglected. It is also assumed that the separation is independent of the flow direction of the permeate. With given selectivity, permeability, feed pressure and permeate pressure the separation process can be calculated. The characteristics of the membrane type chosen are shown in Table 3. PEO has a good relation between permeability and selectivity. The selectivity for CO_2/Ar is not given in the literature. It is assumed here to amount 0.476 of the selectivity for CO_2/N_2, given that [8] published data for Ar/N_2 with 2.1.

Table 3: Separation characteristics of the PEO Membrane.

Membrane type	PEO
CO_2/N_2 selectivity	45 [10]
CO_2/O_2 selectivity	15 [10]
CO_2/Ar selectivity	21.4
Permeability in Nm^3/m^2hbar	1.25 [10]

2.3.1 Pressure Swing Adsorption (PSA)

Another suitable technology for the separation of the CO_2 from the offgas is PSA. In this work zeolite 13X is used as adsorbent in a 6 bed PSA process [9]. It is chosen because activated carbon shows hysteresis when applied to high pressure processes [11]. The data of the chosen process are given in Table 4. To minimise the loss of CO_2 no purging step is applied. To calculate the adsorption performance of the zeolite, the Langmuir isotherms of CO_2 [11], N_2 [11], O_2 [12] and Ar [12] are taken from the literature and implemented into the model. Interaction between the different components is neglected. This estimation is suitable for CO_2 separation processes [11]. With the isotherms, the feed pressure and the evacuation pressure the gas composition of the recovered CO_2 as well as the exhaust gas can be calculated. In addition to that the amount of adsorbent needed for the process can be determined.

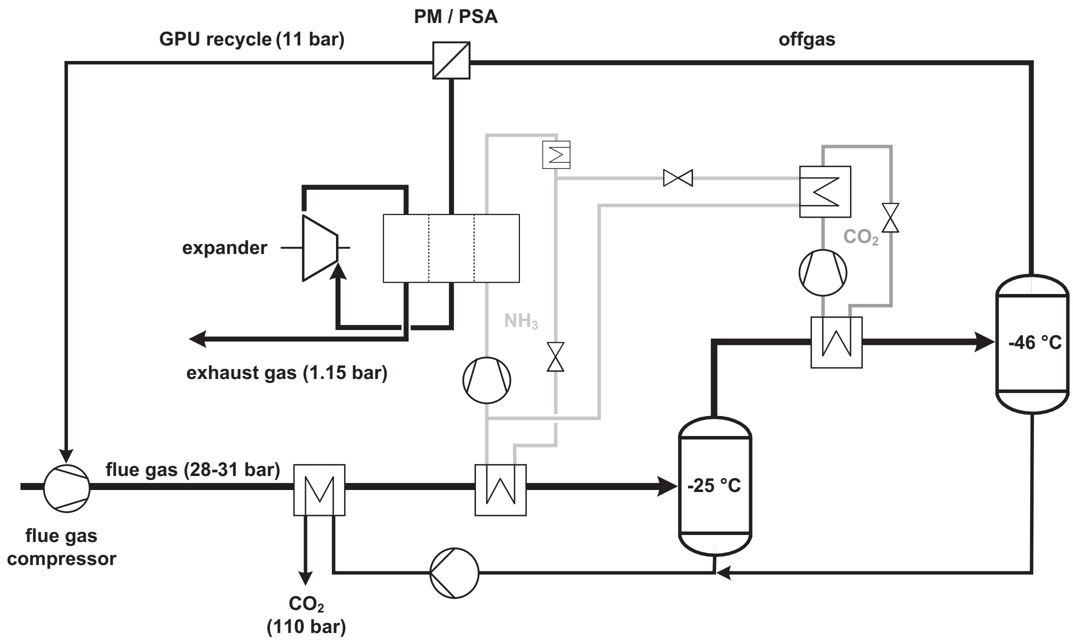

Figure 4: Process flow sheet model of a GPU with additional gas treatment.

Table 4: Process parameters for the PSA process.

Adsorbent	13X
Number of beds	6
Cycle time per bed	120 s
Bed density	0.659 g/cm³ [12]
Length of mass transfer zone length / Bed length	0.2 [9]

2.3.3 Process integration

The constraints for the integration into the process are the same for both technologies. For both processes a driving pressure difference is needed. The offgas is already at high pressure, so the offgas can be fed into the gas treatment without additional compression (see Fig.4). Two streams are leaving the gas treatment, the GPU recycle and the exhaust gas (see Fig. 4). The GPU recycle can be refed to the flue gas compressor depending on the pressure level. The recycle is necessary, because the CO_2 concentration of the GPU recycle is below 90 vol. -% (dry) and would decrease the purity of the product CO_2 stream of the GPU. The pressure ratio over the gas treatment is kept at ~3-4. This constraint is taken to achieve a high CO_2 concentration in the GPU recycle as well as a small membrane area (PM) or bed volume (PSA) and a small pressure difference. There are different possibilities to realise the GPU recycle. In Fig. 4 the offgas enters the additional gas treatment with a pressure of 30.75 bar (PM) and 42.8 bar (PSA) and due to that the GPU recycle is still on a high pressure level and can be mixed with compressed flue gas before stage four of the compressor. In this process design, the pressure ratio between offgas and GPU recycle can vary between 3 and 4 because the feed pressure depends on the CO_2 content of the flue gas compressed in the GPU. An increase in the CO_2 content leads to a lower pressure level requirement for achieving a capture rate of 90 % in the basic GPU process. The GPU recycle is kept constantly at 11 bar. Due to that the GPU recycle can be mixed with compressed flue gas before stage four, a pressure ratio of ~3-4 can be maintained for the recompression.

Another possibility is to expand the offgas to a pressure level of 3.3 bar, before entering the additional gas treatment. With the pressure ratio of 3 the pressure of the GPU recycle arises to 1.1 bar. Therefore the GPU recycle has to be recycled upstream of the flue gas compression. The pressure ratio can be kept at 3 because of the better separation performances of the two gas treatments. The amount of cooling water needed to condense the NH_3 of the cooling cycle in the GPU is minimised, but the GPU recycle has to be compressed further using a pressure ratio of 30.

3. Results

Several characteristic parameters are calculated: the specific energy demand of the GPU, the specific membrane area, the specific bed volume and the overall process efficiency. The first parameter shows the energy demand of the GPU process in relation to the amount of CO_2 mass flow in the CO_2 product stream. The specific membrane area and specific bed volume show the size of the additional gas treatment process needed to achieve the 99% overall capture rate. The overall process efficiency shows the effects of the additional gas treatment on the power plant's overall process. To estimate the potential of an oxygen refeed to the ASU, the overall process efficiency is calculated.

3.1 Partial Condensation

The characteristic values of the GPU without additional gas treatment are shown in Table 2. The specific energy demand amounts to 131 kWh/t_{CO2}. In Table 5 the specific energy demands of partial condensation with additional gas treatment are shown.

Table 5: Specific energy demand of the partial condensation GPU processes with additional gas treatment in dependence of the feed pressure to the gas treatment.

Feed pressure in bar	Specific energy demand (PM) in kWh/t_{CO2}	Specific energy demand (PSA) in kWh/t_{CO2}
3.3	134	134
30.75	133	-
42.8	-	149

For the lower feed pressure there are no differences between PSA and PM. Both are higher than without additional gas treatment. This is expected, as the GPU recycle has to be compressed again, which leads to a higher energy demand. The higher pressure of the GPU recycle is different between PSA and PM, as explained in 2.3.3. For the partial condensation with PM the pressure increase leads to a decrease in the specific energy demand, whereas the specific energy demand of the partial condensation with PSA increases significantly. This is due to the Langmuir isotherms which approach each other on high pressure levels [11]. The reason for the higher feed pressure compared to the partial condensation with PM is the lower CO_2 concentration of the GPU recycle (see Table 6). The PM achieves a higher purity for the pressure ratio given. This leads to a lower CO_2 concentration in the flue gas stream treated in the GPU. So the target CO_2 capture rate of 90 % in the basic GPU can be reached with a pressure of 30.75 bar for the partial condensation with PM and a pressure of 42.8 bar for the partial condensation with PSA.

The size of the membrane area and the amount of bed volume are functions of the feed and product pressure, the composition of the feed gas and the CO_2 capture rate of the additional gas treatment. As the CO_2 capture rate in the additional gas treatment is kept constantly at 90 %, the additional gas treatment depends on the other parameters. In Table 6 the specific membrane area and the specific bed volume can be seen. As expected, the specific membrane area decreases significantly when the feed pressure is higher. The specific bed volume increases compared to the lower pressure level. This can be explained with the higher amount of impurities in the GPU recycle. The impurities demand a higher amount of bed volume and result to a higher mass flow of the GPU recycle.

The flue gas amount treated in the GPU increases due to the GPU recycle. This leads to a higher energy demand in the GPU process. In the first graph of Fig. 5 the influence of the additional gas treatment on the overall process efficiency with a partial condensation GPU is shown. The efficiency loss lies between 0.51 and 1.02 % pts. The results of the specific energy demand suggest that the high pressure GPU recycle of the partial condensation with PM shows a better performance than the low pressure GPU recycle of the partial condensation with PM.

Table 6: CO_2 concentration in the GPU recycle to the partial condensation GPU processes with additional gas treatment and calculated membrane area and bed volume in dependence of the feed pressure to the gas treatment.

Feed pressure in bar	CO_2 content (PM) in the GPU recycle in vol.-% (dry)	CO_2 content (PSA) in the GPU recycle in vol.-% (dry)	Specific membrane area in m²/kg_{CO2}	Specific bed volume in m³/kg_{CO2}
3.3	79.4	79.6	8,825	145.5
30.75	75.66	-	1,165	-
42.8	-	42.1	-	836.1

Though the increase of the overall process efficiency for the partial condensation with PM by an increase of the GPU recycle pressure is small compared with the low pressure scenario, the investment costs can be lowered significantly due to the smaller size of the PM. The PSA processes show that the adsorbent should be used for low pressure processes. An increase of the overall capture rate from 90 % to 99 % leads to an overall efficiency decrease of at least 0.51 % pts.

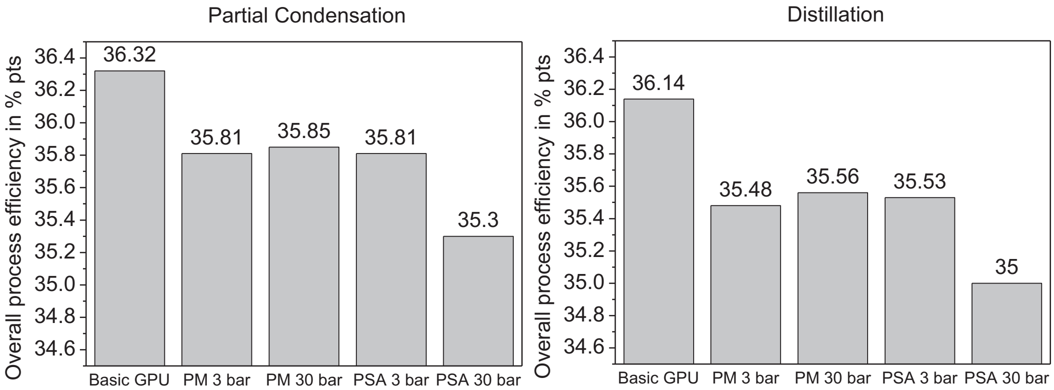

Figure 5: Overall process efficiency for the partial condensation and the distillation GPU processes. The capture rate for the basic GPU process is 90 %. The capture rate for the processes with additional gas treatment are maintained at 99 %.

3.2 Distillation

The results for the basic GPU with a distillation column are shown in Table 2. The specific energy demand of the GPU without additional gas treatment is 138 kWh/t_{CO2}. For the GPU with additional gas treatment the results are shown in Table 7.

Table 7: Specific energy demand of the distillation GPU processes with additional gas treatment in dependence of the feed pressure to the gas treatment.

Feed pressure in bar	Specific energy demand (PM) in kWh/t_{CO2}	Specific energy demand (PSA) in kWh/t_{CO2}
3.3	143	142
28	141	159

Like in the partial condensation GPU case with additional gas treatment the specific energy demand is higher for a low pressure GPU recycle than for a basic distillation GPU. This is due to the same reason as described for the partial condensation with additional gas treatment. The slightly higher specific energy demand for the distillation with PM can be explained with the lower CO_2 concentration in the GPU recycle (see Table 10). For low pressure the distillation with PM results shows a lower CO_2 concentration of the GPU recycle than the distillation with PSA. The distillation concentrates nearly all impurities within the offgas and decreases the partial pressure of the CO_2 of the offgas fed to the PSA/PM. So the distillation with PSA shows a better separation performance with higher amounts of impurities in the offgas when the pressure level of the GPU recycle is low.

For the distillation GPU the purity and the CO_2 capture rate of the basic GPU are set. The column pressure is optimised at 28 bar which is the optimum for the designed capture process under consideration of the temperature, the desired purity and the CO_2 capture rate of the basic GPU. The results for the high pressure GPU recycles show a difference between the CO_2 concentrations of the GPU recycles (see Table 8) comparable to the partial condensation processes. The separation performance of the distillation with PSA decreases significantly with higher feed pressure while the performance of the distillation with PM decreases moderately. This explains the higher specific energy demand for a gas treatment with PSA compared to a gas treatment with PM (see Table 7). The low CO_2 concentration in the GPU recycle of the distillation with PSA leads to a lower flue gas CO_2 concentration and therefore a worse GPU performance. The results show similarity with the results of the partial condensation with PSA.

The specific membrane area and the specific bed volume calculated for the distillation GPU are summarised in Table 8. The specific membrane area needed to separate the CO_2 is higher for both recycle pressure levels than for the partial condensation GPU. This is due to the lower CO_2 concentrations of the GPU recycle compared to the partial condensation processes. The higher amount of impurities in the GPU recycle leads to a higher mass flow of the GPU recycle if the CO_2 capture rate is kept at 90 % in the additional gas treatment. This makes a larger membrane area necessary.

Table 8: CO_2 concentration in the GPU recycle to the distillation GPU processes with additional gas treatment and calculated membrane area and bed volume in dependence of the feed pressure to the gas treatment.

Feed pressure in bar	CO_2 concentration (PM) in the GPU recycle in vol.-% (dry)	CO_2 concentration (PSA) in the GPU recycle in vol.-% (dry)	Specific membrane area in m²/kg_{CO2}	Specific bed volume in m^3/kg_{CO2}
3.3	74.2	79.6	11,406	145.5
28	66.4	43.5	2,114	896

The specific bed volume of the distillation with PSA process shows the same behaviour like the partial condensation with PSA comparing volume and CO_2 concentration of the GPU recycle. For low pressures the CO_2 concentrations of the GPU recycle are similar to the partial condensation results and this leads to similar specific bed volumes needed. For the high pressure level the separation performance decreases significantly. So for high pressure gas treatment the PM again shows better performance while for low pressure gas treatment the PSA shows advantages.

The overall process efficiency for the distillation GPU processes with additional gas treatment is shown in the right graph of Fig.5. Again, there is a significant decrease compared to the basic GPU process. The decrease lies in between 0.58 and 1.14 % pts. Compared to partial condensation the decrease is slightly bigger. This is due to the described performance problems with the PM/PSA at higher impurity amounts in the feed gas of the gas treatment. The values show a significant difference between the high pressure and low pressure recycle, especially for the distillation with PSA.

This suggests that it is preferable to realise the additional gas treatment with PM at a higher pressure level to minimise the overall process efficiency loss and to decrease investment costs, because of the smaller amount of specific membrane area needed for the separation. A PSA gas treatment with a high pressure GPU recycle shows significant disadvantages and is not considered further. The ultimate economic viability of the additional gas treatment, however, depends mainly on the price of the CO_2-emission-certificates.

3.3 Assessment of an exhaust gas recycle to the steam generator for efficiency increase

The exhaust gas contains oxygen due to the air ingress into the overall process and the excess of oxygen fed to the steam generator. For an assumed air ingress of 2 % of the flue gas mass flow upstream of the recycle and a local oxygen ratio in the combustion chamber of 1.15 the amount of oxygen in the exhaust gas arises to 4.3 % of the oxygen supplied by the ASU. This oxygen shall be fed to the combustion chamber to lower the oxygen demand of the process and increase the overall process efficiency. In this study only a calculation to estimate the maximum efficiency increase of such a recycle is made. The process is the same as described in Fig. 4.

In Fig. 6 the potential for the exhaust gas recycle is shown. It amounts to 0.25 to 0.29% pts. for the partial condensation and 0.3% pts. for the distillation. This is the highest possible efficiency increase without taking further gas treatment into account beyond that, needed to separate the oxygen from the offgas. The higher efficiency increase for the distillation can be explained by the higher total amount of oxygen that is in the offgas due to the high purity of the CO_2 product stream leaving the distillation column.

4. Conclusions

The capture of CO_2 from the offgas with PSA or PM decreases the process efficiency. Due to the low CO_2 concentration achieved by the additional gas treatment, the GPU recycle has to be recycled to flue gas compression at a high pressure level. To minimise the energy demand additional compressors or vacuum pumps should be avoided. For a low pressure level at the additional gas treatment PM and PSA show similar results with a slightly higher CO_2 content for the PSA. At a high pressure level only a gas treatment with PM should be realised due to the low separation performance of the used adsorbent. Furthermore, a high pressure for the GPU recycle of the GPU with PM lowers the space requirement for the additional gas treatment significantly. A possible refeed of the residual oxygen in the exhaust gas to the combustion chamber can increase the overall process efficiency. Still the net efficiency would be lower than in the base process.

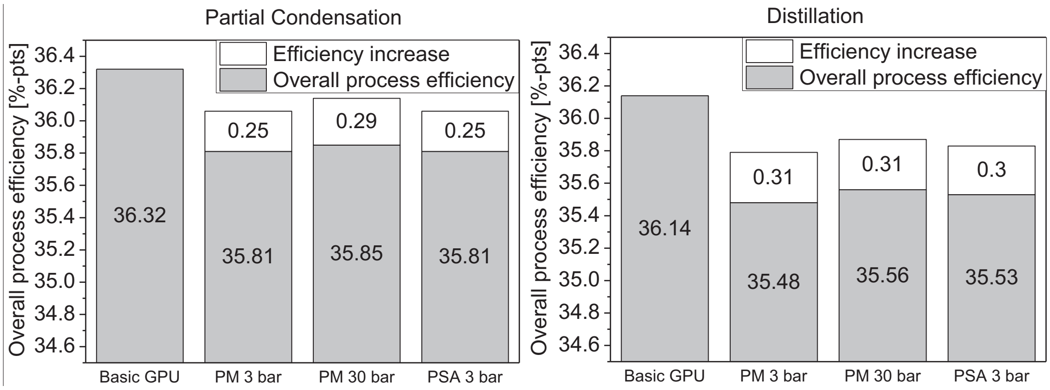

Figure 6: Maximum efficiency increase due to a recycle of the residual oxygen in the offgas to the combustion chamber. No process for separation considered. The capture rate for the basic GPU process is 90 %. The capture rate for the processes with additional gas treatment is kept at 99 %.

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