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The coal-fired Oxyfuel-process with additional gas treatment of the ventgas for increased capture rates

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Abstract

The Oxyfuel-process could be a solution to lower the high specific CO_2 emissions of coal-fired power plants. The carbon capture rate (CCR) in current research is usually set to 90% due to the increasing specific energy demand of the capture process (GPU) and lower CO_2 purity of the product stream at higher CCR. The remaining CO_2 and most of the impurities escape to the environment with the ventgas downstream of the GPU. In current literature it is recommended to increase the CCR from 90% to higher values of up to 98-99%. This can be achieved by adding an additional gas treatment to capture the CO_2 contained in the ventgas downstream of the GPU. At high CCR like 99% the necessary recycle of the captured CO_2 into the basic GPU process leads to an increase of the specific energy demand of the GPU and an efficiency decrease of the overall process. It is possible to lower this efficiency penalty by recycling the remaining impurities downstream of the additional gas treatment to the ASU to regain the contained oxygen. The feasibility of the recycle strongly depends on the capture rate of the additional gas treatment, because it influences the CO_2 concentration in the recycled exhaust gas. In this work an overall process of a coal-fired Oxyfuel power plant with cryogenic ASU and externally cooled GPU as a basic process is modelled. This process is adapted to higher CCR by adding an additional gas treatment by a polymeric membrane (PM) downstream of the GPU. The ASU is modelled as a triple column process. This process enables an exhaust gas recycle downstream of the PM. The GPU is a two stage partial condensation. Furthermore the influence of the additional gas treatment on the GPU process and the overall process for different CCR is examined and the resulting exhaust gas concentrations are calculated to evaluate the possibility for an exhaust gas recycle. This evaluation is necessary, because the remaining CO_2 in the exhaust gas that is recycled to the ASU has to be removed upstream of the ASU. This leads to an additional energy demand to regenerate the molecular sieves upstream of the ASU.

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Keywords: Oxyfuel; Carbon Capture; overall process simulation; Optimisation

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1. Introduction

To alleviate the climate change it is necessary to reduce the anthropogenic CO_2 emissions. Coal-fired power plants have high specific CO_2 emissions compared to other power plant processes. To reduce these emissions carbon capture and storage (CCS) is a possibility. The Oxyfuel-process is a promising process to capture the CO_2 from the flue gas of a coal-fired power plant.

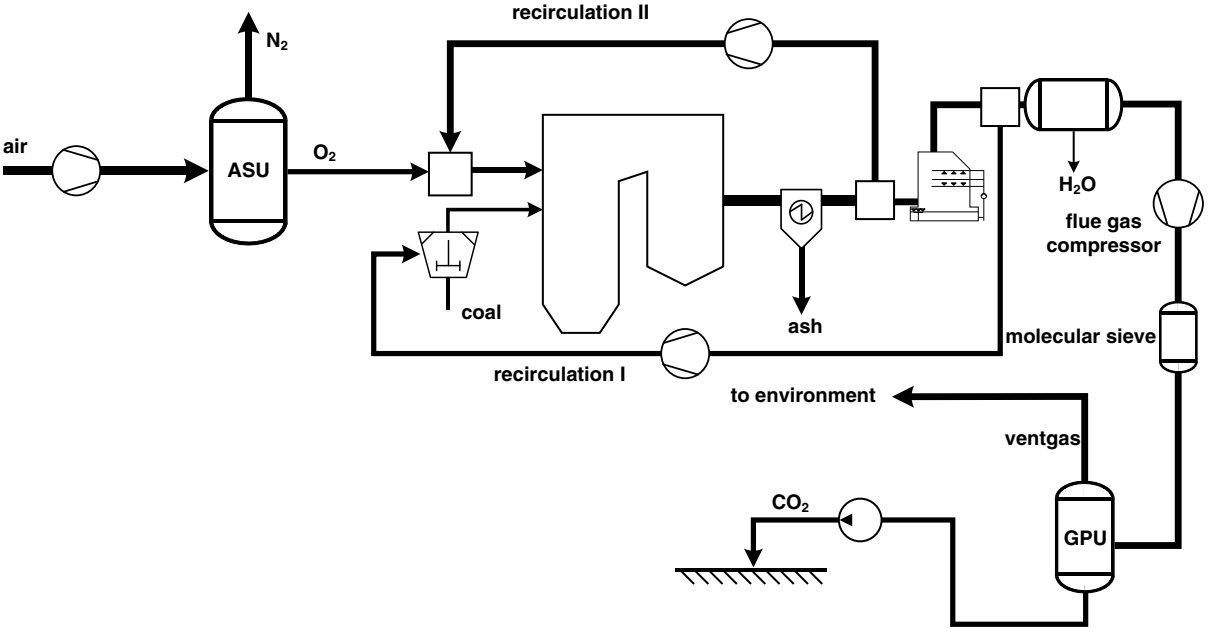

Figure 1: Basic coal-fired Oxyfuel power plant with cryogenic air separation unit (ASU) and a gas processing unit with partial condensation (GPU) to capture the CO_2 from the flue gas.

In Figure 1 the basic Oxyfuel-process with cryogenic ASU is shown. The main target of the process is to achieve a high CO_2 concentration in the flue gas to enable a low energy demand for the GPU process downstream. This is achieved by separating the nitrogen from the air upstream of the boiler in the ASU. The coal is combusted with a mixture of oxygen and recycled flue gas. The flue gas recycle is necessary to keep the temperatures inside the boiler within technical limits of used boiler material [1] [2]. The generated flue gas has a CO_2 concentration of about 81 vol.-% (dry), if an air ingress into the process of 2% is considered. The remaining 19 vol.-% (dry) are impurities (NO_x, SO_x, O_2, N_2 and Ar). Downstream of the boiler fly ash is reduced in an electrostatic precipitator (ESP) and downstream of the recycle SO_x is reduced in a flue gas desulphurisation (FGD) before being dried in the flue gas condenser. The flue gas is then compressed and finally dried. The compressed flue gas is cooled down and partially liquefied. Thereby the CO_2 is enriched in the liquid phase and separated from the flue gas. A CCR of about 90 % can be achieved while maintaining a CO_2 concentration above 96 vol.-% (dry) [3] [4]. 10 % of the CO_2 remains in the ventgas. In the basic Oxyfuel-process the ventgas leaves the process to the environment. In prior research it was shown, that it is possible to achieve highest CCR up to 99 % by installing a PM process for the ventgas [5] [6]. In this work different capture rates of the PM process are considered by varying the membrane area and the pressure ratio Π_{mem} ($\Pi_{mem} = p_{Feed}/p_{permeate}$) of the PM.

The remaining exhaust gas downstream of the PM process contains oxygen which can be recycled to the ASU. This is only feasible if a triple column ASU is used for oxygen supply [7]. For the possible benefit it was taken into account that the ventgas expanders would regain less energy and the oxygen capture rate of the recycled exhaust gas in the ASU would have influence on the ASU process. In this work due to the varying capture rate of the PM process larger amounts of CO_2 can be contained in the exhaust gas recycle. The CO_2 has to be removed in the adsorption bed

of the ASU causing a higher energy demand from the power plant for bed regeneration. An overall process evaluation is necessary to quantify the losses due to the higher energy demand and the effects of the additional gas treatment with exhaust gas recycle on the overall process. The modelling of the overall process is done with *EbsilonProfessional*® whereas modelling of the ASU and the GPU is done with *AspenPlus*®. The PM process is modelled with *AspenCustomModeler*®.

2. Modelling assumptions

The water steam cycle of the power plant is based on the study "Reference power plant Northrhine-Westphalia" [8]. It is a state-of-the-art power plant with bituminous coal as fuel. The main data of the power plant is given in Table 1. The used coal is a bituminous coal from South Africa.

Table 1: Ultimate analysis of the South African bituminous coal and the main data of the air-blown reference power plant.

Ultimate analysis of the coal		Reference 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	Economiser exit temperature (flue gas)	380°C
N	0.016 kg/kg	Efficiency (gross) LHV	49.4 %
O	0.066 kg/kg	Efficiency (net) LHV	45.8 %
S	0.0057 kg/kg	Spec. CO_2-emissions	750.7 g_{CO2}/kWh

2.1. Basic Oxyfuel-process

The Oxyfuel-process is modelled with a cryogenic ASU for oxygen supply and a partial condensation for CO_2 capture to enable a realisation of the process in the near future. The ASU is modelled as a triple column ASU [9] process to enable a possible exhaust gas recycle for oxygen recovery when the additional gas treatment is applied. The compression of the air is adiabatic and the compression heat is used to preheat the oxygen and parts of the low pressure feed water. In Table 2 the main boundary conditions and calculated values for the basic process are listed. The oxygen temperature is 156°C after preheat. This process design leads to a specific energy demand of 178 kWh/t_{O2} at the gear shaft. With an electrical engine efficiency of 97 % the specific energy demand results to 184 kWh_{el}/t_{O2}. With 184 kWh_{el}/t_{O2} the triple column with adiabatic compression has a 15 % lower energy demand than the dual column modelled in [6]. The complicated compression train is a disadvantage. Because of this the heat recovery is not as high as for the dual column and the efficiency only rises by 0.7 %-pts. compared to the Oxyfuel-process with a dual column ASU. The primary recirculation is desulphurised and ash-free. The secondary recirculation is ash-free (see Figure 1). The ESP is modelled for a temperature of 380 °C to enable a hot secondary recirculation. The FGD is modelled as a wet limestone absorption. The flue gas is dried downstream of the primary recirculation. Afterwards the flue gas is compressed in a 6-stage compressor. With the chosen process configuration the flue gas has a concentration of 81 vol.-% (dry). Due to this a GPU is necessary to achieve a higher CO_2 purity. For a CCR of 90 % a pressure level of 30 bar is necessary. The process is externally cooled with NH_3 and CO_2 as refrigerants (see Figure 4). The CO_2 purity has to be maintained above 96 vol.-%. The liquefaction temperatures are set to -25 °C for the first separator and -46 °C for the second separator. This process design leads to a specific

energy demand of 128 kWh/t_{CO2} at the gear shaft. With an electrical engine efficiency of 97 % the specific energy demand results to 132 kWh_{el}/t_{CO2}. The dependence of the CO_2 purity and the CCR on the pressure level downstream of the flue gas compressor is shown in Figure 2 on the right. The CO_2 purity decreases and the CCR increases with higher pressure levels. At pressure levels higher than 37 bar the CO_2 purity drops below 96 vol.-%. So with this process configuration and flue gas quality the highest CCR that can be achieved is 92 %. At higher CCR the CO_2 purity is not sufficient for storage purposes. The specific energy demand has a minimum in the considered pressure range at 34 bar. The CO_2 concentration in the ventgas is also influenced by the pressure level of the flue gas compression (see Figure 2 left). At high capture rates and pressure levels the concentration of CO_2 in the ventgas drops due to the high amount of CO_2 captured in the product stream. For the chosen basic process the overall process leads to a net efficiency of 37.5 % and specific CO_2 emissions of 93 g/kWh_{el}.

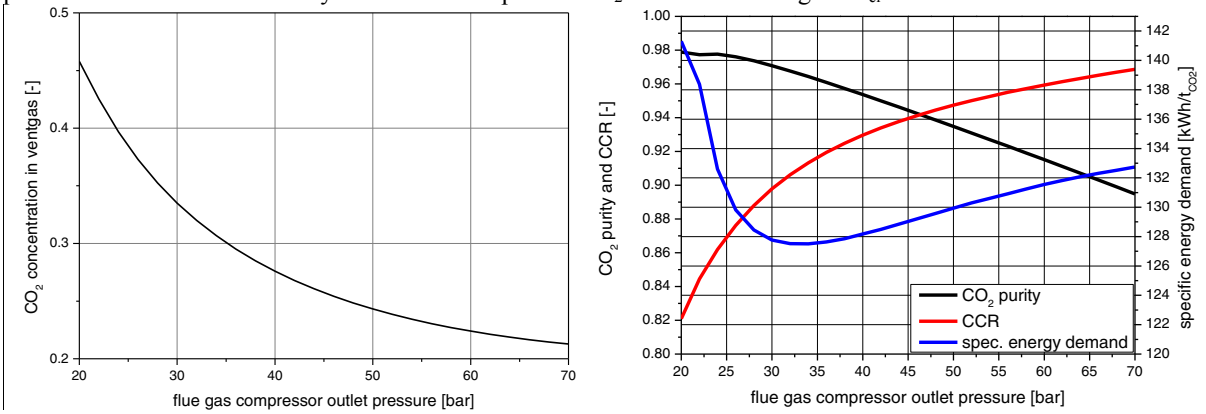

Figure 2: Left: CO_2 concentration in the ventgas depending on the outlet pressure of the flue gas compressor for the constraints given in Table 2. Right: Process data of the basic GPU process depending on the outlet pressure of the flue gas compressor.

Table 2: Boundary conditions and calculated values for the basic Oxyfuel-process.

ASU (triple column with adiabatic compression)		Overall process		GPU (partial condensation)	
O_2 purity	95 vol.-%	Power (gross)	600 MW	CCR	90 %
O_2 capture rate	97.3 %	Power (net)	446.5 MW	CO_2 purity	> 96 vol.-%
O_2 preheat temperature	156 °C	Furnace exit temperature	1250°C	Pressure after compression	30 bar
Feed water preheat temperature	104 °C	Evaporator exit temperature	470°C	Liquefaction temperatures	-25°C/-46°C
Compressor efficiency	85 %	O_2 excess (local)	15 %	Specific energy demand	132 kWh_{el}/t_{CO2}
Steam demand for adsorber regeneration	6 kg/s	Air ingress	2 %	Engine efficiency	97 %
Specific energy demand	184 kWh_{el}/t_{O2}	Efficiency (net)	37.5 %		
Engine efficiency	97 %	Spec. CO_2-emissions	93 g_{CO2}/kWh_{el}		

2.2. Oxyfuel-process with increased CCR

To achieve higher CCR than 92 % while maintaining the CO_2 purity at high levels a PM can be applied to the ventgas [5] [6]. The PM model was designed with *AspenCustomModeler*® as shown in [10]. The modelled

Polyactive® membrane is commercially available [11]. The main data is shown in Table 3. It has a high selectivity for CO_2 over the inert gases and a high CO_2 permeance.

Table 3: Data of the *Polyactive*® membrane modelled in this work [11].

PM data	Value at T=25°C
CO_2 permeance	3 m³(STP)/m²hbar
CO_2/N_2 selectivity	50
O_2/N_2 selectivity	2.8
Ar/N_2 selectivity	2.8

The amount of CO_2 in the ventgas has an effect on the PM process. For the basic process the ventgas has a CO_2 concentration of 34 vol.-% (see Figure 2 left). The behaviour of the PM for different CO_2 concentrations in the ventgas can be seen in Figure 3 left. A larger membrane area leads to a higher capture rate of the PM. The same can be seen for a higher CO_2 concentration in the ventgas.

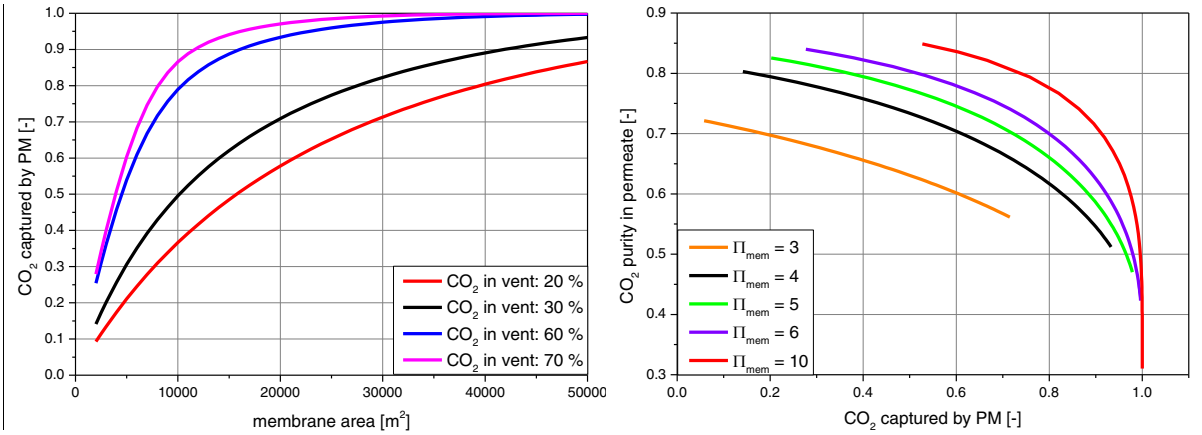

Figure 3: Left: CO_2 captured by the PM for different CO_2 concentrations in the ventgas at a Π_{mem} of 4. Right: CO_2 purity in the permeate depending on the CO_2 capture rate by the PM for different Π_{mem} and a CO_2 concentration in the ventgas of 30 %.

The values are for a constant Π_{mem} of 4. The effect of the applied Π_{mem} on the capture rate of the PM and the CO_2 purity in the permeate is shown in Figure 3 right. A higher capture rate leads to a decreasing CO_2 concentration in the permeate while a higher Π_{mem} leads to a higher permeate purity and capture rate of the PM. The CO_2 concentration in the ventgas is 30 %. In Figure 4 the dual partial condensation process is shown. The PM process can be added as shown with the dotted line. The feed temperature to the PM has to be maintained at 25°C, to ensure that the PM can achieve the performance assumed in Table 3. The captured CO_2 in the permeate is recycled to the flue gas compression and mixed with the flue gas. The remaining exhaust gas is used for cooling demands and decompression in the GPU. For a more detailed description of the GPU see [5] [6]. Π_{mem} as well as the size of the PM are varied.

The additional capture process leads to an efficiency loss [5] depending on the amount of CO_2 captured by the PM process. In [6] it was proposed that the exhaust gas (see Figure 4) can be recycled to the ASU to recover the oxygen contained in it. For a triple column this can be beneficial to the energy demand of the ASU and lower the efficiency loss caused by the additional capture process [7]. The energy recovery potential can be up to 4 % of the

total energy demand of the ASU depending on the O_2 purity supplied by the ASU, the O_2 excess in the combustion chamber, the air ingress and the GPU process.

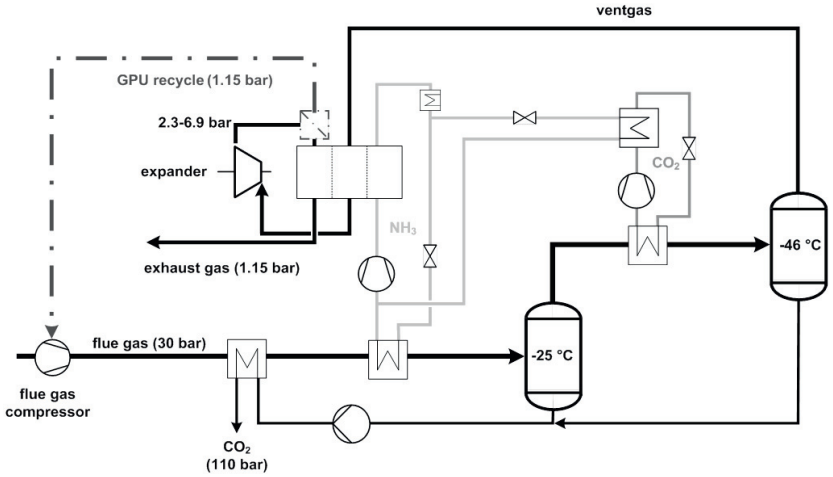

Figure 4: Dual stage partial condensation with external cooling by NH_3 and CO_2 and a PM in the ventgas system for higher CCR.

3. Results

3.1. Effects of the PM process on the GPU

The influence of the PM process on the GPU is evaluated concerning different parameters. The purity of the captured CO_2 product stream can be seen in Figure 5 depending on the outlet pressure of the flue gas compression for a GPU with and without a PM applied. The purity of the captured CO_2 is only slightly lower if a PM is installed.

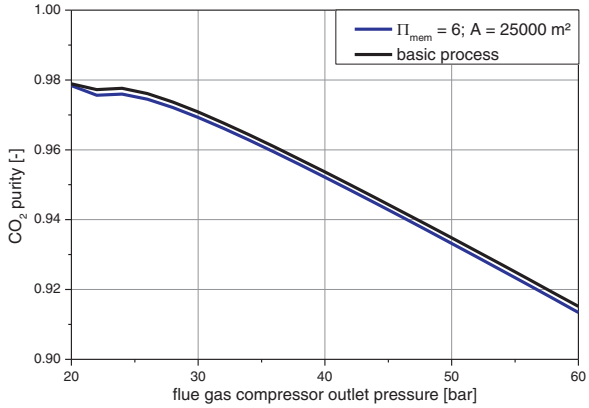

Figure 5: CO_2 purity of the CO_2 captured in the basic GPU process and of the CO_2 captured in the GPU with PM for a membrane size of 25000 m² and a Π_{mem} of 6. The values are shown over the outlet pressure of the flue gas compressor.

The difference is still very small even for a high Π_{mem} of 6 and a large membrane area of 25000 m². So the concentration of the CO_2 in the permeate does not have a big influence on the purity of the CO_2 product stream due to its small size compared to the total flue gas stream. The purity can be maintained close to the level of the basic process even for higher CCR.

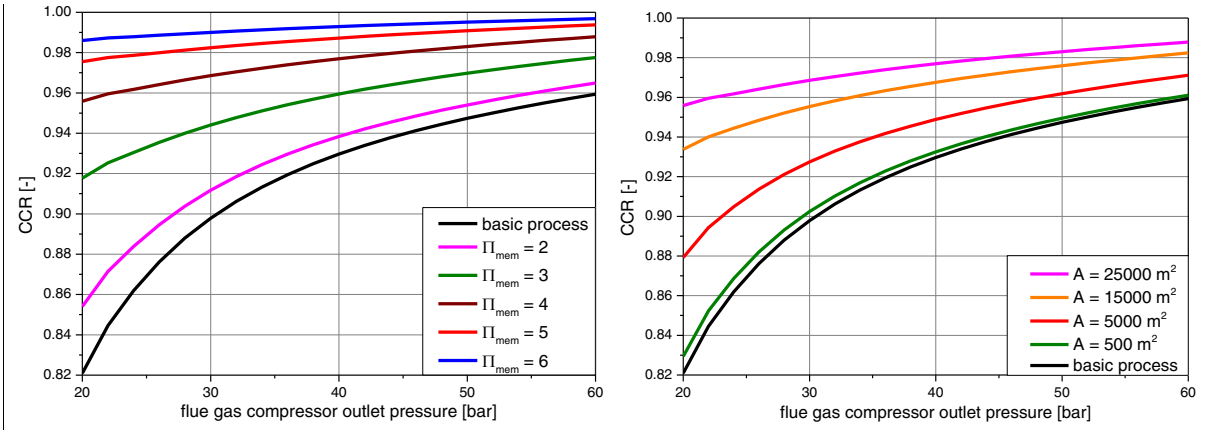

Figure 6: Left: CCR of the GPU process with and without additional PM for different Π_{mem} and a membrane size of 25000 m². Right: CCR of the GPU process with and without additional PM for different membrane sizes and a Π_{mem} of 4.The values are shown over the outlet pressure of the flue gas compressor.

The CCR for a GPU with and without an additional PM is shown in Figure 6. On the left the CCR is shown depending on Π_{mem} and the outlet pressure of the flue gas compressor. The CCR rises with higher pressure levels at the flue gas compressor outlet. A higher Π_{mem} also leads to an increased CCR. Depending on Π_{mem} and the outlet pressure of the flue gas compressor it is possible to achieve a CCR of over 99 % for the chosen process design with a membrane area of 25000 m². In Figure 6 right the CCR is shown as a function of the outlet pressure of the flue gas compressor and the membrane size for Π_{mem} = 4. The increase of the CCR for a constant outlet pressure of the flue gas compressor lessens with a rising membrane area. In addition the CCR increases with a bigger membrane area. The effect is decreasing with a larger membrane area. With Π_{mem} = 4 a CCR of above 99 % is possible.

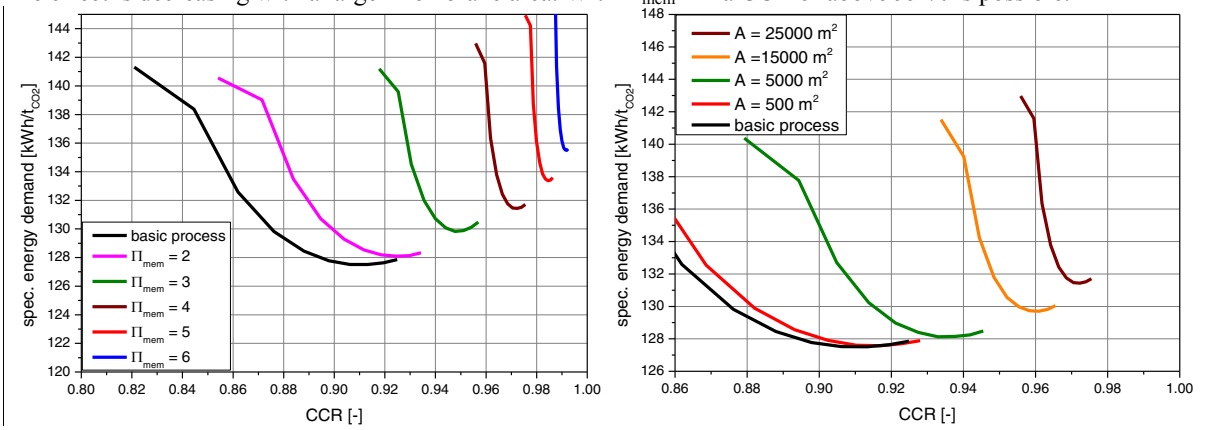

Figure 7: Left: Specific energy demand of the GPU with and without PM depending on the CCR for different Π_{mem} and a membrane area of 25000 m². Right: Specific energy demand of the GPU with and without PM depending on the CCR for different membrane areas and Π_{mem} = 4.

The third parameter evaluated here is the specific energy demand of the GPU depending on membrane area and Π_{mem}. The curves in Figure 7 show a similar behaviour to the basic process. All of them have a minimum. These minima can be found at higher CCR but also with increasing specific energy demand for increasing Π_{mem} and for increasing membrane area. The curves stop shortly after the minima. This is due to the constraint of maintaining a CO_2 purity in the captured CO_2 above 96 vol.-%. The lines stop where the purity drops below 96 vol.-%. If a lower

purity would be accepted, the curves would show a similar behaviour like in Figure 2 right.

3.2. Effects of the PM process on the overall process

The PM does not give any feedback to the overall process except for an increased energy demand. As shown in Figure 8 the net efficiency of the overall process decreases with a rising CCR. The efficiency penalty caused by the PM mostly depends on the amount of recycled gas that has to be compressed again in the flue gas compressor. In addition to that the captured gas stream cannot be expanded anymore and cannot be used for cooling demands and energy recovery in the GPU (see Figure 4). The efficiency penalty has a similar trend to the curves shown in Figure 7 left and right. In Figure 8 the efficiency loss due to the higher capture rate is shown. The line shows the efficiency loss for the basic process when the CCR is increased by an increased outlet pressure of the flue gas compressor. The dashed line shows the CCR when the CO_2 purity for the basic process drops under 96 vol.-%. The points represent the minima of the curves in Figure 7 for different PM processes. The points are shown for a membrane size of 5000 m² and 25000 m². Π_{mem} is varied between 2 and 6. The basic process has the minimum of the specific energy demand at the CCR of around 91 % (see Figure 7). With increasing CCR the efficiency loss also rises. For the PM processes the efficiency loss due to higher CCR is comparable with the basic process efficiency losses.

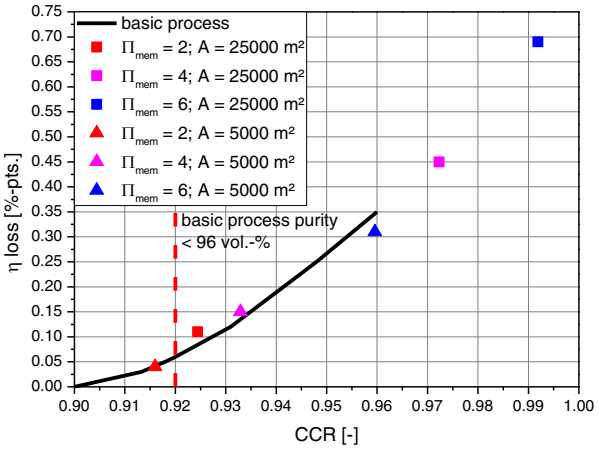

Figure 8: Efficiency loss due to the increased CCR for the membrane sizes 5000 m² and 25000 m² and Π_{mem} 2-6.

Despite the very high CCR with the PM processes, the purity is maintained at >96 vol.-% while the basic process cannot maintain this purity at CCR above 92 %. Therefore a CCR of over 92 % can only be achieved with a PM process when the CO_2 purity has to be higher than 96 vol.-%. For CCR below 92 % the basic process is preferable to the membrane process because of the simpler process design.

3.3. Oxygen recovery from the remaining exhaust gas downstream of the PM

The exhaust gas downstream of the PM contains excess oxygen from the overall process. For the considered process configuration with a membrane area of 5000 m² and 25000 m² and a Π_{mem} of 6 the oxygen contained in the exhaust gas varies depending on the outlet pressure of the flue gas compressor. For the PM with an area of 5000 m² 2.6-4.3% and for the PM with an area 25000 m² 2.4-4.2% of the oxygen supplied by the ASU is contained in the exhaust gas. This can be recycled to the ASU to recover the oxygen and lower the energy demand of the ASU. To generate a benefit for the ASU process the exhaust gas has to be recycled to the ASU on a pressure level of 5 bar. Due to this only PM configurations with a high Π_{mem} can be considered. Therefore the recycle results in a lowered energy recovery in the expanders of the GPU (see Figure 4). Depending on the flue gas compressor outlet pressure

the CO_2 concentration in the exhaust gas varies from 11.2-27.5% for the 5000 m² PM or 2.3-6.3 % for the 25000 m² PM. This CO_2 has to be captured by the molecular sieve upstream of the ASU to prevent it from freezing in the heat exchangers of the ASU. For the regeneration of the molecular sieve steam from the water-/steam-cycle is used. These influences on the energy saving potential are calculated. The results are shown in Table 4. The total efficiency potential is comparable for both membrane sizes with 0.22-0.23 %-pts. efficiency gain. The losses in the ASU process are comparable as well for both membrane sizes. The expander losses are higher for the smaller PM due to the larger retentate mass flow that cannot be expanded anymore to ambient pressure in the GPU. The efficiency loss because of the molecular sieve regeneration shows an advantage for the larger PM. The CO_2 contained in the exhaust gas is less than for the process with the smaller PM as can be seen in the higher CCR (see Figure 8).

Table 4: Efficiency gain by an exhaust gas recycle to the ASU for oxygen recovery.

	A = 5000 m², Π_{mem} = 6	A = 25000 m², Π_{mem} = 6
Total efficiency potential	0.23 %-pts.	0.22 %-pts.
Loss in ASU	-0.035 %-pts.	-0.033 %-pts.
Loss in GPU expanders	-0.115 %-pts.	-0.097 %-pts.
Molecular sieve regeneration loss	-0.166 %-pts.	-0.031 %-pts.
Efficiency gain	-0.086%-pts.	0.059 %-pts.

There is small efficiency gain by an exhaust gas recycle if the PM captures most of the CO_2 contained in the ventgas. For the PM process with a membrane area of 25000 m² and a Π_{mem} of 6 the total efficiency loss due to the additional capture process could be reduced from 0.69 %-pts. to 0.63 %-pts. For smaller membrane sizes there is no benefit by an exhaust gas recycle due to the high amount of CO_2 that is still contained in the exhaust gas and due to the higher losses of the expander train.

4. Conclusion

A PM process as considered in this work has the potential to increase the CCR of the Oxyfuel power plant up to over 99 %. It shows a good performance especially at highest CCR. The main advantage is that the CO_2 concentration of the captured CO_2 can be maintained above 96 vol.-% while the net efficiency loss is in a similar range as the basic process. The efficiency loss results in 0.04-0.69 %-pts., depending on the CCR achieved. For highest CCR with only small amounts of CO_2 remaining in the exhaust gas downstream of the PM process a recycle of the exhaust gas to the ASU for oxygen recovery can be applied. Thereby the efficiency loss due to the higher CCR can be reduced. However this is highly complicated for the overall process and the achieved efficiency gain is small and only works when a triple column ASU is used. Therefore it is recommended to check carefully if an exhaust gas recycle should be applied.

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