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Integration of Oxygen-containing Exhaust Gas into the Air Separation Unit of an Oxyfuel Power Plant

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Abstract

The Oxyfuel process is one possible solution to lower the high specific CO_2 emissions of coal-fired power plants. The flue gas generated by this process has a high CO_2 content, due to the missing nitrogen. However the dried flue gas still contains approximately 15 vol% (dry) impurities (NO_x, SO_x, O_2, N_2 and Ar). To further increase the CO_2 purity the flue gas needs to be treated in a gas processing unit (GPU). Additional downstream gas treatment approaches such as Polymeric Membranes or Pressure Swing Adsorption to capture CO_2 from the offgas of the GPU, can raise the capture rate from 90% to 99%. The remaining CO_2 and most of the impurities contained in the offgas downstream of the GPU are discharged to the environment with the exhaust gas of the GPU. The O_2 contained in the exhaust gas downstream of the GPU amounts to 4-5% of the oxygen supplied by the ASU. The energy demand of the ASU can be lowered if the O_2 contained in this exhaust gas was captured and recycled back to the firing system. The energy saving potential of such a recycle would amount to 4-5% of the ASU energy demand, but only if all O_2 is captured from the exhaust gas and no energy is needed for the capturing process. So the capturing process and its integration into the overall process has a substantial influence on the achievable energy saving. Target of this work is to integrate and model the capture and recycle of the oxygen from the exhaust gas back into the Oxyfuel power plant process.

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Keywords: CO_2 capture; Oxyfuel; Optimisation

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1. Introduction

To alleviate climate change it is necessary to reduce anthropogenic climate gas emissions. The application of carbon capture and storage (CCS) technologies is a solution to lower the high specific CO_2 emissions of coal-fired power plants. One of these CCS technologies is the Oxyfuel process.
In Fig.1 the Oxyfuel process with cryogenic air separation unit (ASU) is shown. The coal is burned in a mixed atmosphere of O_2 and recycled flue gas. The flue gas recycle is necessary to keep the furnace temperatures within technical limits of used boiler materials[1], [2]. The change of atmosphere in the boiler leads to a different combustion behaviour and different pollutant formation than in air firing [2]. Due to the missing nitrogen and the recirculation, the amount of all other gas components is increased compared to air firing [1].

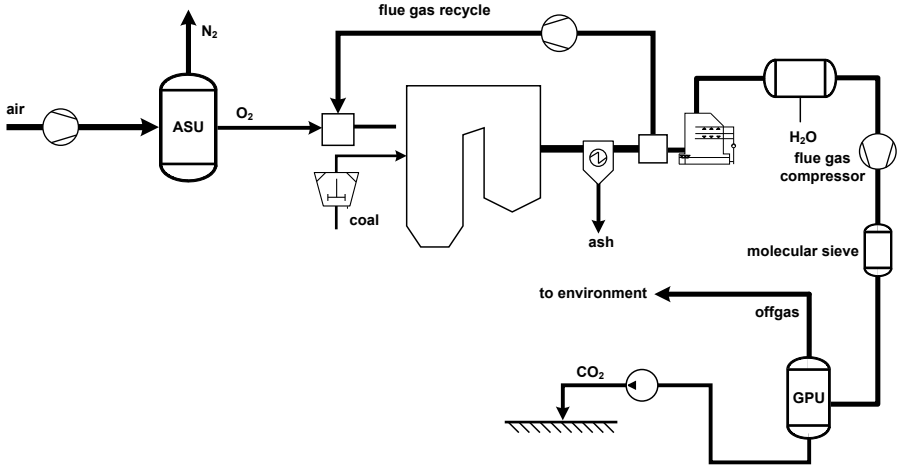

Figure 1: Basic Oxyfuel power plant with cryogenic air separation unit and CO_2 capture through externally cooled partial condensation..

The flue gas generated has a high CO_2 content of about 85 vol% (dry), because of the missing nitrogen. However, the dried flue gas still contains approximately 15 vol% (dry) of impurities (NO_x, SO_x, O_2, N_2 and Ar). To purify the CO_2 further for storage or other utilisation, the flue gas needs to be treated in a gas processing unit (GPU). The mostly assumed CO_2 capture rate of the process itself is 90%. Current research suggests an additional gas treatment downstream of the GPU. Approaches such as Polymeric Membranes (PM) or Pressure Swing Adsorption (PSA) can be applied to capture CO_2 from the offgas of the GPU (see Fig. 2) [3], [4], [5], [6]. These can raise the capture rate from 90% to 99% [7], [8]. The remaining CO_2 and most of the impurities contained in the offgas downstream of the GPU (see Fig. 2) are released to the environment with the exhaust gas of the GPU.
The exhaust gas contains oxygen, that amounts to 4-5 wt% (dry) of the oxygen originally supplied by the ASU. If captured and recycled to the steam generator, this oxygen can reduce the energy demand of the ASU by the same amount. The possible overall process electrical net efficiency increase from such a recycle are examined in this work. Furthermore the possibilities to capture this oxygen out of the offgas are compared. Modelling of the overall process is done with the commercial software *EbsilonProfessional®* whereas modelling of ASU and GPU is done with the commercial software *AspenPlus®.*

2. Basics and modelling assumptions

The water steam cycle of the Oxyfuel processes modelled is based on a state-of-the-art air blown bituminous coal-fired power plant with an electrical power output of 600 MW gross [9]. The main assumptions for this reference plant and the bituminous coal are given in Table 1. The Oxyfuel adaption is modelled with matured, state-of-the-art technologies, to ensure that a realisation in the near future is possible.

2.1 Overall process modelling

The oxygen for the combustion is supplied by a cryogenic ASU with a purity of 95 vol% (dry). The ASU is a dual column with a liquid oxygen (LOX) boiler and an adiabatic compression operated main air compressor. The oxygen is preheated to a temperature of 183 °C with the waste heat of the compression within the ASU island. Further waste heat of the compression is integrated into the power plant by preheating parts of the condensate. While the coal is transported with recycled flue gas and fed into the furnace the rest of the recycle is mixed with supplied oxygen.

Table 1: Ultimate analysis of the South African coal and the assumptions of the basic air blown power plant process.

Ultimate analysis		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	Ambient temperature	15 °C
N	0.016 kg/kg	Efficiency (gross)	49.4%
O	0.066 kg/kg	Efficiency (net)	45.8%
S	0.0057 kg/kg	Spec. CO_2-emissions	750.7 g_{CO2}/kWh

The part of the flue gas mixed with the oxygen upstream of the furnace is cleaned in a hot electrostatic precipitator (see Fig.1). The flue gas used to dry and transport the coal (primary recycle) is extracted downstream of a wet flue gas desulphurisation to protect the coal mill from corrosion.

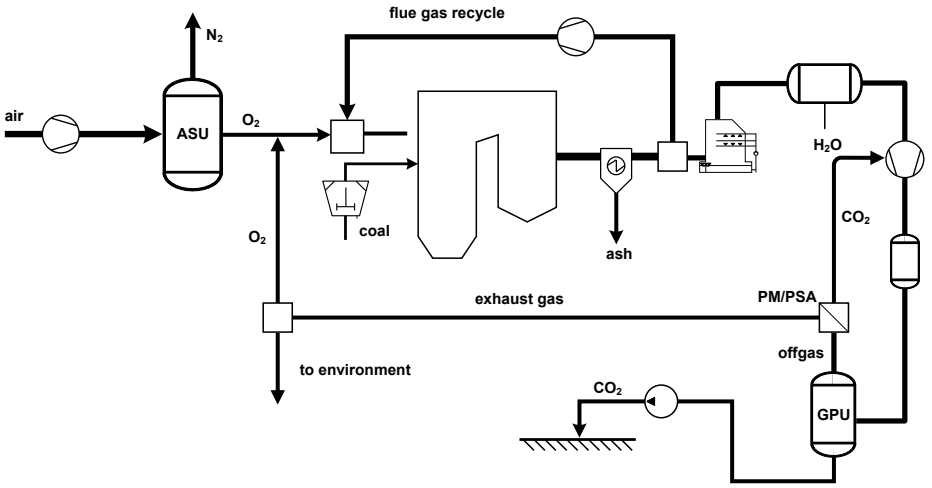

Figure 2: Oxyfuel power plant with additional gas treatment and O_2 recycle back to the steam generator.

After drying, the remaining flue gas is compressed and completely dewatered in a molecular sieve before entering the GPU. Inside the GPU at first the flue gas is cooled down to a temperature of -25 °C. At these conditions CO_2 starts to condensate. The liquid CO_2 contains only a very small amount of impurities. The remaining vapour is cooled down in a second stage to -46°C where a second CO_2-enriched liquid fraction can be separated. Both liquid fractions, which contain about 90% of the total CO_2, are mixed and pumped up to a pressure of 110 bar. The supercritical stream is used to cool the flue gas downstream of the compression. The remaining offgas is expanded

and used in the GPU for cooling. The main data of the GPU are given in the left column of Table 2. Since the offgas of the GPU still contains about 10% of the total CO_2 and is available at high pressure and low temperature it is suitable for an additional gas treatment with membrane or pressure swing adsorption [6], [8], [3], [5], [7] (see Fig.2).

In previous research [7] the additional offgas treatment is realised with a PEO membrane [10] (see Table 2) that is applied directly downstream of the CO_2 capture within the GPU. The pressure ratio from feed to permeate is kept constant with a value of three. As the captured CO_2 has a purity similar to the flue gas upstream the GPU, it should be recycled to a compressor stage with a slightly lower inlet pressure. Moreover this recycling is necessary to liquidise the captured CO_2 again. The recycle leads to an increase in the GPUs energy demand. This is due to the increased amount of gas treated and the reduced mass flow used for power recovery in the expanders and the cooling in the GPU [7].

Table 2: Process data of the GPU with and without offgas treatment. Given gas concentrations are for the offgas w/o offgas treatment and for the exhaust gas for the scenario with offgas treatment. Parameters of the membrane used for the additional CO_2 capture are given below.

Process parameters	w/o offgas treatment	With offgas treatment
CO_2 purity in vol% (dry)	97	97
Capture rate in %	90	99
Specific energy demand GPU in kWh/t_{CO2}	131	133
Overall process efficiency in %	36.9	36.4
Gas concentrations downstream GPU in vol% (dry):	Offgas	Exhaust gas
CO_2	33.4	5.3
N_2	36.5	52.3
O_2	16.3	22.7
Ar	13.8	19.7
p in bar	30	29.5
T in °C	-46	25
$\dot{m}$ in kg/s	27.98	17.25
Membrane Data PEO		
Selectivity CO_2/N_2		50
Selectivity O_2/N_2		2.8
Selectivity Ar/N_2		2.8
Permeance CO_2 in m³(STP)/m²hbar		3
Membrane area in m²		5982

The efficiency penalty in comparison to the reference power plant for the basic Oxyfuel process amounts to 8.9 %-pts. and to 9.4 %-pts. for the Oxyfuel process with additional gas treatment. The difference between both scenarios is 0.5 %-pts. To lessen this penalty the oxygen contained in the exhaust gas could be recovered and used to decrease the energy demand of the ASU. This potential energy saving amounts to 4.1% (see Table 4) of the energy demand of the ASU. This corresponds to the amount of air oxygen that would no longer be needed and still supply the same amount of oxygen to the combustion chamber as in the basic scenario. In Table 2 the composition of the exhaust gas is displayed. Composition, pressure and temperature of the exhaust gas are important to determine the most suitable process to capture this oxygen. As can be seen the pressure is still about 30 bar at a moderate temperature. Though separating the oxygen from the exhaust gas is problematic, because of the relatively high amount of argon contained in the exhaust gas. Ar and N_2 have to be removed, otherwise they would accumulate within the process. As Ar is more difficult to remove from O_2 than N_2, different processes can be used for this gas separation.

The selectivity of O_2/Ar for membranes is reported in the literature as close to 1[10]. Therefore a separation of the oxygen with membranes would not be effective. The same problem is reported for adsorbents in pressure swing adsorption [11]. That is why the most effective possibility for the separation of oxygen from the exhaust gas is to recycle the exhaust gas to the ASU. This solution is also discussed in literature [4], [6]. The separation of Ar and O_2 still remains more complicated than the separation of N_2 and O_2 [12].
It has to be kept in mind that the high pressure (see Table 2) is only available theoretically, because the gas is used for cooling and power regain in the process. If the gas shall be used at the highest pressure and is not used for cooling and power regain in the GPU, the specific energy demand of the GPU increases.

2.2 Air separation unit

As described in the previous section a cryogenic ASU unit for oxygen supply is used in the process. The ASU process modelled for the basic Oxyfuel process is a dual column ASU with a LOX-boiler and a dual reboiler (see Fig.3). The product is GOX with a purity of 95 vol% (dry) [13], [8]. The air compression can be either adiabatic to maximise the overall process efficiency or isothermal for a simpler process integration.
The air is compressed from ambient pressure up to 4.54 bar. If the compression is adiabatic, the heat of the compression is integrated as far as possible into the oxygen and condensate preheating. Afterwards a direct contact cooler cools down the air to 12 °C. Water and CO_2 are captured in molar sieves upstream of the main heat exchanger. Downstream the air is split into two streams. One is used stream to balance the heat losses of the cold box to the environment through decompression in an expander. The mechanical power is transformed into

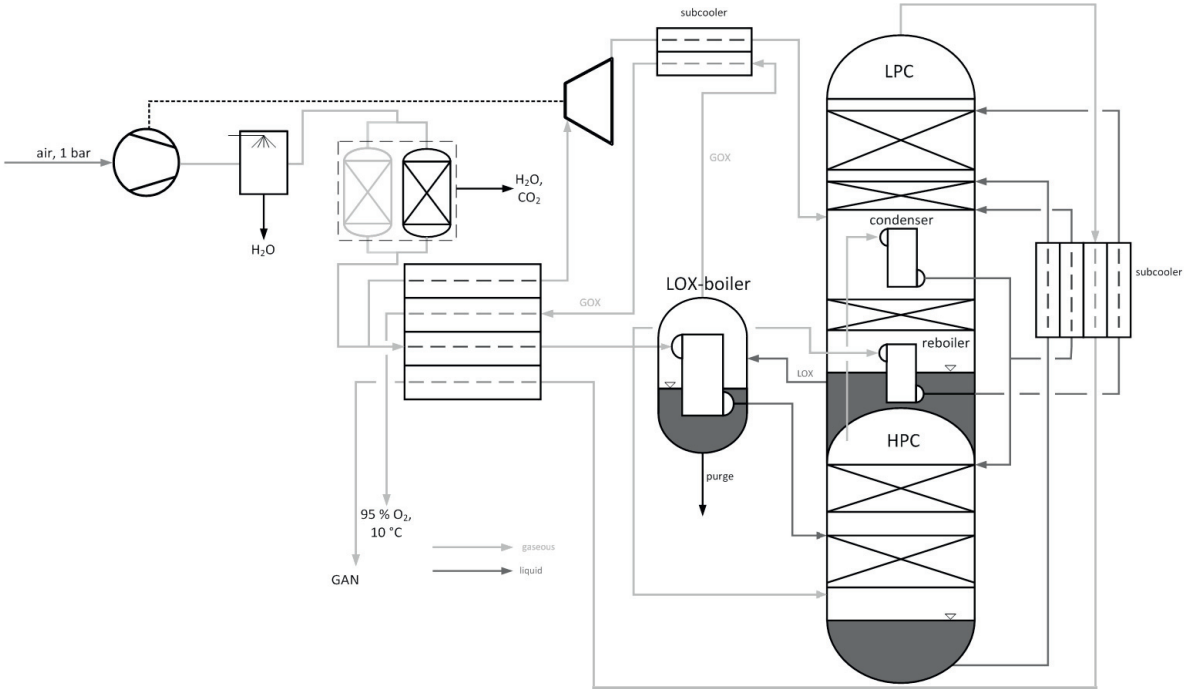

Figure 3: The basic process for the air separation is a dual column with LOX-boiler and dual reboiler which supplies oxygen with a purity of 95 vol% (dry).

electrical power and used to lessen the energy demand of the main air compressor. The stream to the expander amounts to about 11% of the total air flow and it is cooled in the main heat exchanger before entering the expander. The stream is fed to the low-pressure column (LPC) downstream of the expander. The main air flow is cooled in

the main heat exchanger. Downstream the air is split into three streams. One part is condensed in the LOX-boiler, where the liquid oxygen product (LOX) of the low pressure column (LPC) is vaporised. The liquefied stream is then fed to the high pressure column (HPC). The second fraction is fed to the reboiler of the LPC, where it condensates. The liquefied stream is subcooled by the nitrogen product of the LPC and fed to the low pressure column. The remaining air is fed directly to the HPC. The oxygen-enriched bottom product of the HPC is subcooled and fed to the LPC.
The nitrogen-enriched top product of the HPC is condensed, subcooled and fed to the LPC. The LPC products are heated up and leave the process via the main heat exchanger. The main assumptions of the process are summarised in Table 3. The ASU columns are simulated with equilibrium theoretical stages and structured packings. The pressure loss is set to 0.7 mbar per theoretical stage [14]. The specific heat losses of the coldbox to the environment are set to 30 kJ/kmol of air processed in the ASU [12]. The amount of oxygen produced by the ASU is kept constant to ensure comparability of the results. In the basic case the number of theoretical stages in the LPC is set to 47 and for the HPC to 30. These numbers are chosen to avoid high investment costs and pressure losses while achieving a high O_2 capture rate. The feed stages of the different streams into the column are optimised aiming at a low specific energy demand of the ASU.

Table 3: Process assumptions for all ASU scenarios examined. The polytropic efficiencies differ, because the adiabatic compressor is considered to be an axial compressor whereas the isothermal compressor is a geared radial compressor.

Process parameters			
Number of parallel ASU streams	3	Compressor mechanical efficiency	0.98
Oxygen product flow in kg/s	31.95	Engine efficiency	0.97
Air temperature upstream coldbox in °C	12	Engine mechanical efficiency	0.998
Head pressure LPC in bar	1.34	Expander efficiency	0.84
Oxygen product outlet pressure	1.16	Expander mechanical efficiency	0.98
LMTD heat exchangers in K	2	High pressure column (HPC) theoretical stages	30
Compressor polytropic efficiency adiabatic/isothermal	0.89/0.85	Low pressure column (LPC) theoretical stages	47

2.3 Integration of the exhaust gas into the ASU

To obtain energy savings from the recycle it is necessary to recycle the exhaust gas at least at the pressure of the ASU, which in the basis scenario would be 4.54 bar. For the integration of the exhaust gas, there are different possibilities:

- Scenario A: Direct mixture with the compressed air upstream of the direct contact cooler without changing the feed stages into the columns to optimise the separation process
- Scenario B: Direct mixture with the compressed air upstream of the direct contact cooler with optimisation of the feed stages into the columns to optimise the separation process
- Scenario C: Separate cooling and feed to the columns without changing the feed stages into the columns to optimise the separation process
- Scenario D: Separate cooling and feed to the columns with optimisation of the feed stages into the columns to optimise the separation process

Both scenarios A and B have in common that the mixture of the exhaust gas to the pressurised air is realised upstream of the direct contact cooler. The concentration of oxygen in the exhaust gas (see Table 2) is similar to the concentration of oxygen in air. Therefore direct mixing could be sufficient. Furthermore the temperature of the exhaust gas is 25 °C, that leads to a similar cooling demand like for the compressed air. This would be the easiest and least expensive way concerning capital and operating costs to integrate the exhaust gas into the ASU.

Scenario C and D show, whether it is preferable to realise a direct feed to the columns. Scenario A and C have the advantage, that retrofitting of an Oxyfuel power plant would be possible, because there are no changes in the column design necessary. A change in the column design would result in higher capital costs for the ASU. In addition to that it is possible to close the recycle if necessary, without much impact on the operation of the ASU.

3. Results and discussion

3.1 Integration of the exhaust gas into the ASU

The results of the modelled and calculated scenarios are displayed in Table 4. The basic scenario has a pressure level of 4.54 bar. The CO_2 stream that has to be captured in the molar sieve is 0.106 kg/s. The concentrations of the impurities N_2 and Ar in the O_2 product stream of the ASU are equal. The process configuration achieves an O_2 capture rate (captured oxygen in the product stream compared to the total oxygen fed to the ASU) of 98.48%. This results in an overall specific energy demand of about 213 kWh/t_{O2}. The exhaust gas that is recycled for the scenarios A-D is kept at the pressure level necessary to bypass the air compressors and to avoid this energy demand.

Table 4: Calculated results for the different integration scenarios tested as regards the feeding of the exhaust gas into the ASU with an isothermal compression.

Process parameters	Basic process	Scenario A	Scenario B	Scenario C	Scenario D
Outlet pressure air compressor in bar	4.54	4.71	4.66	4.69	4.57
CO_2 captured in molar sieve of the ASU in kg/s	0.106	0.52	0.51	0.51	0.51
Spec. amount of gas fed to expander	0.11	0.11	0.11	0.05	0.05
Ar in product stream in %	2.4	4.86	4.28	4.3	2.89
N_2 in product stream in %	2.6	0.14	0.72	0.7	2.11
Abs. energy demand engine of the main air compressor in kW	24488	25415	24039	24296	23896
O_2 capture rate in %	98.48	93.49	97.9	98.26	98.27
Spec. energy demand in kWh/t_{O2}	213	221	209	211	208
Potential energy saving in %	-	4.1	4.1	4.1	4.1
Achieved energy saving in %	-	-3.8	1.9	0.9	2.4

Scenario A envisages the direct mixing of the exhaust gas stream with the pressurised air stream prior to direct contact cooler. The feed stages are not optimised for the new concentrations of the gas entering the coldbox. The pressure level is increased compared to the basic process. The reason for this increase can be seen in the change of the concentrations of the impurities in the O_2 product stream. The impurities almost completely consist of Ar. In addition to that the capture rate decreases in magnitude of 5%. The results indicate, that the high amount of Ar (increase of about 70%) that is fed to the ASU cannot be separated in the same configuration as in the basic process. To achieve a purity of 95 vol% (dry) it is necessary to increase the heating duty in the reboiler of the LPC. This causes a higher amount of O_2 being caught in the head product of the column. Consequently the O_2 capture rate decreases significantly and there is no benefit left out of the recycle. Instead the recycle leads to an increased specific energy demand of 221 kWh/t_{O2}.

Scenario B also is designed to have a direct mixing of the exhaust gas with the pressurised air upstream of the direct contact cooler. In this scenario the feed stages of the streams to the columns have been varied to achieve the highest possible energy saving compared to the basic process. Again the concentration of Ar in the product rises significantly suggesting also here difficulties in separating the Ar from the O_2. The O_2 capture rate decreases only

to 97.9%. Consequently there is a benefit for the specific energy demand which decreases to 209 kwh/t_{O2}. The energy saving achieved compared with the theoretical maximum energy saving is 40 % for this scenario.
Scenario C is modelled with separate cooling of the exhaust gas stream instead of a mixture upstream of the direct contact cooler. The exhaust gas is cooled down as far as possible in the main heat exchanger and downstream it is depressurised in an expander. Then it is fed to the LPC. The concentrations show again a high amount of Ar in the product stream. However, the capture rate decreases only a little compared to the basic process. The specific energy demand decreases to 211 kWh/t_{O2}. This scenario indicates, that a separate feed to the coldbox leads to a better separation performance of the columns and furthermore it profits from the smaller amount of compressed air that has to be fed to the expanders (see Table 4).
Scenario D is similar to scenario C except that the feed stages of the columns are optimised to yield the lowest specific energy demand. The concentration of Ar in the impurities of the O_2 product stream is slightly increased compared to the basic scenario. But it is still much lower than in scenario A, B and C. The capture rate is also high with 98.27%. This shows again that the process profits from a separate feed to the columns. Scenario D achieves the lowest specific energy demand with 208 kwh/t_{O2}. This is 60% of the potential energy saving.

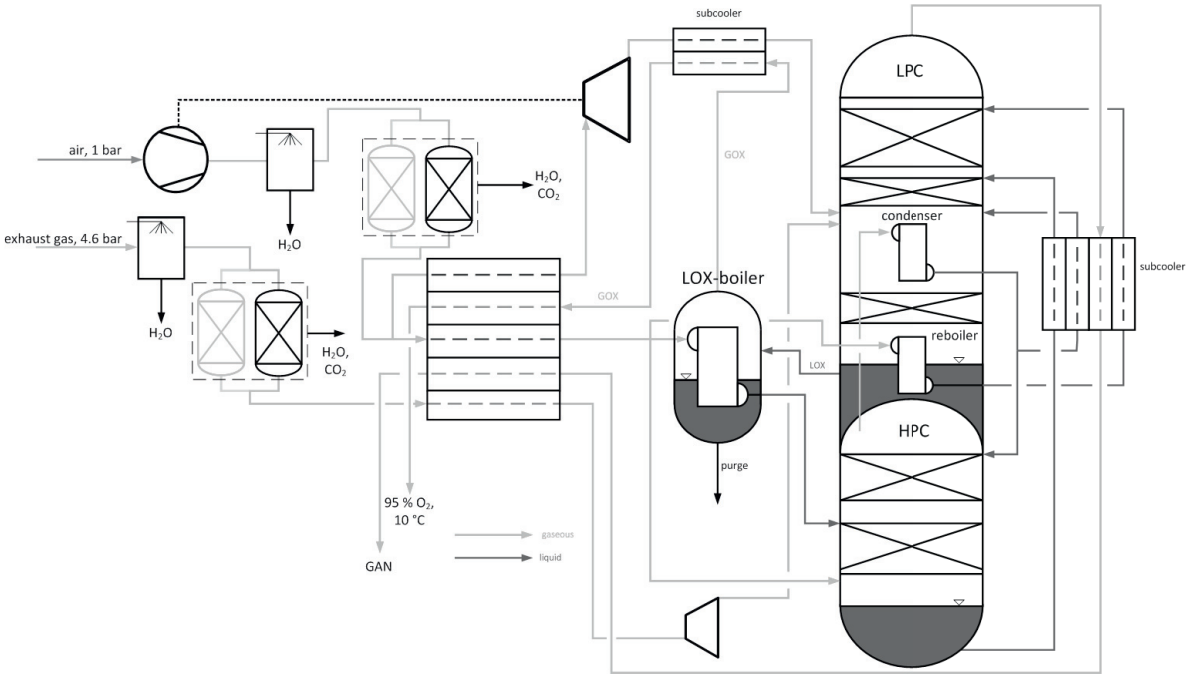

Figure 4: Dual column dual reboiler ASU with LOX boil and exhaust gas recycle. The exhaust gas is separately fed to the coldbox.

The results of the four scenarios examined here indicate, that a dual column has difficulties to separate the oxygen from the exhaust gas, if the ASU process design were to remain unaltered. More precisely the high amount of Ar contained in the exhaust gas causes separation difficulties. The results show an advantage for a separate feed of the compressed air and the exhaust gas to the coldbox. Still the energy saving utilisation is with highest 60% not as high as expected. So, even with feed stage adjustment that would cause a lowered flexibility of the ASU and of the overall power plant from the fact that the exhaust gas recycle configuration is open or closed, the benefit cannot exceed 60%. Still it has to be examined however, if this benefit is high enough to increase overall process efficiency and whether it is economical or not.

3.2 Influence on the overall process

The scenarios B, C and D show a benefit of the exhaust gas recycle to ASU on the specific energy demand of the power plant. Still it has to be integrated into the overall process for achieving a final assessment of the potential. The recycle is at a pressure level of 4.6-4.8 bar. As described in section 2.1 this pressure influences the energy demand of the GPU, because the decompression cannot be used in the expanders of the GPU anymore. To see if there is a benefit to the overall process it is simulated with the software used in this study.
The process flow sheet is the same as in Fig. 2 and the process characteristics are still the same as in Table 2. The compression of the ASU implemented into the overall process is adiabatic. The energy demand for the compression increases. The compression heat is integrated into the process by preheating a condensate bypass. In this manner an overall benefit from an adiabatic compression can ensue. The benefits due to the O_2 recycle are the same as for the isothermal compression, because there are no changes in the coldbox. The specific energy demand of the basic ASU with adiabatic compression is 231 kWh/t_{O2}. With a recycle of the exhaust gas, the specific energy demand can be lowered down to 225 kWh/t_{O2}. The results of the simulation show that the overall process efficiency does not rise though. The efficiency for the scenario with additional gas treatment, but without exhaust gas recycle is 36.4 %-pts. (see Table 2). The exhaust gas recycle leads to an overall process efficiency of 36.38 %-pts. This efficiency decrease derives from the mentioned problem with the GPU expander. The exhaust gas recycle leads to a total energy saving of only 1.8 MW_{el} for the three parallel streams in the ASU for the scenario B, which here shows the highest energy saving potential. If the exhaust gas exits the expander with the pressure of 4.6 bar, the expander of the GPU produces about 2 MW_{el} less. So the benefit for the ASU results in a decrease in efficiency for the overall process. The break even for the overall process would be achieved when the energy saving in the ASU is 2 MW_{el}. This corresponds to an achieved relative energy saving of 66% of the possible energy saving. These results show that an economical evaluation for such a recycle with a dual column ASU is not necessary, because there is no benefit for the overall process even with highest energy saving in the ASU.

4. Conclusion

The results show that for an Oxyfuel coal-fired power plant recycling of the exhaust gas to the ASU is a complicated option, because of the high concentration of Ar contained in the exhaust gas. Ar influences strongly the capture performance of the ASU. As a consequence of an exhaust gas recycling always leads to a lowered O_2 capture rate (see Table 4). Therefore recycling the exhaust gas without changing the basic column design results in a higher energy demand than the basic process. But even if the column and the feed stages of the different streams were adapted to an exhaust gas recycle, the benefit still is lower than expected. This benefit is not sufficient to balance the energy losses in the GPU because of the lower energy recovery in the expander.
This study shows that from the maximum available energy saving 60% has to be regained to achieve a higher overall process efficiency. For a dual column it is not recommended to build such a recycle of the exhaust gas. Current research with the triple column indicates a better performance in the separation of the Ar and the O_2, which constitutes the main problem of the dual column. This is now examined further and promises a higher benefit for the specific energy demand than the dual column. For a dual column process no economic benefits can be gained.

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