Oxyfuel Process for Hard Coal Power Plants with CO_2-Removal

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Introduction

The removal of CO_2 from the flue gases of conventional hard coal power stations, where volumetric concentrations of about 15 % are reached, imposes very high energy demands. The Oxyfuel process proposes to increase the CO_2 concentration in the flue gas by burning coal with pure oxygen instead of air. An upstream cryogenic Air Separation Unit provides the required oxygen. To reduce temperatures in the boiler, flue gas is recycled. Here, various configurations for the location of the recycle branching within the flue gas treatment are possible. With Oxyfuel combustion it is possible to achieve CO_2 concentrations of above 90% in the dry flue gas and the liquefaction of the exhaust gas now can be managed with a lower energy input. Taking into account that a direct compression of the exhaust gas would result in high impurities in the CO_2 stream, one has to study alternative options. The research at the Hamburg University of Technology (TUHH) investigates the cryogenic separation of the flue gas, leading to a relatively pure CO_2 stream, which will supposedly not interfere with the geologic formations where it is to be stored. Therefore, the Oxyfuel process represents an attractive technology for relatively efficient removal of the CO_2 from the flue gas of fossil fuel combustion plants and preparing it for sequestration.

Current research work at Hamburg University of Technology aims to identify the key factors which have the most significant influence on establishing and running such power plants efficiently and to analyse their interaction. The analyses are explicitly performed under operating conditions which correspond to those occurring in actual power plants.

Boundary Conditions

The research at the TUHH is based on state of the art components and developments in power plant technology, which are described in the so called Reference Plant North-Rhine Westphalia, a study of an advanced plant in 2004. This study defines the latest technology when it comes to planning a hard coal plant in Germany. The main parameters are:

- Steam parameters: 600 °C / 620 °C / 292,5 bar
- Gross Power Output: 600 MW
- Net Efficiency: 45,9%

One of the considerations concerning Oxyfuel combustion is to reduce the present-day excess oxygen rate of 17% in modern hard coal plants down to 5%. This idea came up in order to reduce the amount of impurities in the flue gas. In Oxyfuel combustion it seems possible to reduce the excess oxygen because of the higher oxygen concentration in the boiler. It has to be noticed, that the amount of excess oxygen is not an issue of the combustion rate only but also of mixing gas with pulverised hard coal in the combustion chamber. Here considerable fluctuations occur which lead to temporary low stoichiometric conditions. Therefore, the research at TUHH limits the lowest permissible amount of excess oxygen to 10 % and defines 15 % as design case (this equals a stoichiometric air ratio of $\lambda = 1{,}15$). With the reference plant as a starting point one can get first conclusions concerning an Oxyfuel power plant of similar size. With a thermal capacity of 1200 MW, an amount of 48 kg coal per second will be burned. This will require an oxygen flow of 100 kg/s, which is 2-3 times as much as the present-day's largest Air Separation Unit (ASU) could provide and the total amount of CO_2 produced will be as much as 115 kg/s.

Recirculation and Oxygen Fraction

One of the main issues being addressed is the necessary amount of recirculation of cooled-down flue gas back to the steam generator. This recirculation is necessary to limit the temperatures in the combustion chamber which would be prohibitively high when burning the coal with pure oxygen. To narrow the realistic combinations of oxygen excess and oxygen fraction in the boiler to a reasonable extend, the recycle ratio that is required to achieve an adiabatic flame temperature that corresponds to air operation was calculated for different oxygen excess rates and recycle temperatures.

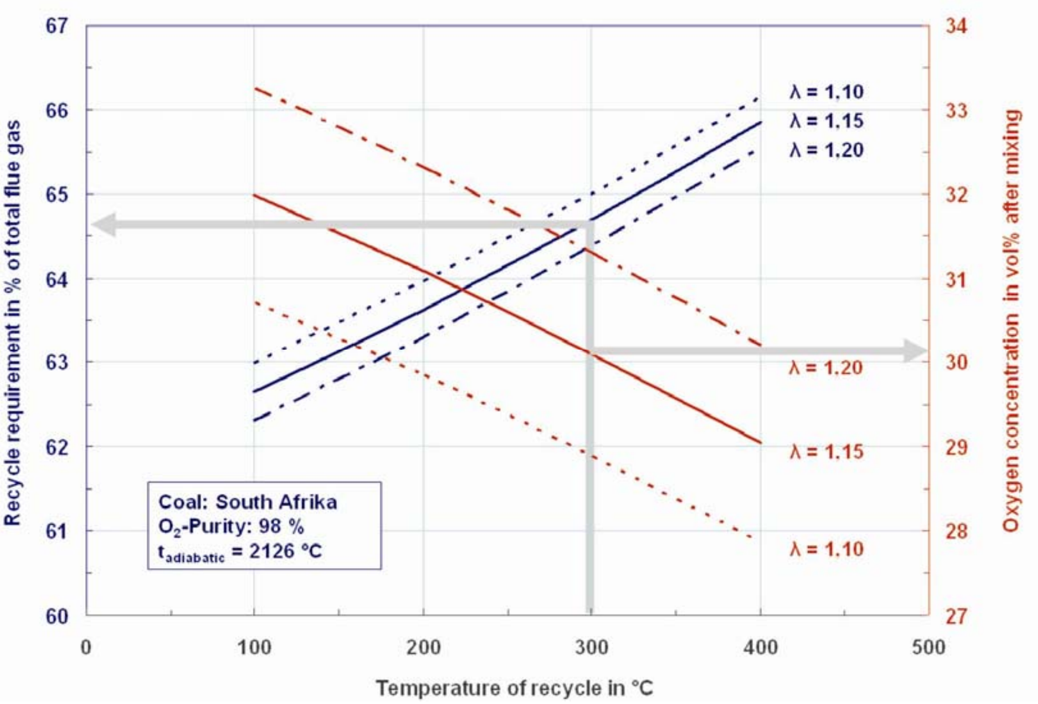

Figure 1: Dependence of required recirculation amount and oxygen concentration on recycle temperature and oxygen excess

The calculations are based on a complete burnout of the coal and include the amount of recycled flue gas. As the flue gas composition is incorporated, the recycle ratio and the oxygen fraction in the boiler are inevitably linked, just like in a proper Oxyfuel process. The determination of the adiabatic flame temperature is done using an energy balance of the boiler, taking into account gas temperatures, gas compositions, and coal properties. Consequently, air and oxygen preheating, the recycle temperature and the gas composition from the ASU can be varied. The aim was to determine the recycle ratio that leads to the same adiabatic flame temperature as the coal would reach in an air fired boiler.

As can be seen from Figure 1, South African coal reaches an adiabatic flame temperature of 2126 °C when the combustion with preheated air takes place with an oxygen excess of 15 %. In order to reach the same temperature in Oxyfuel combustion with a recycle temperature of 300 °C and an oxygen excess of 15 %, 64,5 % of the flue gas need to be recycled and the oxygen concentration in the mixture of recycled flue gas and oxygen would reach 30,5 % by volume.

These calculations were done for various coals and form the base for the combustion experiments in the vertical combustor, which are currently being undertaken at TUHH. Here, emission characteristics will be examined by comparing the results of Oxyfuel combustion to those in an air operated case. The tests will provide the oxygen concentration that will result in a similar temperature profile. This will allow for validating the assumptions mentioned above and for being able to include the results in the boiler design.

Air Leakage

A problem area identified in the Oxyfuel process is air leakage, which is already known in conventional boilers but has no major impact on the process there. As in the Oxyfuel process the CO_2 purity in the flue gas influences the energy demand of the CO_2 separation, the effect of leakage air was investigated.

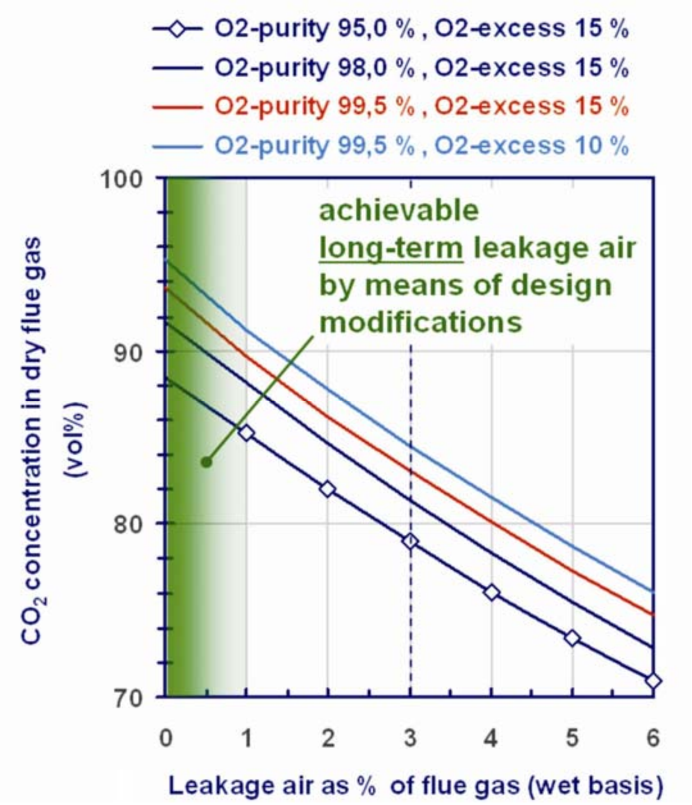

Figure 2: CO_2 purity in the flue gas as a result of leakage air, oxygen excess in the combustion chamber and oxygen purity from the ASU

In conventional air fired boilers, leakage air can rise to an amount of 10 % (related to the total flue gas volume) after some years in operation. Figure 2 shows the effect of leakage air on the CO_2 purity, which decreases rapidly due to the dilution by incoming nitrogen. The reference case with an oxygen concentration of 99,5 % and an oxygen excess of 15 % shows that even 1 % of leakage air will lead to a CO_2 purity below 90 %.

Cryogenic CO_2 Separation

As mentioned before, the cryogenic separation is a promising technique to achieve almost pure liquefied CO_2, which can be sequestrated. Until today there are still no reliable data for limits available regarding the amount of impurities for different geologic storage sites. Still, common opinion shows that especially oxygen, sulphur compounds, and NO_x are seen as critical components. Therefore, the cryogenic separation and the possibilities of an energetic integration into the plant concept are investigated. The process is based on condensation by lowering the temperature and increasing the pressure of the flue gas. Note that the water has to be separated prior to the CO_2 liquefaction because of freezing and the formation of hydrates. Drying will not be possible by condensation only but will require non neglible number of molecular sieves.

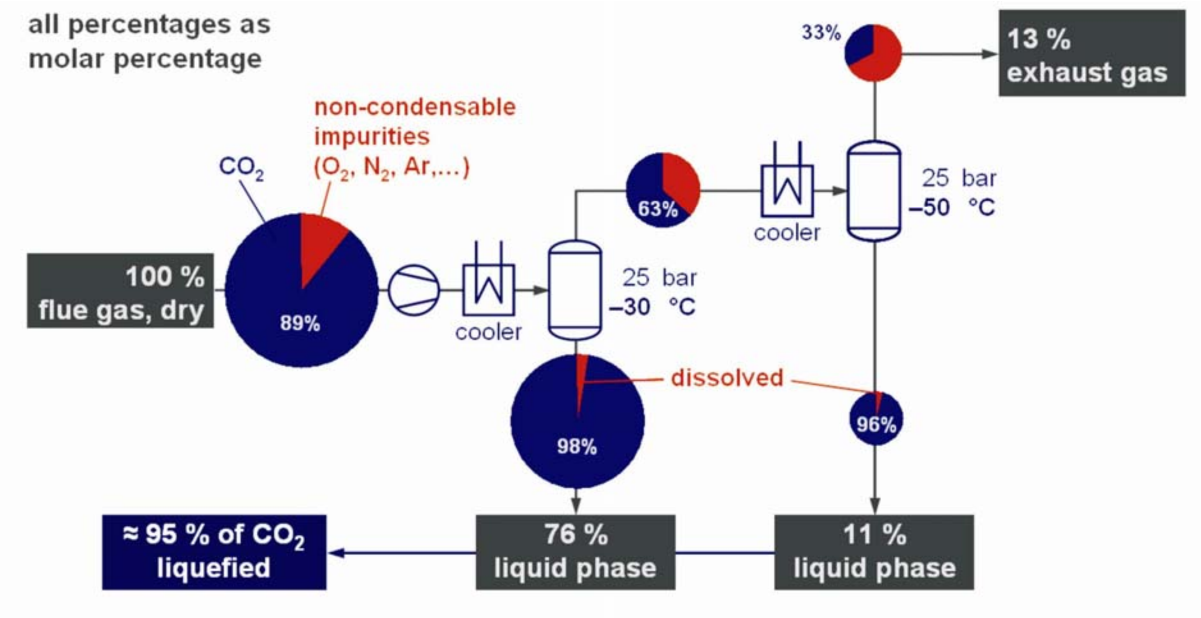

Figure 3: Scheme and approximate mass flows and CO_2 concentrations

Figure 3 presents the main process of the cryogenic liquefaction. The results were generated with the simulation software ASPEN Plus™. Therefore, the exact results for purities and mass flows will have to be verified by experimental phase equilibriums, which are currently investigated at TUHH. From the basic separation design one can already see that a complete liquefaction of the CO_2 in the flue gas stream is not possible. Further, one can identify a certain amount of impurities still included in the liquefied stream. The exact amount is a major issue of current research and is due to dissolving of a certain amount of non-condensable gas in the CO_2 stream, depending on pressure and temperature. Even in this simplified scheme there are various imaginable combinations of parameters, which for certain reasons are of no technical importance, as following figure will explain.

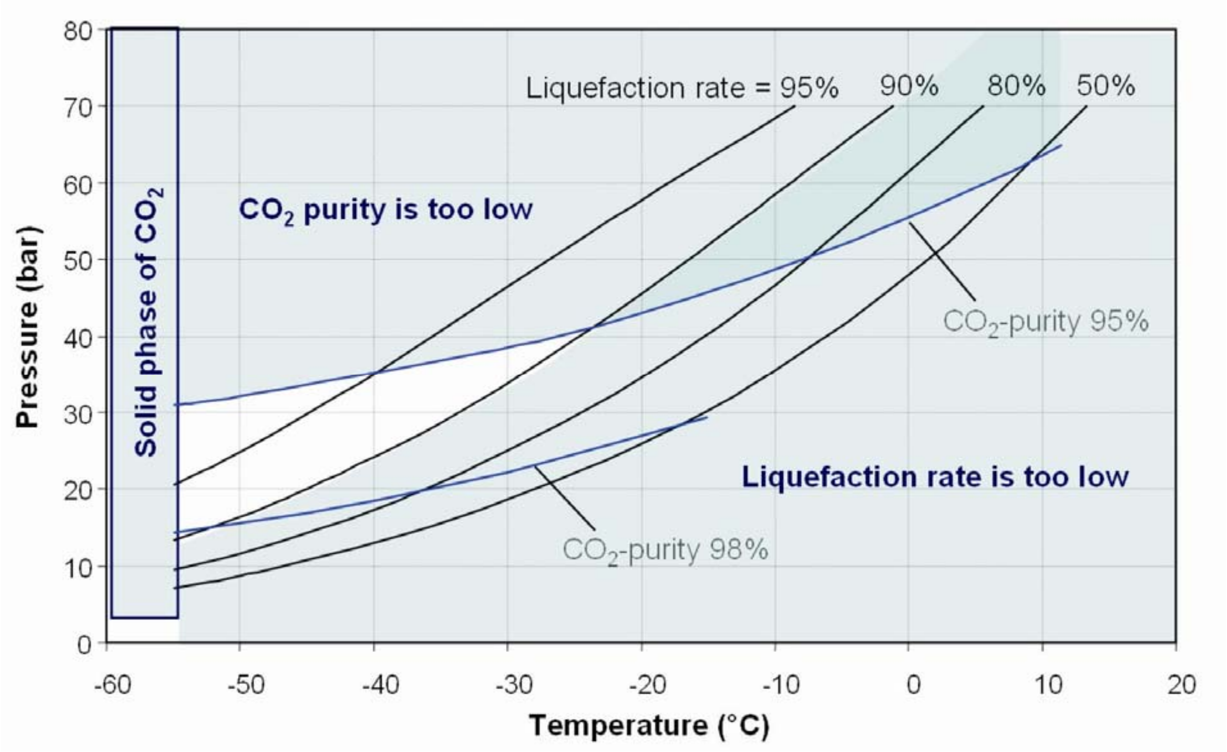

Figure 4: Dependence of cryogenic liquefaction performance on pressure and temperature

Figure 4 shows the results of a proposed cryogenic liquefaction where pressure and temperature are varied. The simulation was done using South African coal and an oxygen excess of 15 %, the presented boundaries will be specific for each set of parameters. Temperatures of -55 °C result in the formation of dry ice as CO_2 reaches its solid phase. A high pressure in the process does increase the liquefaction rate, just as low temperatures do, but leads to dissolving of impurities due to higher partial pressure of non-condensable components. As one wants to have a high amount of CO_2 liquefied and ready for sequestration, one should try to achieve a liquefaction rate of more than 90 %. These considerations result in the fact that realistic parameters for the liquefaction will be around -45 °C and 25 bar.

Additional Power Demand Due To The Oxyfuel Process

One of the key power consumers in an Oxyfuel power plant is the CO_2 separation unit, whereby the power demand is mainly depending on the process chosen for the refrigeration unit. Three processes were investigated and compared to direct flue gas compression, subsequently referred to as “zero emission”.

1. The first investigations were done on the internal cooling process. It stands for compression of the flue gas and expansion of the liquefied CO_2. This process is similar to a CO_2 refrigerator.
2. The second approach is to integrate a two-stage ammonia refrigerator to provide the required low temperatures.
3. The third possibility investigated is the use of an absorption cooler. First calculations were made with a simplified and not well integrated set-up, using only waste heat from the flue gas and a pass-out stream at 8 bar.

The results of the process simulation are presented in Figure 5, where the specific energy demand for the four possibilities is plotted for varying CO_2 purities. The liquefaction rate is held at 90 % for all process designs except the zero emission case where CO_2 and all impurities can be sequestrated.

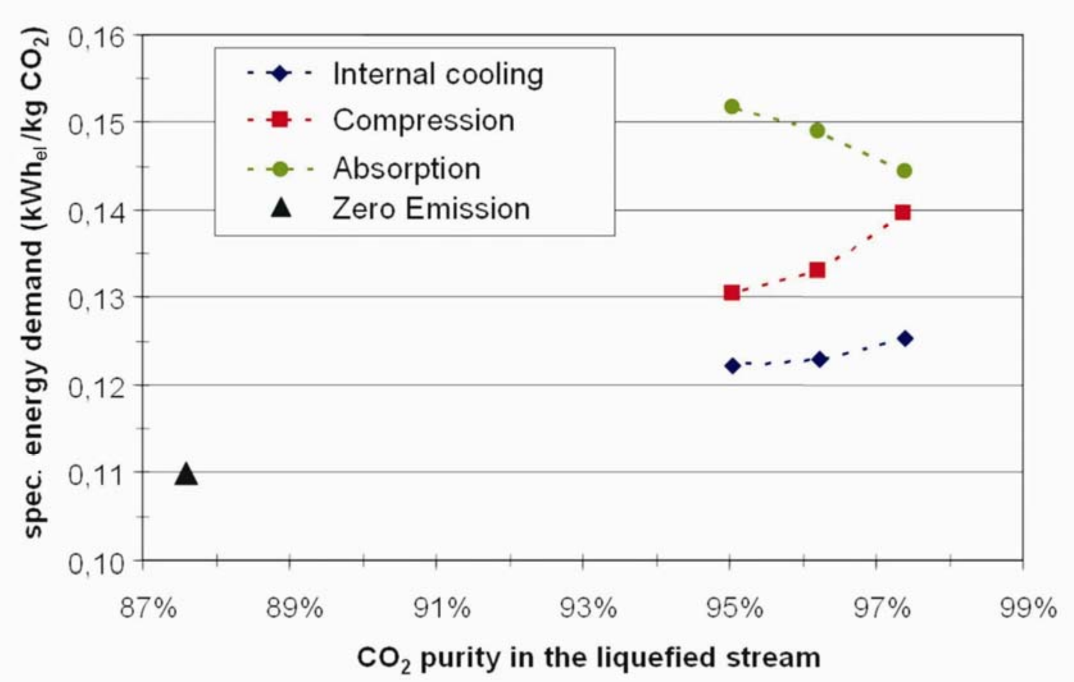

Figure 5: Comparison of the specific electric energy demand of the CO_2 separation techniques

These processes are now to be applied to the mentioned reference plant with a thermal capacity of 1200 MW. Note that the figure 5 presents the specific electric energy demand related to the amount of captured CO_2. As the aim is to reach a high CO_2 purity in the liquefied flow, the cases are to be compared with purities of more than 98 %. Under these boundary conditions, the internal cooling yields the lowest power demand, where about 45 MW_{el} are needed. The zero emission case has a similar demand but doesn't reach the specified purity. The compression and absorption cases require about 52 MW_{el}. Still, the absorption case can be a promising alternative as this process allows a high integration since the main input is heat at a relatively low temperature level.

The other key power consumer of an Oxyfuel power plant is the Air Separation Unit, which alone would consume about 90 MW_{el} when using a standard two-column process. One promising way to reduce the demand is to use an alternative Air Separation Unit concept based on the so-called three column process. This could reduce power consumption to approximately 70 MW_{el} at the expense of lower oxygen purity and higher power consumption for the CO_2 separation.

These two main additional units in the power plant process have a remarkable impact on the net efficiency of the power plant. The currently investigated cases lead to a net efficiency of about 36 %, which is a drop of almost 10 % (absolute). When you consider not only the liquefaction rate of 90 % but also the increased coal demand due to the efficiency loss the amount of captured CO_2 compared to the reference plant is 87 %.