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Comparative Thermodynamic Analysis and Integration Issues of CCS Steam Power Plants Based on Oxy-Combustion with Cryogenic or Membrane Based Air Separation

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Abstract

When realizing CSS steam power plants based on oxy-combustion, the energy demand for oxygen production is one of the main causes for efficiency losses. This comparative study focuses on the impact of the air separation technology - cryogenic as well as high temperature membrane based - on the efficiency of a coal-fired oxyfuel steam power plant. As a result of this study both show comparable efficiency potentials whereas the membrane based technology needs a higher degree of integration into the power cycle to compete efficiencies of the oxyfuel power plant with cryogenic ASU.

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1. Introduction

To recover and store carbon dioxide from flue gases of fossil fuel power plants, processes based on oxy-combustion appear to be promising. Concept of the technology is the combustion with commercially pure oxygen to achieve high CO₂ concentrations in the flue gases for the final CO₂ separation. The required oxygen is supplied by an air separation unit where the nitrogen is separated from the air. A great portion of the flue gases has to be recycled to substitute the removed nitrogen. This measure is inevitable to maintain the temperature level in the combustion chamber and in particular not to increase the heat transferred to the membrane walls of the steam generator which is limited by material parameters.

The major part of the efficiency losses within the CO₂ capture process chain of this technology is caused by the oxygen supply for the combustion. According to today's state-of-the-art, cryogenic air separation is the best available technology when realizing large-scale oxyfuel power plants. Since requirements are different compared to today's typical field of application, these cryogenic processes show a considerable potential for energy savings. In contrast, oxygen supply based on high temperature membranes is often suggested as a future option [1, 2].

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This article outlines the impact of the oxygen supply by cryogenic air separation units (C-ASU) and by high temperature membrane air separation units (HTM-ASU) on net efficiency of the oxyfuel steam power plant. The hard coal fired reference power plant North Rhine-Westphalia (RPP-NRW, net efficiency 45.9 %, [3]) provides a basis for this study. Ebsilon Professional[®] is used for the overall process simulations whereas the separation equipment of both air separation technologies, the flue gas treatment system as well as the CO₂ conditioning unit are modeled as black-box models.

2. Oxygen supply by air separation

When separating oxygen from air, purity of the products is of particular importance. Affecting the oxygen consuming process, the quantity of impurities such as e. g. nitrogen and argon is qualified by the purity of the oxygen x_{O_2} . On the other hand impurities of oxygen in the nitrogen by-product causes losses of the target product oxygen.

In this context, the oxygen recovery rate R is usually defined as the ratio of the oxygen mole flow rate in the oxygen product exiting the ASU to that entering the unit with the air to be processed. An oxygen recovery rate of 100 % would imply a totally oxygen free nitrogen product whereas a recovery rate of for instance 50 % results in a doubled air flow rate for the same amount of oxygen produced.

2.1. Cryogenic air separation units

The central unit operation of the cryogenic air separation process is the low temperature multistage distillation which is typically realized by means of a double-column system. For comprehensive process descriptions reference can be found at [4, 5].

The energy consumption of C-ASUs mainly consists of the power requirement for the compression of the feed air. Thermal energy, which is utilized to regenerate molecular sieves, adds roughly 10 % to the energy demand of the unit. Before air enters the cold parts of the C-ASU these molecular sieves remove components from the air (H₂O, CO₂, hydrocarbons etc.) that would interfere with the cryogenic process. Typical state-of-the-art C-ASUs produce oxygen with a purity of 99.5 % by volume at an oxygen recovery rate of 97.85 % [6]. Delivering the oxygen at approximate atmospheric pressure, these ASU show a specific power consumption of approximately 0.25 kWh² per kg of pure gaseous oxygen contained in the oxygen product. This specific power demand must not be taken as a fixed value – it is depending on both the chosen compression process (i.e. type of compressor, interstage cooling, unit size etc.) and other internal parameters of the ASU process. The outlet pressure, for instance, is influenced by the thermal interconnection of the double-column system besides equipment pressure losses. It can be decreased by decreasing the oxygen purity or lowering the temperature difference of the interconnecting heat exchanger (already in the range of 1 – 3 K) of the double-column system.

Adapting the C-ASU for the application in oxyfuel steam power plants, decreasing the purity of the oxygen product seems to offer the highest potential to lower its energy demand. Concurrently, it has to be considered that the energy demand of the CO₂ conditioning unit is increasing with a growing level of impurities. For lower product purities, optimized air separation processes offer additional opportunities for further reduction in energy demand (compare e.g. [7]). In [8] Air Liquide specifies a specific power demand of 0.2 kWh per kg with a future saving prospect to 0.16 kWh per kg for an optimized cryogenic air separation process with an oxygen product purity of 95 % by volume.

Possible options for thermal integration of C-ASU into oxyfuel steam power plant are limited to the recovery of the heat of compression due to the highly integrated cryogenic process design. With interstage cooling intentionally omitted the waste heat temperature level rises, accepting an increased specific power demand for compression. Feasible heat sinks within the oxyfuel power plant process are condensate or oxygen preheating.

² Calculated value. As far back as 1985, Linde specifies total specific energy consumption (including pre-purification unit) of 0.26 kWh/kg O₂ in [4].

2.2. High temperature membrane air separation units

Key component of the oxyfuel process with high temperature membrane air separation unit (HTM-ASU), which is in the stage of development, is a dense membrane made of ceramic materials. These materials begin to conduct oxygen ions above a material dependent temperature (usually above 700 °C). Driving force for the mass transport is the differential oxygen partial pressure across the membrane, while the oxygen flux is enhanced with decreasing membrane thickness and rising temperature. As only oxygen permeates the membrane, 100 % pure oxygen could be produced provided that air leakage within the membrane module is avoided. For further details regarding membrane materials, references [9, 10] are recommended.

The basic idea of the HTM-ASU, as illustrated schematically in figure 1, is the elevation of the oxygen partial pressure on the air side with an air compressor. The partial pressure difference across the membrane can be further enhanced by lowering the oxygen partial pressure on the oxygen receiving side of the membrane by sweeping with flue gas³, which contains only a small amount of oxygen.

As temperatures at the compressor outlet are not sufficient to activate the membrane material's conduction mechanism, the air needs to be preheated with counter current oxygen enriched flue gas. To recover parts of the spent energy for compression, the oxygen depleted air is expanded in a turbine. As the off-gas leaves the HTM-ASU at still elevated temperatures, the heat can be recovered in the power plant cycle. The energy demand of the HTM-ASU is determined by the required high temperature heat. In addition, mechanical driving power is needed or produced depending on the ASU process parameter design.

Regarding the process design, an analogy between the HTM-ASU and the externally fired gas turbine cycle can be drawn. According to the general behavior of gas turbine cycles, the maximum thermal efficiency is increasing with rising turbine inlet temperature while the optimal compressor outlet pressure is increasing at the same time. However, the thermal efficiency of the HTM-ASU is dependent on the oxygen recovery rate as the turbine receives a reduced mass flow for expansion. Consequently, the thermal efficiency is decreasing with rising oxygen recovery rates. For instance a decreasing oxygen recovery rate that approaches 0 % results in a maximized thermal efficiency of the ASU process but on the other hand the energy demand for the same amount of oxygen produced is growing ad infinitum. However, since the main task of the HTM-ASU is the oxygen supply, the maximization of the HTM-ASU's thermal efficiency is not of interest. In fact together with the power cycle an overall process optimization has to be conducted to find the optimum efficiency.

An important factor for the technical and economical realization of an oxyfuel steam power plant with HTM-ASU is the required membrane surface area that is increasing with lowered differential oxygen partial pressure (equivalent to increasing oxygen recovery rate and decreasing compressor outlet pressure).

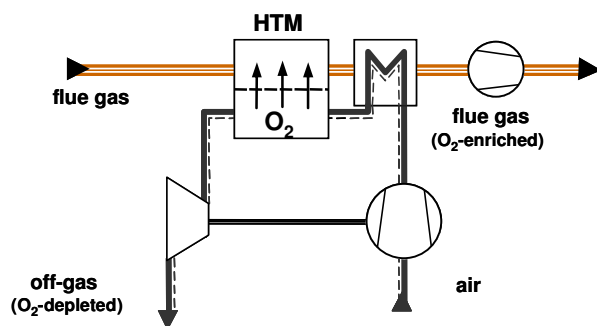


Figure 1: Scheme of an air separation unit based on high temperature membranes (exemplary flue gas swept).

³ It shows that materials enabling high permeation rates tend to be more sensible to flue gas components compared to those with inferior permeation rates [9]. Accordingly the high CO₂ concentrations of the oxyfuel flue gases can totally disrupt the oxygen permeation in some cases. Moreover sulfur components and alkali metal components can cause permanent chemical destruction of the atomic structure which is vital for the ion conduction.

As the specific energy demand per kilogram of pure oxygen produced by the HTM-ASU is heavily dependent on the process parameters and also on the economical boundary conditions, it is not possible to specify a general figure at this stage. Specific figures will be stated later in this paper for each studied case.

3. Integration of air separation into the oxyfuel power plant

For this study the concept of the RPP-NRW is adopted as the underlying base process, whereas the flue gas path is changed for oxyfuel combustion conditions. The integration of the air separation into the overall power generation process is the object of investigation in this study. Therefore the specific power demand for the CO₂ conditioning unit is assumed with 130 kWh_{el} per ton CO₂ separated, where a CO₂ separation rate of 90 % with a CO₂ purity greater than 95 % and a pipeline handover pressure of 100 bar is taken as a basis. Closer considerations to the issue of CO₂ separation processes for oxyfuel flue gases and their related energy demand can be found at [11]. For the simulation, comparable components are used with the same parameters (e.g. efficiencies of pumps, fans, electrical drives etc.). An air ingress rate referring to flue gas mass flow at the steam generator exit of 2 % is assumed.

3.1. Integration of the cryogenic air separation unit

The drive element for the air compressor can be realized either with electromotive drive or by means of a steam turbine. Within this study an electromotive drive was assumed. To keep the flow sheet design simple, the heats from air compression and from the CO₂ compression are not recovered and fully rejected (cf. fig. 2).

Concerning the boundary conditions of the firing system, the same adiabatic combustion temperatures as in the air based combustion of RPP-NRW (air/fuel ratio of 1.15) is being used [12]. Accordingly, a total flue gas recirculation rate of 67 % at a temperature of 370 °C is needed. The remaining flue gas is cooled down only to approx. 245 °C in order to avoid corrosion. This heat is transferred to a boiler feed water stream that is arranged in parallel with the high pressure preheating train. After a water quench, the flue gas is fed into the flue gas desulphurisation unit and is finally passed on to the CO₂ conditioning unit.

For an oxyfuel power plant, supplied by a cryogenic ASU consuming 0.2 kWh per kg O₂ with a purity of 95 %, a net efficiency of 36.4 % is determined. If the full potential of the low purity cryogenic air separation processes of 0.16 kWh per kg O₂ can be realized, the net efficiency rises by 1.1 % points. Employing a typical state-of-the-art ASU with a purity of 99.5 %⁴ and a related specific power demand of 0.24 kWh_{el} per kg O₂, a net efficiency of ca. 35.5 % is the result.

Further options for optimisation of the oxyfuel power plant with cryogenic ASU are oxygen preheating, advanced heat recovery of the flue gases or the recovery of the heat of compression (ASU or CO₂ conditioning unit).

3.2. Integration of the high temperature membrane air separation unit

Within this section the integration options of the HTM-ASU regarding the flue gas side are addressed at first. Then subsequently the impact of different heat recovery options of the ASU off-gas is shown for a selected example.

To operate the HTM-ASU, high temperature heat well above 700 °C has to be provided. In a steam power plant this heat can only be supplied by the flue gases. Various integration options are imaginable. In the concept of the Oxyfuel-AC process [1], the recycled flue gas serves two purposes simultaneously: to provide the required high temperature heat as well as to enhance the oxygen flux by sweeping the membrane (cf. figure 2). Approximately 70 % of the flue gas is fed to the HTM-ASU at operation temperature (700 – 1000 °C) and is recycled to the combustion chamber at temperatures of 500 – 700 °C after oxygen enrichment. This implies a high temperature gas cleaning unit for removal of dust and flue gas components that are incompatible to the membrane material as well as a recirculation fan designed to operate in the high temperature range of 500 – 700 °C.

⁴ Because of the corresponding lowered level of CO₂ impurities in this case a specific energy demand for the CO₂ conditioning unit is assumed of only 125 kWh per ton of CO₂ separated.

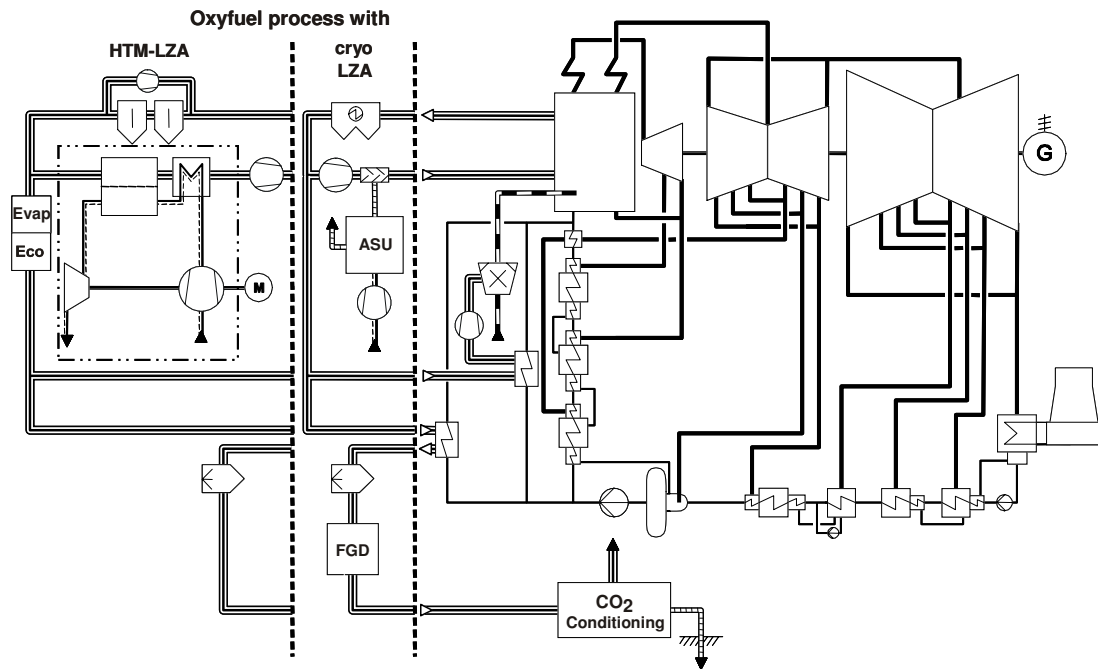


Figure 2: Process flow schemes of the studied oxyfuel power plant supplied by HTM-ASU and cryogenic ASU.

In a study of the Foster Wheeler Power Group [2] a process design is suggested where the double role of the flue gas (heating medium and sweep gas) is uncoupled in order to make use of a conventional flue gas treating system. The required heat to reach the membrane operating temperature is transferred to the compressed air by an additional convection type heating surface within the steam generator. Leaving the steam generator at approx. 370 °C, the flue gas is further cooled down and is then treated by a conventional flue gas treating system. The major part of the cleaned flue gases that has to be recycled to the combustion chamber, is reheated to be used as sweep gas for the HTM-ASU. Furthermore, the low temperatures of the cleaned flue gas allow for the deployment of conventional fans for the recirculation. The above mentioned uncoupling enables virtually freely adjustable oxygen concentrations in the mixture before combustion. With the Oxyfuel-AC concept, this concentration is limited since the flue gas has to provide at least the heat demand of the HTM-ASU.

Depending on the membrane material used, CO₂ for instance can cause the breakdown of the oxygen flux. Obviously, in that case, the flue gas sweeping is not advantageous. By applying the so called dead-end flow scheme to the membrane module, the oxygen produced is 100 % pure (at the cost of lower driving forces). Afterwards it is mixed with the recycled flue gas stream. With the same membrane parameters this option results in a lower power demand as the pressure drop of the membrane module can be avoided for the flue gas stream.

For the following statements regarding various off-gas heat recovery options, the flue gas integration of the Oxycoal-AC concept is being used (cf. figure 2). Similar to the process with oxygen supply by cryogenic ASU, the flue gas is beneficially cooled down to 245 °C with boiler feed water that runs parallel to the regenerative high pressure preheating train. The membrane module, being operated in counter current flow pattern, is described as a black-box model in the simulation.

To investigate the impact of changed membrane parameters, the membrane temperature starting from 850 °C is varied by ± 50 K as well as an oxygen recovery rate of 90 % is assumed and altered by ± 5 %. Being the driving force for the conduction of oxygen, the oxygen partial pressure ratio across the membrane substantially influences the required surface area. By adjusting the flue gas recycle rate and therefore the amount of sweep gas, the average oxygen pressure ratio is adapted. The average pressure ratio between membrane inlet and outlet $\bar{\pi}$ was assumed to

be 25 and changed subsequently by ± 2.5 . In addition, the outlet pressure of the air compressor is systematically varied to find the particular optimum of the overall net efficiency with the preceding parameters.

Without recovering any heat from the off-gas of the HTM-ASU, the efficiency is lower to that of the oxyfuel power plant with cryogenic ASU with 36.4 % (cf. figure 3 upper row). For the initial values of the membrane parameters ($T = 850\text{ }^{\circ}\text{C}$, $R = 90\%$, $\bar{\pi} = 25$), an optimal efficiency of approx. 35.1 % at an outlet pressure of 24.2 bar (absolute) is calculated (cf. boxed curves in figure 3 upper row). For a power plant with an electrical gross power output of 600 MW, 29.6 MW of driving power is needed to run the HTM-ASU. Accordingly a specific energy demand for oxygen supply of 0.48 kWh per kg O_2 of high temperature heat and 0.08 kWh per kg O_2 of electrical power is computed. Due to the high flue gas recirculation rate of 75 % overall at $580\text{ }^{\circ}\text{C}$, the recirculation fan requires an input power of 28.7 MW.

On the combustion-side, the oxygen content of the secondary gas is enriched to 25.7 % by volume (equal to 21.7 % by volume within the complete mixture before combustion). Compared to the unmodified RPP-NRW, the adiabatic combustion temperature is lowered by 200 K.

Following correlations are identified by the variation of the membrane parameters (cf. fig. 3 upper row):

1. Both, the decrease of the average oxygen partial pressure ratio as well as the decrease of oxygen recovery rate yield an increase in net efficiency. This effect is ascribable to the high power duty of the recirculation fan.

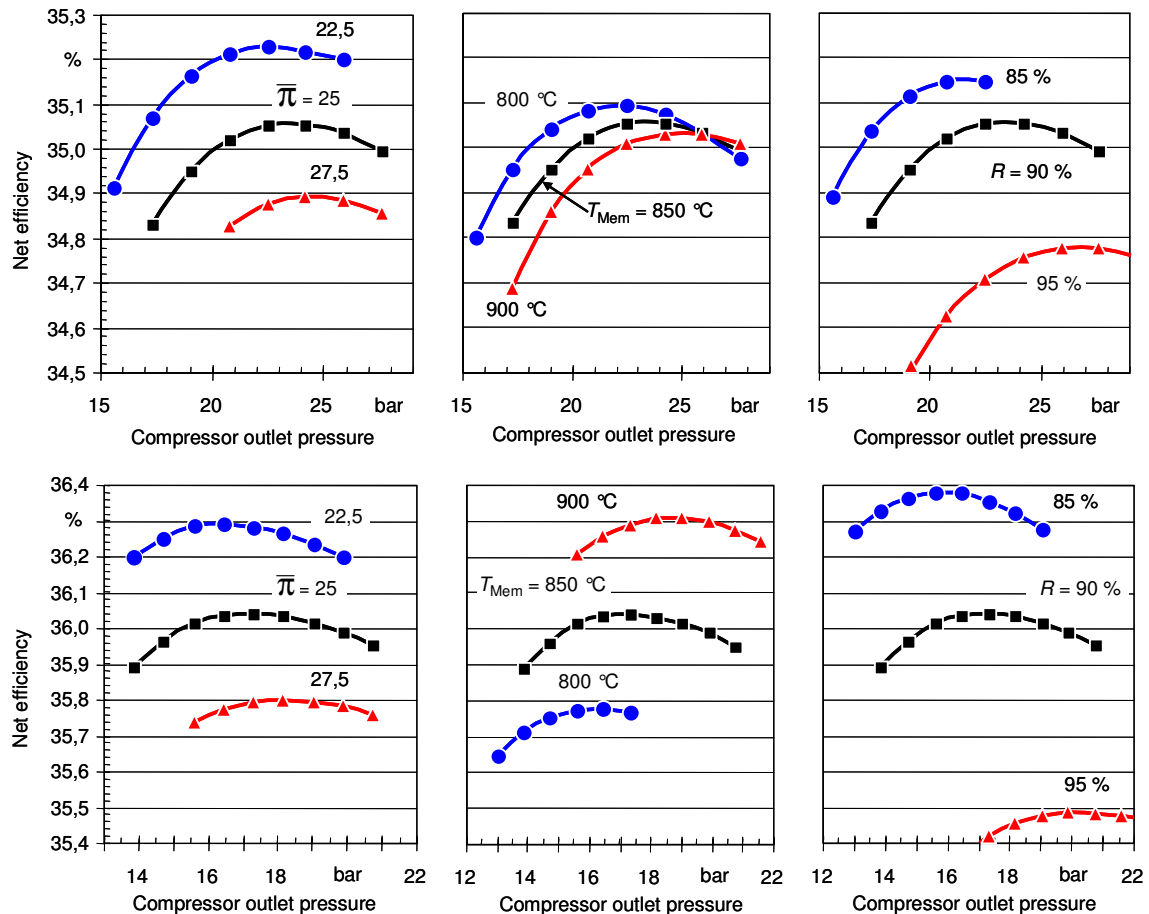


Figure 3: Net efficiencies of the examined oxyfuel process with HTM-ASU versus air compressor outlet pressure. Varied membrane parameters: average oxygen partial pressure ratio (left), membrane temperature (mid) and oxygen recovery rate (right). Top row: process without off-gas heat recovery. Bottom row: process with off-gas heat recovery in parallel with the high pressure boiler feed water preheat. Basic membrane parameters $T_{Mem} = 850\text{ }^{\circ}\text{C}$, $R = 90\%$, $\bar{\pi} = 25$ (boxed curves).

When transferring less oxygen from the air to the flue gas at constant air pressure a smaller amount of sweep gas is required to obtain the same π (cf. fig. 3 top right). Likewise less sweep gas is sufficient when π is reduced at constant air side conditions (cf. fig. 3 top left). Using less flue gas for sweeping results in reduced power duty of the recirculation fan. This again is affiliated on the one hand to the lowered mass flow and on the other hand to the decreased temperatures, as the required (marginally raised) heat duty to operate the HTM-ASU has to be supplied by this reduced flue gas mass flow. The latter is also associated with lowered temperature differences of the high temperature heat exchanger.

2. Increasing the membrane operating temperature without off-gas heat recovery induces a loss of efficiency (cf. fig 3 top row, centre) as the off-gas temperature is rising and thus the exhaust gas losses of the power plant are increased.

Preheating another portion of high pressure boiler feed water in parallel with the regenerative preheating train to cool down the off-gas to 245 °C (likewise the remaining flue gas) at basic membrane parameters results in a reduced exhaust loss. Therefore an enhanced optimal efficiency by 1 % point of 36.1 % is calculated (cf. boxed curves in figure 3 bottom row). The lowered optimal pressure of 17.3 bar is associated with a decreased driving power of 6.3 MW for the HTM-ASU. The specific energy consumption for oxygen supply is now 0.585 kWh thermal energy plus 0.02 kWh electrical power per kg of produced oxygen. The further increased recirculation rate of 81 % in total at 640 °C costs a considerably higher fan power duty of 43.9 MW. The firing system has to cope with an oxygen content of 17.7 % by volume in the secondary gas (equal to 15.9 % in the total mixture). A lowered adiabatic combustion temperature by 450 K compared with the unmodified RPP-NRW is associated.

Regarding the variation of the membrane parameters, the process shows the same qualitative behaviour as the process without off-gas heat recovery besides a higher sensibility (the curves are set wider apart). In opposition to the case without off-gas heat recovery a growth in membrane temperature now increases the net efficiency. The inversion of this correlation can be illustrated with the above mentioned analogy between the HTM-ASU and the gas turbine cycle. The heat recovered from the off-gas is displacing fractions of the steam turbine bleed streams for the regenerative feed water preheating and induces a competing combined cycle process parallel to the main steam cycle. Similarly to the gain in efficiency with increased turbine inlet temperatures of combined cycles, enhanced membrane temperatures show positive effects on the net efficiency.

As the off-gas is oxygen depleted air, no corrosion problems seem to occur with further heat recovery. Continuing off-gas cool down to 100 °C to exclusively preheat the condensate, a comparable efficiency as with the just described high pressure feed water preheat can be realized. However, the membrane is operated at a higher pressure of 20.7 bar. Finally a higher oxygen concentration of the secondary gas of 21.7 % by volume (18.8 % in the total mixture) as well as a more controllable combustion conditions can be achieved.

The highest efficiencies are obtained when employing a double-staged off-gas cooling at first to 245 °C by boiler feed water and finally to 100 °C for condensate preheating. By this means, the net efficiency can be increased by 0.7 % up to 36.8 %.

In principle, the mentioned dependencies regarding altered heat recovery options and in particular the membrane parameters are transferable to other high temperature heat integration concepts of the HTM-ASU. Especially the server influence of the hot end flue gas recirculation fan on the overall efficiency has to be regarded.

4. Conclusions

The analyses of the oxyfuel process with HTM-ASU indicate a characteristic dependency on the heat recovery of the off-gas leaving the expansion turbine. For overall process optimization, minimization of thermal and electrical energy demand to run the HTM-ASU is irrelevant. Moreover, a minimal degree of integration regarding the heat recovery is inevitable to compete with the net efficiency of the oxyfuel process supplied by a state-of-the-art cryogenic ASU (purity 99.5 % by volume) without any integration.

The latter shows a loss in net efficiency towards the RPP-NRW of 10.4 % points. However, the energy savings due to the application of cryogenic ASU supplying 95 % pure oxygen reduces this efficiency penalty to 9.5 % points. If the specified energy saving potential by means of further optimised air separation processes can actually be achieved in practice the efficiency drop can be minimized to 8.4 %.

In contrast the studied oxyfuel process with HTM-ASU shows an efficiency loss of 10.8 % without any off-gas heat recovery at the chosen base membrane parameters. This loss is reduced to 9.7 % when heat recovery is

implemented by exclusively either boiler feed water or condensate preheat. The latter process design option shows less challenging combustion conditions. The minimal efficiency drop of 9.1 % is calculated when applying a combined condensate and boiler feed water preheat. Nevertheless with increasing optimal pressure ratios of the air compressor at constant membrane parameters, combustion conditions are significantly tightened by decreasing the oxygen concentrations and hence reducing adiabatic combustion temperatures.

Summarizing, it can be stated that the efficiency losses of 8.4 to 10.4 % for the oxyfuel process with oxygen supply based on cryogenic ASU compared to 9.1 to 10.8 % for the oxyfuel process with HTM-ASU do not imply a significant difference concerning the process efficiency potential.

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