Integrated Low-Temperature CO_2 Capture from IGCC Power Plant by Partial Condensation and Separation of Syngas

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As an alternative to baseline technology a low-temperature syngas separation process has been proposed for pre-combustion CO_2 capture from coal-derived syngas for an IGCC plant. Through systematic process synthesis – sequencing of separators and location of compressors and expanders, followed by Pinch Analysis and energy targeting – a process design has been proposed and the net power consumption has been estimated. From a targeted power consumption of 21.9 MW the figure for the proposed design is increased to 22.9 MW mainly due to increased load for auxiliary refrigeration at low temperature. The power consumption of the proposed design represents further improvement from earlier designs that have already been shown to have higher energy efficiency than baseline technology.

1. Introduction

CO_2 capture by pre-combustion decarbonisation of syngas has potential of high energy efficiency, compact process units and highly competitive capture cost. Unlike post-combustion capture processes where CO_2 is captured from diluted flue gases at atmospheric pressure, pre-combustion capture conditions are characterised by higher CO_2 pressure and concentration. Since physical solvents such as Selexol and Rectisol are commercially proven technologies for CO_2 removal from syngas, these are considered to be baseline technologies for pre-combustion capture. As a promising technology alternative to physical solvents the present work presents a low-temperature CO_2 removal unit for IGCC application. Pre-combustion capture based on partial condensation of CO_2 and phase separation into a H_2-rich gaseous phase and CO_2-rich liquid phase has earlier been proposed by Brouwers and Kemenade (2010). A benchmarking by Berstad et al. (2013) showed the low-temperature capture process to be more efficient than Selexol in an IGGC power plant.

The process principles for the low-temperature CO_2 capture unit are mainly based on cooling, partial condensation and phase separation of the syngas, and are generally similar to those used in CO_2 separation from oxy-fuel flue gases (Soundararajan and Gundersen, 2012). Although a dedicated desulphurisation unit may be needed upstream of the capture, the syngas separation process is independent of the use of solvents. The syngas is required to be dehydrated before entering the low-temperature unit to prevent ice formation in heat exchangers. Subsequently the dry feed stream is compressed to specified separation pressure before cooling in a heat exchanger network against other process streams and auxiliary refrigeration. When the final separation temperature is reached, typically between 217 K and 220 K dependent on process specifications, the partially condensed gas is separated into a H_2-rich gaseous phase and a CO_2-rich liquid phase. In order to ensure high energy efficiency the process design requires effective heat integration between the hot syngas stream and cold H_2 and CO_2 product streams, as well as recovery of shaft power from expanders.

As shown in Berstad et al. (2013) the CO_2 capture rate (CCR) obtainable by low-temperature syngas separation is highly sensitive to syngas CO_2 concentration as well as separation pressure. Separation temperature, which is limited by the required margin between separation temperature and CO_2 freeze-out temperature, also impacts the obtainable CCR. The separation temperature constraint is, however, not a

critical issue as increased separation pressure can be used to compensate for restrictions on minimum separation temperature. Both Berstad et al. (2013) and Mori and Forsyth (2013) have indicated 85 % to be a practical upper limit with respect to CCR obtainable with low-temperature capture from IGCC.

2. Energy targeting and process design

As the low-temperature capture unit requires efficient heat integration Pinch Analysis is the obvious tool for targeting the energy requirement and initiating process design. However, classical Pinch Analysis has its well-known limitations such as the inability to take into account the degrees of freedom associated with changing the pressure of process streams between supply and target state. As stream pressure levels play a particularly strong role in low-temperature and cryogenic processes, the need for an extended methodology for process synthesis was pinpointed by Aspelund et al. (2007) who proposed the Extended Pinch Analysis and Design (ExPAnD) methodology including pressure-related aspects and targeting minimum irreversibilities in processes. The ExPAnD methodology takes on the degree of freedom in process design related to intermediate pressure and phase changes of process streams between supply and target state. For instance, by allowing process streams to act as cold utility streams in low-temperature processes, process paths for high-efficiency process-to-process heat integration can be obtained. Furthermore, changes in pressure, primarily through the use of compressors, pumps and expanders, require integration of shaft power in order to enable maximal utilisation of the thermomechanical exergy of process streams.

2.1 Boundary conditions

For the low-temperature capture unit, feed and target specifications for material streams are given in Table 1. As can be observed for the fuel and CO_2 product streams, target specifications are not very rigorously defined. The H_2-rich fuel should be of highest possible purity as this would be the obvious consequence of high CCR. However, as the gas turbine, assumed to operate with low-NO_X combustor, will require pre-mixed H_2 as feed, the fuel concentration of inerts such as nitrogen, CO_2 and argon is not a critical issue. For captured CO_2 the purity should also be as high as possible.

Table 1: Feed and target specifications for syngas feed after desulphurisation and dehydration, fuel product and captured CO_2

	Flowrate	T	P	Composition						
	kg/s	K	bar	CO_2	H_2O	CO	H_2	N_2	Ar	H_2S
Syngas feed	114.2	303.15	35.6	0.380	0.000	0.016	0.538	0.057	0.009	0.000
Fuel			25	Subject to CCR						
CO_2			150	As pure CO_2 as possible						

2.2 Process synthesis and the impact of selected pressure levels

As mentioned in the introductory section the CCR of low-temperature capture unit is sensitive to separation pressure. This is illustrated in Table 2 where the estimated CCR as well as product-stream data for three different pressure levels are shown. As can be observed, the separation pressure must be above 110 bar in order to obtain a CCR of 85 % for the current syngas composition.

The highest separation pressure in Table 2 is assumed to apply for the further analysis and process design. A consequence of performing the syngas separation at high pressure is the H_2-rich fuel product stream leaving the main separator at low temperature and an excessively high pressure level, as the target pressure for this stream is 25 bar. As emphasised by Aspelund et al. (2007) the pressure-based exergy of this process stream should to a highest possible degree be utilised for heat integration by process-to-process heat exchange through pressure change. Moreover, generation of useful shaft power and its integration back into the low-temperature process should be carried out for maximising overall efficiency. As expansions from 111.5 bar to about 25 bar, a pressure ratio of roughly 4.5, can be utilised for power and refrigeration, at least two expander stages should be employed in the process. A single-stage expansion across such high pressure ratios for the fuel gas with as low molecular weight as 8.9 will not be efficient. There are numerous options for the path of the H_2-rich fuel stream from its supply state after separation, to its target state. However, a direct expansion after leaving the main separator is not possible as CO_2 would freeze out immediately due to temperature drop below the freezing point. As further compression beyond 111.5 bar does not seem rational, the first step is thus to heat the stream in at least one heat exchanger before expansion.

Table 2: Product streams and CCR from the phase separators for different separation pressures

P separator bar	CCR %	Product streams	Flowrate kg/s	Composition				
				CO_2	CO	H_2	N_2	Ar
32.6	61.5	H_2-rich gas	56.1	0.191	0.021	0.702	0.074	0.012
		CO_2-rich liquid	63.5	0.987	0.001	0.007	0.004	0.001
		Recycle stream	5.4	0.867	0.012	0.072	0.037	0.012
		CO_2 after purification	58.1	0.999	0.001*			
60.3	77.6	H_2-rich gas	40.8	0.121	0.023	0.763	0.081	0.013
		CO_2-rich liquid	81.0	0.975	0.002	0.014	0.007	0.003
		Recycle stream	7.6	0.778	0.019	0.123	0.060	0.019
		CO_2 after purification	73.4	0.998	0.002*			
111.5	84.8	H_2-rich gas	33.9	0.085	0.023	0.794	0.084	0.013
		CO_2-rich liquid	90.1	0.954	0.004	0.026	0.012	0.004
		Recycle stream	9.8	0.676	0.026	0.188	0.084	0.026
		CO_2 after purification	80.3	0.997	0.003*			

* Cumulative fraction of CO_2, H_2, N_2 and Ar

For the liquid CO_2 stream leaving the bottom of the main separator, direct pumping to target pressure (150 bar) may seem like an energy-efficient solution due to the low compression power required by liquid pumping. Moreover, one of the heuristics by Aspelund et al. (2007) also suggests this: "A fluid with $P_s < P_t$ should be compressed in liquid phase if possible to save compressor work." However, there are also good reasons for not following the heuristics at this particular point. As can be observed in Table 2 the estimated concentration of H_2 in the CO_2 stream is 2.6 mol% and corresponds to a potentially energy loss rate of about 13 MW based on lower heating value (LHV). From the viewpoint of energy efficiency this rate of lost fuel represents a major penalty and a process step for the recovery of this fuel should thus be included in the process design. Hence, instead of using the heuristic rule for liquid pumping of CO_2 at the main separator liquid outlet, instead the S-layer of the diagram in Figure 1a is complemented with an additional H_2 recovery separator.

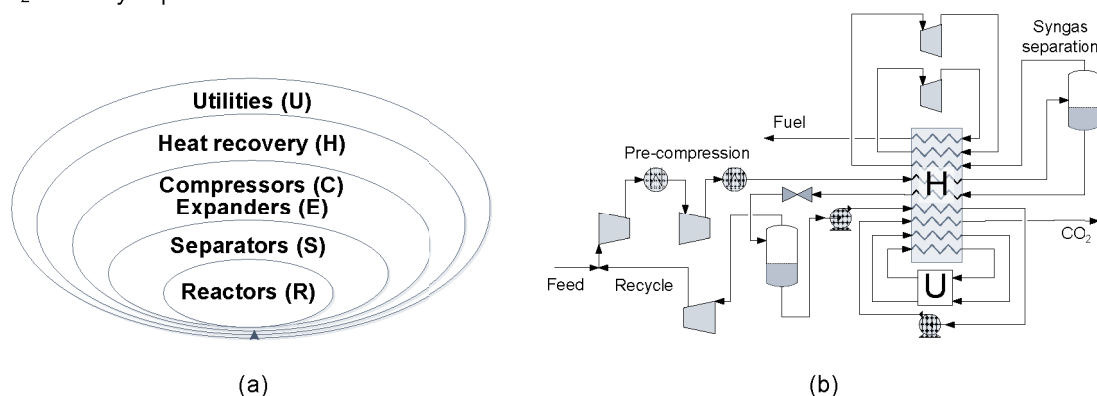(a)   (b)

Figure 1: (a) Process design hierarchy based on extended definition by Linnhoff et al. (1982), also known as the Onion diagram; (b) General process flow diagram for the low-temperature capture unit with S, C and E components before heat recovery system (S) and utilities (U) have been designed

H_2 recovery is carried out by throttling and flashing the liquid CO_2 stream. However, as is the case for the gaseous H_2-rich product stream, the liquid CO_2 cannot be expanded directly after separation due to CO_2 freeze-out. Instead, the stream first has to be heated prior to flashing. The new introduced intermediate target temperature before flashing represents another degree of freedom in the process design, as is the case for the separation pressure. The pre-flash reheat temperature has been specified to 231 K and the pressure for each case is in turn adjusted on order to obtain a flash separation temperature of 217 K, equivalent to the minimum allowed process-stream temperature in the capture unit. The resulting purification of CO_2 is considerable and the flash gas made up of mainly CO_2 and H_2 is recycled to the pre-compressor train. After this purification step the heuristic rule of liquid pumping is correctly followed by pumping the purified liquid CO_2 to 95 bar and further to 150 bar after heat integration.

With the selected process structure, process-stream paths and parameters the R, S and C&E layers from the 'onion' diagram in Figure 1a have been defined. The general process structure prior to designing layers

H and U is shown in Figure 1b. Although the core of the low-temperature CO_2 capture unit – separators, compressors and expanders – is defined, the degree of heat integration in the heat recovery system (H) and resulting utility (U) requirements will have significant impact on the overall energy requirement.

2.3 Energy targeting

With the selected process paths for syngas feed and separation product streams the resulting composite curves for heat integration have been plotted in Figure 2a. As can be observed the low-temperature capture unit is not self-contained with respect to cooling duty as a net cooling requirement of about 10.2 MW has been identified. Although a self-contained process could be obtained by recycling and re-expanding captured CO_2 in a semi-closed auto-refrigeration loop as is commonly done for oxy-fuel CO_2 processing units (White et al., 2010), the current process design assumes liquid pumping of CO_2 to transport pressure and otherwise no recycling of CO_2 apart from the CO_2- and H_2-rich recycle stream from the flash separator. On the one hand this CO_2 process path eliminates the requirement for a CO_2 compressor train but on the other, auxiliary refrigeration is required due to the cold-side duty deficit.

The process pinch, specified to 3 K temperature approach, is located around the point of CO_2 condensation, where the single hot syngas stream "sees" four different cold process streams: pressurised cold CO_2, high-pressure fuel, intermediate-pressure fuel and low-pressure fuel. Above pinch the number cold streams is reduced from four to two, as only the low-pressure fuel and liquid CO_2 streams are heated above pinch. Below pinch the four cold streams contribute down to about 220.5 K while the high-pressure fuel stream contributes to 217 K.
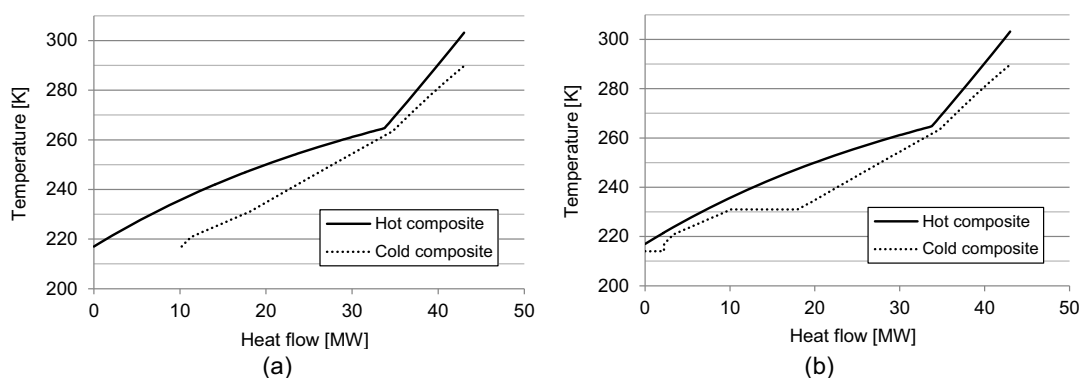

(a) (b)

Figure 2: (a) Composite Curves for syngas cooling (Hot Composite) and heating of separation product streams (Cold Composite) based on specified process paths; (b) Energy targeting Composite Curves for the low-temperature CO_2 capture unit including propane and ethane auxiliary refrigeration

As assumed in earlier works by the authors (Berstad et al., 2013), a propane-ethane cascade refrigeration cycle as shown in Figure 3a provides auxiliary cooling in order to meet the net cold duty in the capture unit. In this work the refrigeration cycle is assumed to be a two-stage propane cycle with open intercooler in cascade configuration with a single-stage ethane cycle. Analogously to meeting heating requirements with isothermal condensation of steam different pressure levels in high-temperature processes, the cooling duties in the capture unit can be provided by isothermal evaporation of propane and ethane at different pressures and temperatures. The selection of pressure levels for the auxiliary refrigeration cycle is in principle subject to optimisation but current pressure levels have been based on practical considerations. In order to ensure a cold-end Pinch of 3 K the ethane evaporation temperature is specified to 214 K, corresponding to an ethane low-pressure level of 3.92 bar. Furthermore, a propane low-pressure level of 1 atm has been chosen, corresponding to an evaporation temperature of 231 K. The high-pressure level for propane is set to 12.1 bar based on available intercooler outlet temperature (303 K) and required sub-cooling (3 K), and the intermediate pressure is set to 3.46 bar to ensure relatively even pressure ratio distribution between the compressors. As the low-pressure propane evaporator must be heat-integrated with the ethane condenser in the cascade configuration, the selected temperature difference between the condensing and evaporating liquids determines the ethane high-pressure level. This difference is specified to 5 K, resulting in an ethane condensation temperature and pressure of 236 K and 8.54 bar.With all process stream compressions and expansions specified the overall energy targeting now depends on the process-to-process and auxiliaries-to-process heat integration and resulting energy penalties. Regarding auxiliary integration, the ethane and propane evaporators are assumed to provide the cooling duty at 214 K and 231 K, respectively. For this configuration, composite curves are plotted in Figure 2b and the

overall energy results are given in Table 3. Based on the energy targeting the potential minimum auxiliary consumption is now targeted to be 2.24 MW and 7.94 MW at 214 K and 231 K. With the given auxiliary refrigeration process the net power consumption target calculates to 5.29 MW.

2.4 Process design

Although the target power consumption has been identified, the final results will depend on the selected heat integration network, in other words the efficiency of thermal energy integration, as well as the efficiency of power integration. As can be observed in Table 3 the total shaft power from the two fuel expanders is about 9 MW in the energy targeting case and the integration of this power is vital for the overall efficiency. There are two main options for integration of the generated shaft power, either by conversion to electric power for driving compressors in the process or by direct power integration with the pre-compression train. The first option has been arbitrarily chosen in this work and is assumed to operate with an overall transmission efficiency of 90 %.
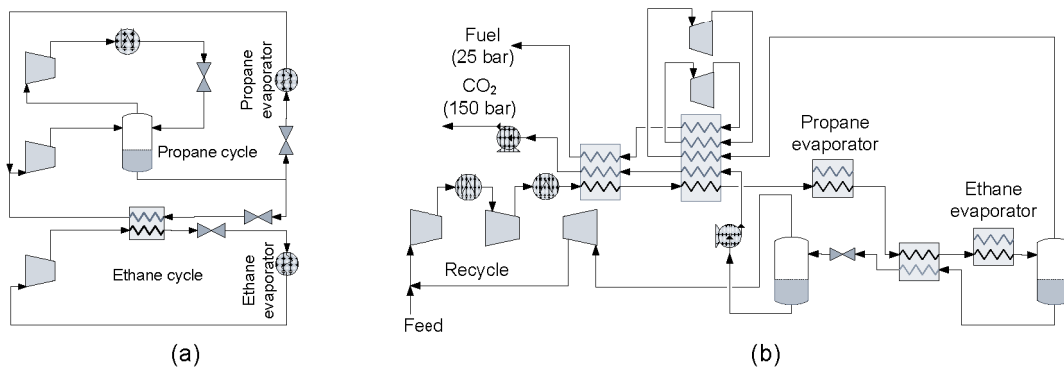

<div align="center">(a)</div> <div align="center">(b)</div>

Figure 3: (a) Structure of the propane-ethane auxiliary refrigeration cycle; (b) Process flow diagram for the proposed process design

The flow diagram for the proposed process design is shown in Figure 3b and the resulting heat transfer diagram is plotted in Figure 4. Comparing this heat transfer diagram with the composite curves in Figure 2b it can be observed that the heat integration and use of auxiliary refrigeration in the proposed design differ from that assumed in the energy targeting. Whereas the total cooling duty for the propane evaporator is decreased from 7.94 to 4.03 MW, the use of ethane is increased from 2.24 MW to 6.59 MW. Instead of using propane as indicated in the energy targeting, a five-stream heat exchanger with three fuel streams and CO_2 on the cold side is proposed to provide the entire cooling duty in the range 240–270 K (hot-side temperature) in order to reduce the number of required heat exchangers. Hence, propane will only contribute to cooling of syngas from 240 K to 234 K as well as for ethane condensation. As a result the total auxiliary cooling duty has increased from a targeted 10.2 MW to 10.6 MW and as can be observed in Table 3, the power consumption for refrigeration has increased from 5.29 to 6.40 MW due to the shifted duties of auxiliary propane and ethane refrigeration.
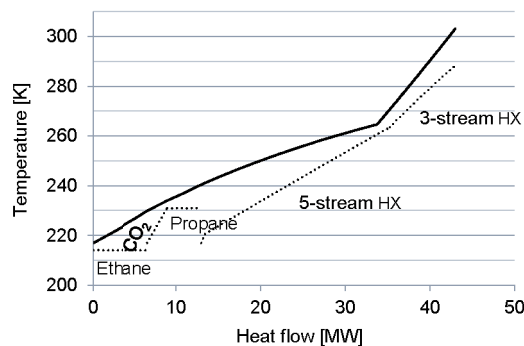

Figure 4: Heat transfer diagram for the proposed process design

2.5 Overall results

Power figures given in Table 3 are results of process design as well as a set of process assumptions such as the efficiency of compressors and expanders, pressure drop in intercoolers and heat exchangers, ambient temperature and other important parameters. The assumptions for machinery efficiencies are mostly equal to those assumed in Berstad et al. (2013) and considered to be well within realistic bounds. Regarding pressure drop 0.5 bar is assumed for all intercoolers, 1 bar for CO_2 per heat exchanger and 0.5 bar for other streams per heat exchanger. Cooling water pumping power is assumed to be 5 kW_{el} per MW of intercooler heat rejected. As mentioned in the introduction, Berstad et al. (2013) found the low-temperature capture to have higher energy efficiency than baseline technology for 85 % capture-unit CCR, with 0.8 %-points improvement in net electric efficiency relative to Selexol. The overall power requirement for the capture unit was about 24 MW and with the current process design this has been reduced to about 23 MW. This implies a further reduction in electric efficiency penalty by about 0.1 %-points, mainly through reducing the auxiliary refrigeration power consumption by adding an intermediate propane pressure level.

Table 3: Power figures [MW] for energy targeting figures and proposed process design

	Syngas compr.	Recycle compr.	CO_2 pumps	Auxiliary refr.	Fuel exp.	CW pumps	Net power
Energy targeting	22.1	1.12	1.41	5.29	9.17	0.21	21.9
Proposed design	22.1	1.12	1.40	6.40	9.25	0.22	22.9

3. Conclusions

Pinch analysis and energy targeting for a low-temperature CO_2 capture unit for IGCC has been performed. From the energy targeting a process design including heat exchanger network and auxiliary refrigeration has been proposed and the overall power consumption has been estimated. From the initially targeted power consumption of 21.9 MW the resulting figure for the proposed design is increased to 22.9 MW mainly due to increased load for auxiliary refrigeration at low temperature. The power consumption of the proposed design represents a further improvement from earlier designs, which have already been shown to be more energy efficient than baseline technology. On-going and further work includes design of central process units such as multi-stream plate-fin heat exchangers, auxiliary refrigeration utilities, compressors and expanders. Moreover, operational aspects such as off-design operation and control will be addressed.

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