ALTERNATIVE COOLING SYSTEM FOR CO$_2$ CAPTURE TECHNOLOGY

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A Thesis submitted in partial fulfilment of the requirements for the degree of
Master of Engineering in Mechanical Engineering

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2015
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ABSTRACT

A new CO₂ capture technology for the IGCC power plant of 500MW capacity was proposed by Surovtseva, Amin and Barifcani in the year 2011. This technology required a cooling system to cool the flue gas from 35°C to -55°C for effective carbon dioxide capture. The initial work estimated that the energy required for cooling will be in the order of 13 MW through basic thermodynamic analysis, but the detailed analysis of the refrigeration system was not covered. The cooling system for this technology was further analysed and improved by Qi Tian (CDU Student-2014-2015). But it required energy in the order of 50MW after heat recovery for operation. This made the capture technology an energy intensive one as it consumed 10% of the plant capacity when compared to 5-7% for the existing technologies. The objective of this thesis is to research for multiple alternatives for the cooling system and to design an effective cooling system whose energy requirement is in the order of 13 MW as estimated by the initial works.

The first alternative involved integration of a conventional JT based cooling cycle with a multistage heat exchanger system. For this case the energy required for achieving the cooling was found to be in the order of 130MW. This approach did not perform as expected because of the high influence of negative JT coefficient of Hydrogen in the flue gas.

The second alternative researched was redesigning the cooler, used after the heat recovery system, with optimum refrigerant combinations at the operating conditions and with different refrigeration cycles. This also did not lead to the expected result as the energy consumption by the system was in the order of 60 MW to 80MW.

The third alternative researched was the sub-cooling of the separated CO₂, which exits the heat recovery system, in a JT valve and using it as a cold source to further reduce the feed gas temperature. This further reduced the cooling load on the final stage chiller substantially and brought down the power consumption to 11.7MW which is even lesser when compared to initial prediction of 13MW thus arriving the thesis objective. The additional heat transfer units were designed as another outcome of this thesis.
INTRODUCTION

The threat of climate change is considered to be one of the major environmental issues in contemporary science and engineering. Combustion of fossil fuels produces large amount of CO₂ across many industries today. It is the main source of carbon dioxide emissions and it accounts for 95% of total annual CO₂ emissions (Chen, et al., 2013). An economically viable CO₂ capturing technology is therefore deemed essential to any power plant utilising fossil fuels. Numerous studies show that in the case of Integrated Gasification Combined Cycle (IGCC) power plants, carbon capture can be a viable option given an appropriate sustainable technology is developed (David, 2001).

One system using cryogenic techniques was proposed by Surovtseva, Amin and Barifcani in the year 2011, which can be applied to IGCC power plants and other industrial gasification facilities. Initial laboratory testing proved that sufficiently low concentration of CO₂ can be achieved through this method; however, the energy requirements have been estimated only roughly, and no detailed analysis has been conducted for refrigeration system. A cooling system for the above CO₂ capture technique was proposed by Ms Qi Tian, a former student from CDU (2014). But after the performance analysis of the proposed system, it was found that the energy requirement for the cooling system was very high making it economically unviable. Alternatives for the proposed design are being researched in this thesis with an aim to design a cooling system with lower energy consumption. This would aid in the implementation of the CO₂ capture technology on large scale and hence reduce their emissions into the atmosphere and hence positively impacting the environment.
LITERATURE REVIEW

This literature review was done to understand the CO₂ capture technology that is being studied here and the energy benchmarks of other CO₂ capture technologies that are available. It is also made to understand the current cooling system and the other cryogenic cooling system in the market which would enable us to develop a low energy consuming cooling system for the CO₂ capture technology under study here. Literatures on performance improvements of the auxiliary devices are also studied which would be helpful in improving the developed cooling system in the later stage.

Review of existing CO₂ capture technologies

Capture of CO₂ from fossil fuel burning power plants is gaining importance in these days as an effective greenhouse gas control technology. A number of processes for pre- and post-combustion CO₂ capture from modern power plants such as IGCC are currently under investigation.

Chen et. al studied the absorption rate of CO₂ using different absorbents like Selexol, Rectisol and water and found that out of these three Rectisol is having 15% higher absorption rate when compared to water and 10% higher absorption rate when compared to Selexol (Chen, et al., 2013). Membrane separation technology is another CO₂ capture technology which is getting widespread because of their large scale implementation capacity (Luis, et al., 2012).

Amine based CO₂ capture technology is being widely researched and developed. Anusha Kothandaraman, a research scholar of Massachusetts Institute of Technology, did her thesis on the performance of different solvents that can be used for carbon capture and sequestration through absorption method. Three commercially used solvents Monoethanolamine, Potassium carbonate and chilled Ammonia were compared, using a framework designed through ASPEN plus. It was concluded that Monoethanolamine is the best solvent for the current existing carbon capture plants and potassium carbonate can be used for the future plants for better performance. It was also found that the energy requirement for CO₂ capture was is in the range of 0.01572 KWh/gmol CO₂ for Monoethanolamine based and 0.021 KWh/gmol for ammonia based absorption systems (Kothandaraman, 2010). This research gave better idea of the energy requirements for the chemical absorption techniques that are already commercialized.
Rao and Rubin in their research found that implementing a CO$_2$ capture plant for an existing power plant would be cost intensive when compared to the cost of implementation for a new power plant basically because of the less efficient heat integration (Rao & Rubin, 2002).

Overall performance and cost of carbon per mass CO$_2$ abated including the cost of CO$_2$ compression for transportation are quite well documented for most of these and summarised in Table 1 for coal-fed facilities (Rao & Rubin, 2002).

<table>
<thead>
<tr>
<th>Technology</th>
<th>US$/ton CO$_2$</th>
<th>In terms of % of plant capacity</th>
</tr>
</thead>
<tbody>
<tr>
<td>IGCC (inc. GE, Shell and ConocoPhillips)</td>
<td>18-42</td>
<td>15%</td>
</tr>
<tr>
<td>Pre-combustion capture by amines</td>
<td>29-62</td>
<td>25%</td>
</tr>
<tr>
<td>Post-combustion capture by amines</td>
<td>40-74</td>
<td>19%</td>
</tr>
<tr>
<td>Oxy-combustion capture</td>
<td>35-72</td>
<td></td>
</tr>
</tbody>
</table>

In all cases the associated capital cost increases by 30-40% when carbon capture is implemented, and the overall efficiency of the plant reduces by as much as 10%.

**CO$_2$ capture through Cryogenic Condensation and Hydrate Formation**

A carbon dioxide capture system using Cryogenic condensation and hydrate formation technique was developed by Surovtseva et al. A simplified process diagram of the developed system is shown below in figure 1.

![Figure 1: Flow process of CO2 absorption cycle (Surovtseva et al., 2011)](image)

The composition of the feed gas is shown in the figure 2 below.
Flue gas at 55-60 bar passes through a cooling system, as shown in figure 3, to achieve -55°C, at which most of CO₂ is separated. The remaining overhead gas enriched in hydrogen is then directed into a hydrate reactor, where the remaining CO₂ is absorbed by an aqueous solution to form hydrate slurry. The condensed CO₂ is utilised to pre-cool the feed gas as shown in figure 2, and it was suggested that up to 80% of cooling can be achieved through this heat recovery system (Surovtseva, et al., 2011). But according to the initial evaluation, the final stage chiller E105 would require energy in the order of 13MW for a 500 MW plant utilizing IGCC combustion technique with 38% overall efficiency (Surovtseva, et al., 2011). But the details of the cooling system including the analysis were not discussed.

**Figure 2: IGCC-GE Flue gas composition**

<table>
<thead>
<tr>
<th>Component</th>
<th>Mole Fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Methane</td>
<td>0.02%</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>3.50%</td>
</tr>
<tr>
<td>CO₂</td>
<td>40.30%</td>
</tr>
<tr>
<td>H₂S</td>
<td>0.22%</td>
</tr>
<tr>
<td>Hydrogen</td>
<td>55.14%</td>
</tr>
<tr>
<td>Argon</td>
<td>0.79%</td>
</tr>
</tbody>
</table>
Qi Tian redesigned and optimized this cooling system and the details are shown in figure 4. The goal of this research was to design and optimise the cooling system for the lowest possible input energy to the final stage cooler. This method also consisted of the input flue gases being precooled using the cold liquid CO₂ which is exiting the system (Tian, 2014). All heat exchange devices included in this scheme (Fig 4) were appropriately sized using UNISIM simulation software. Peng-Robinson equations of state were used to evaluate fluid properties, to ensure consistency with the original research.
Considering the arrangement shown in Figure 3, energy efficiency is directly related to the energy required by the last cooler of the system E105. After the heat recovery system consisting of four heat exchangers (E101, E102, E103, E104) was optimised, the energy required for the last cooler was found to be in the order of 50MW for a feed flow rate of 500cc/min. Table 2 summarises the findings in comparison with the original predictions (Surovtsev et al., 2011).

Table 2: Energy Performance Comparison (Tian, 2014)

<table>
<thead>
<tr>
<th></th>
<th>IGCC GE, ( \text{H}_2/\text{CO}_2 = 1.37 )</th>
<th>IGCC Shell, ( \text{H}_2/\text{CO}_2 = 1.53 )</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Result from Original Research</td>
<td>Optimised system</td>
</tr>
<tr>
<td>Cooling load on E105 (Fig 4), MW</td>
<td>13.7</td>
<td>49.6</td>
</tr>
<tr>
<td>Energy recovery, % total load</td>
<td>82.6</td>
<td>36</td>
</tr>
</tbody>
</table>

It has to be noted that the energy requirement is very high when compared to proposed requirement. This is because; the proposed design could recover only 35% of energy when
compared to 80% in the initial research. Hence the energy requirement is high (10% of plant capacity) making the CO$_2$ capture technology, economically unviable.

Two IGCC configurations have been investigated, GE and Shell, flue gas in each consisting primarily of Hydrogen and CO2. It has to be noted here that in Shell configuration, the flue gas is supplied at substantially lower pressure than required for Cryogenic Condensation and Hydrate capture method, and therefore additional capital and operational expenditure is required to compress this stream (Surovtseva, et al., 2011). This cost was not included in estimations presented in Table 2.

This thesis therefore aims to achieve lower energy consumption (less than 13 MW proposed in the initial research) by optimising the cooling system. The various options for the cooling system are discussed in the upcoming sections.

**Cryogenic Cooling Techniques**

There are number of systems which use Joule Thompson effect to create cryogenic temperature. Joule Thompson effect describes the temperature change of a gas when is adiabatically forced through a valve or other pressure reducing devices. The amount of cooling obtained for a given pressure drop of any gas is given using the Joule Thompson Coefficient ($\mu_{JT}$) which is given by

$$\mu_{JT} = \left(\frac{\partial T}{\partial P}\right)_H$$  \hspace{1cm} (Ravishankar, et al., 2014)

Where $\partial T$ is the isenthalpic temperature drop achieved for the pressure drop $\partial P$ at the temperature $T$(Ardhapurkar, Sridharan, & Atrey, 2014). Thus a positive JT coefficient means a reduction in temperature with increased reduction in pressure. The JT coefficient becomes negative above a particular temperature called the inversion temperature. Hence after this temperature the temperature starts increasing for increased changes in pressure which is not desirable in case of cooling. CO$_2$ from the capture system can be considered to be used as a refrigerant because of its high JT coefficient.

Extensive works were done in the field of cryogenic cooling system by in the later 1980’s. One such system, which is similar to the system, proposed here, had compressor in series with a precooling system and a Joule Thompson valve as shown in the figure below originally patented by Ball Corporation in 1988(Benedict, Lester, & Linenberger, 1989).
The system designed was capable of achieving a temperature in the range of 60K to cool the load shown here. The working gas here was nitrogen and it was shown that precooling the gas using the cold gas exiting the load reduced the power consumption by 4.5 times (Benedict et al., 1989).

Valencia experimented on the CO\textsubscript{2} and H\textsubscript{2}S separation from the natural gas and encountered problem because of CO\textsubscript{2} freezing. The problem was addressed using a technology called controlled freezing zone (CFZ) which was developed by him. (Northrop & Valencia, 2009) This technology can be implemented in our system when we face any problems while handling CO\textsubscript{2} in solid form. The other main factor discussed here was to avoid water vapour in the stream to avoid problems related to blocking due to ice.

Clodic et. al discussed thermal swing process that freezes CO\textsubscript{2} on the surface of heat exchangers. Fouling of heat exchanger is one major problem faced by them during their research (Clodic D, 2005). Parallel heat exchangers were used to sort this issue. This method can be used in this thesis; we can operate parallel heat exchangers to operate the process. When one heat exchanger is cooling the other would be heated to remove the CO\textsubscript{2} from surface which causes fouling.

A method of expanding CO\textsubscript{2} in an expander causing freezing and avoiding surface fouling was experimented by Castrogiovanni et. al. This method eliminated the fouling problems of heat exchanger. (Castrogiovanni, Balepin, Robertson, & Calayag, 2012). They also used the exit CO\textsubscript{2} particles to cool the incoming flue gas. This point is very relevant to this thesis as the exit CO\textsubscript{2} is at very low temperature and it can be used to cool the inlet gas stream.
**Vapour Compression Refrigeration Cycle**

The objective of a vapour compression cycle is to remove the heat from a cold space and transfer it to a hot space. It is also more efficient when compared to a system of heat exchangers when we are operating at higher temperature differences (Rao, 2008). It is achieved through a refrigerant which performs that function at different pressure and temperature stages. The vapour compression cycle is shown below.

![Image of Vapour Compression Refrigeration Cycle](image)

**Figure 6: Vapour Compression Refrigeration Cycle (Rao, 2008)**

Refrigerant which is partially saturated (4) enters the evaporator which absorbs heat and makes the refrigerant completely saturated (1). The saturated vapour is compressed to a high pressure vapour (2). This high pressure vapour is cooled down and condensed to saturated liquid at the same pressure in a condenser (3). The high pressure liquid is expanded into a partially saturated liquid (1) at low pressure through expansion and the cycle is carried out again. The TS diagram is also shown in the figure. In ideal cycle, it is assumed that the compression process is isentropic. The input work for the whole cycle is $W_{in}$ which is the work supplied to the compressor. It is the work required to transfer heat from a low temperature source to a relatively high temperature surroundings.
Cascade Refrigeration Systems

Cascade refrigeration systems consist of two Vapour compression cycles coupled to each other. The condenser of lower cycle acts as the evaporator for the cycle above. The overall cycle is shown in the figure below.

![Cascade Refrigeration Cycles](image)

Thus it can be seen that we can use different refrigerants for different stages. There are temperature ranges that each refrigerant can operate. This overcomes the temperature range that we can operate using conventional VCR cycle as we use multiple refrigerants with different temperature ranges to suit the application. The energy penalty for this cycle is less when compared to the single vapour compression cycle (IIT Kharagpur, 2013).

Suitability of the Cycles

Castillo et al. analysed and explored the advantages and disadvantages of the different precooling stages of the known LNG processes. In these processes the natural gases are cooled using the processes such as pre-cooling, liquefaction and sub-cooling to temperature as low as -161° C. The C3MR process analysed here pre-cools LNG using a multistage refrigeration cycle at different pressure levels and at -35 °C. Liquefaction-sub cooling stages uses refrigerant in multiple phases. Flash separators were used to separate the vapour and liquid states. The liquid phase is used to further cool down LNG to the desired temperature. The two phases are further mixed and compressed and condensed to repeat the cycle. A part of the propane, from the pre-cooling stage, act as the condenser for this process at different pressure levels.
While working with the concept of cascade refrigeration, “temperature cross” was the error message that was received repeatedly. On analysis, it was found that temperature cross occurred because there was both sensible and latent heat exchange occurring at the heat exchanger. This made it difficult for the software to handle. One method of solving it is having different heat exchangers for different phases for computation. Other method was to make sure that there is only completely sensible heat transfer or latent heat transfer happening in the heat exchanger. (W.R, 2007)

The Mixed fluid cascade process is a three stage cooling process with the refrigerant from each stage cooling the natural gas stream. A part of the refrigerant from each stage is used to cool the next stage(s) as well. Each cooling stage is further cooled down by its own throttled stage being compressed.

Both the types of systems were compared and it was found that C3MR process has lower power consumption for the same operating temperatures when compared to the other cycle in a warm environment. It also has almost similar cooling load in each cycle which is operation wise better because of less or similar inventories (L, et al., 2013). Based on this we can apply multistage cascade refrigeration over single stage refrigeration for cooling the feed gas in this thesis as minimum power consumption is our main objective.

Castillo and Dorao compared mixed refrigerant cycles and pure cycles and came up with few recommendations as to designing a precooling stage for a LNG cycle. They recommended, the process definition should start with compressor as it is capitally and operationally cost intensive. They also found that pure cycle is thermodynamically more efficient but involves more equipment than mixed refrigeration cycles and hence it is capital intensive (Castillo & Dorao, 2012). These were their findings which is applicable for the cooling system that we are designing here.

Taking this into account in this thesis, energy consumption is the major criterion, and hence it should be better if we go with a pure refrigeration cycle rather than mixed refrigeration cycles.

**Performance Improvements and Auxiliary Devices**

Several researches were conducted on the methods of improving the performance of the refrigeration system. Piotr A. Domanski conducted a research on the effect of adding liquid line/suction line heat exchanger or economizer or ejector on the throttling losses to a Rankine
cooling cycle. These modified cycles were compared with the regular Carnot and Rankine cycles and 38 different fluids were used for the study. It was found that, in all the three cases, the performance of the overall performance was improved with the smallest performance improvement observed in the liquid line heat exchanger cycle. The performance improvement was highest for the suction line ejector cycle provided for maximum ejector efficiency, which is not feasible in normal operating cycles (Domanski, 1995). This research gives an insight into the various options for the cooling system that can be used to the thesis undertaken here.

Similarly, Dabas conducted research on the effects of capillary tube size on the performance of the vapour compression refrigeration cycle. It was estimated that larger capillary tube sizes are more suitable for very low temperature applications (below -10 °C) and smaller capillary tubes were more suitable for normal refrigeration applications (-5 °C). He also stressed the significance of optimising the capillary tube length and size in achieving high performance from the system (Dabas, 2011).

Xing et al proposed a vapour compression cycle with an ejector sub cooling and compared it with a conventional vapour compression system in the temperature range of -10°C to -40 °C. It was experimentally determined that, the refrigeration capacity improved by 11.4% and COP increased by 9.5% when using R404A as the refrigerant. This proved that ejector sub cooling increased the performance of the refrigeration cycle. But it was also mentioned that the performance of the cycle depends on the operating pressures of the ejector (Xing, 2015).

Several articles were found on improving the performance of shell and tube heat exchangers. Feng Yang et al compared combined parallel two shell pass, combined series two shell pass shell and tube heat exchangers with a regular segmented baffle shell and tube heat exchanger. The heat exchangers were compared using a computer simulation for similar operating conditions and it was found that combined serial two shell arrangement provided the maximum performance than the other two arrangements (Yang, 2015).

Douglas T Reindl, has done extensive research on the waste heat recovery from industrial refrigeration systems and using the recovered heat for various purposes like space heating and underfloor heating. It was also explained that the quantity of heat removed and used plays a major role in justifying the capital investment for the system. The significance of quality and timely availability of heat were also discussed. The major sources that were identified in this research were screw compressor oil cooling system, reciprocating compressor head cooling system and discharge of the compressors. (Reindl, 2007). This gives
an idea of using the various waste heats from the process in an effective way. It may also help with one of the project milestones, improving auxiliary devices performance.

Jian Wen et al introduced a ladder type fold baffle shell and tube heat exchanger and compared it with conventional shell and tube heat exchanger with helical baffles. It was determined experimentally that, for the same operating boundaries and targets the ladder type heat exchanger had an increased overall heat transfer coefficient of 22.5%, which is very significant. Thus ladder type baffles can improve the performance of a baffled shell and tube heat exchangers. (Wen, Yang, Wang, Xue, & Tong, 2015)

Thus based on the literature review, it was decided to pursue either JT effect incorporation to the system or pure refrigeration cycles to the chiller and sub-cooling CO₂ for optimising the energy performance of the CO₂ capture system.
PROBLEM STATEMENT

CO₂ capture technology proposed by Surovtseva et. al (Surovtseva, et al., 2011) had good CO₂ capturing capacity but the energy consumption for the cooling system was later estimated by Qi Tian to be 10% of power plant capacity, which is very high.

The goal of this thesis is to research for various alternatives for the cooling system for the CO₂ capture technology, followed by designing and optimising it for minimum energy consumption. UNISIM software is to be used for simulation, performance evaluation of the cooling system and optimizing the whole system for minimum energy input. The construction and field testing of the cooling system would not happen as a part of this thesis. The objective of the cooling system is to reduce the feed gas temperature at 35°C (at 55 bar) to -55°C (at 55 bar).

The objective is planned to be achieved by the following methods-

- Investigating JT effect incorporation to the system
- Optimal design of Chiller
- Sub-cooling of exit CO₂ and using it for heat recovery

Hence the goal is planned to be achieved through following objectives.

1. Optimising the heat recovery system for maximum energy recovery. The target heat recovery is 80% as proposed in the initial research
2. Minimizing the overall energy input to the cooling system of the CO₂ capture technology. The target energy consumption is 13MW as proposed in the initial research.

The following milestones are expected to be met during the thesis

1. Creating a process flow diagram for the new cooling system.
2. Sizing the equipment.
3. Evaluation of the energy performance of the cooling system and optimising it for minimum energy consumption.
4. Comparison of the system with the cooling systems designed by Qi Tian and from the initial works.
5. Improving auxiliary devices performance.
INVESTIGATION OF INCORPORATING JT EFFECT

Multiple models were analysed before finalising on one model based on the cryogenic cooling system proposed by Benedict (Benedict, et al., 1988), which was discussed in the literature review section. It is designed based on the plan of taking advantage of the JT effect which was discussed earlier. The overall system model for the alternatives is shown in the figure 8 below.

![Figure 8: Cooling system modified from Benedict's Cryogenic-Cooler](image)

There are two alternatives that were finalised to be executed for this thesis based on the system discussed above. The gas is planned to be compressed and then precooled using the cold streams from the process and this low temperature gas can be further throttled to 55 bar across the JT valve which should result in massive reduction of temperature. The compression stage might be one or two resulting in two alternatives which are discussed here. The systems were modelled in UNISIM. Peng Robinson Equation of state was used here to as discussed earlier to confirm with the consistency from the original research. They are also more suitable for gas condensate systems that we are discussing here.
System Design

The compressor-heat exchangers – expander which was developed for this thesis is shown below in figure 9. This used single stage compression.

![Diagram of Alternative cooling system 1](image)

It can be seen that the cooling system contains a system of heat exchangers in between a compressor and a Joule Thompson (JT) valve, followed by an externally powered heat exchanger (chiller).

The pre-cooler system here is a system of heat exchangers using cold stream from the process to reduce the temperature of gas before throttling. Shell and tube heat exchangers with one shell and two tube passes were selected as discussed in the initial research mainly to reduce the pressure drop across each stage.

The GE feed which is entering in has a pressure of 55kPa. The feed gas was compressed to different pressures mainly to 100bar, 150bar and 200bar. The polytrophic efficiency of compressor was set at 75% as it is the average efficiency of the compressors available in the market (Kurz, et al., 2010). The JT valve opening was set at 50%. An additional powered cooler was employed in the system to ensure that the feed gas is cooled to the desired temperature if the temperature is not as expected at the exit of JT valve. Also the amount of temperature drop achieved for higher pressures were insignificant to the power consumed by
the compressor. That is the reason to limit the maximum pressure to 150 bar. It is expected that the desired temperature would be achieved through minimum energy consumption of 13MW (Surovtseva, et al., 2011) as set in the original research.

The second alternative was similar to alternative 1, except for the fact that the compression was multistage with a powered intercooler to reduce the temperature of the gas before entering the heat exchanger system. It is designed as shown in figure 6 below.

![Figure 10: Alternate cooling system 2](image)

It can be seen that there are three input energy streams for this system when compared to the earlier one. But the temperature at the exit of compressor two is expected to be lesser and hence the energy consumption is expected to be relatively lesser at the cooler side.

As this system is basically an improvement from the previous system, this system will be pursued after proving the effectiveness of the first system.

**Results and Discussion**

The system (alternative 1) was simulated using UNISIM and the results are shown in table 4. The table also shows the energy consumed for operating the system at various temperatures and pressures which helps us in assessing the effectiveness of the system.
Table 3: Power Consumption of Alternative 1

<table>
<thead>
<tr>
<th>Compressor</th>
<th>JT valve</th>
<th>Additional cooling on E105, MW</th>
</tr>
</thead>
<tbody>
<tr>
<td>$P_{exit}$, bar</td>
<td>Load, MW</td>
<td>$\Delta P$, bar</td>
</tr>
<tr>
<td>100</td>
<td>17.3</td>
<td>45</td>
</tr>
<tr>
<td>200</td>
<td>43.5</td>
<td>145</td>
</tr>
<tr>
<td>500</td>
<td>90.5</td>
<td>445</td>
</tr>
</tbody>
</table>

It can be seen that for an exit pressure of 100 bar, the pressure drop across the JT valve is in the order of 45 bar. For this high pressure drop the temperature drop is only 11 °C which is lower when compared to the work done. Hence the temperature that is achieved at the exit of the JT valve is in the range of -27 °C which is very far from -55 °C, which is the desired temperature.

Hence the total energy required to achieve the exit temperature of -55 °C is around 65 MW. This is very high when compared to the system without JT effect designed by Qi Tian, which consumed only 50 MW. Hence, the compressor-JT valve system is not adding any additional value to the system and they are actually downgrading the system performance.

With increase in compression ratio the amount of temperature drop we get is very less when compared to the energy consumption or compression. For example for an exit pressure of 200 bar the compressor load is 43.5 MW and the additional cooling load is 38.2 MW. Hence the total energy required for cooling is of the order of 82 MW.

The obtained results were not as expected and hence extensive research was carried out on the performance of the major gases across the Joule Thompson valve. As discussed in the literature review section, Joule Thompson coefficient is the amount of temperature drop per unit pressure drop across the JT valve. It is a property of the particular gas and it varies with both temperature and pressure.

The major gases in our system are carbon dioxide (40%), hydrogen (56%) and Nitrogen (3.5%). They have the maximum influence on the temperature across the valve. Hence their properties are particularly analysed. The various operating pressure of the system which we will be studying the properties are 1 bar, 50 bar, 80 bar, 100 bar, 150 bar and 200 bar.
The variation of JT coefficient with temperature for different pressure was arrived from the NIST website (NIST, 2015). The results were compiled as variation of inversion temperature for different pressure and they are shown below.

![Figure 11: Variation of Inversion Temperature with pressure](image1)

**Figure 11: Variation of Inversion Temperature with pressure**

![Figure 12: Pressure Vs JT coefficient](image2)

**Figure 12: Pressure Vs JT coefficient**

**Effect of Hydrogen**

It can be noted from figure 11 and 12 that hydrogen inversion temperature and JT coefficient are both decreasing with increase in pressure. Hence at higher pressure, the inversion temperature is lower suggesting that the gas temperature would rise across the JT valve. Above 150bar, there was no inversion temperature for hydrogen, showing that, the JT coefficient is always negative, increasing the temperature of the fluid across the valve always.
Effect of Carbon Dioxide
On the other hand, Carbon dioxide is having relatively higher inversion temperature than hydrogen and the value increases with pressure. But still the inversion temperature is very close to the operating temperature (-18°C) at operating pressures (50-100bar). Also from figure 12 it can be seen that the JT coefficient of CO₂ at operating pressure is a weak negative value. Hence the temperature of the gas is slightly raised across the valve.

Effect of Nitrogen and Argon
The Joule Thompson coefficient for Argon and Nitrogen for different temperature at 70bar which is the operating pressure is shown in figure 10 below.

It still has to be noted that, the other gases in the stream namely Nitrogen (3.5%) and Argon (1%) are having a high JT coefficient value (0.2 to 0.4) (Green & Perry, 1979), which is very high when compared to the negative coefficients of H₂ and CO₂. Hence all these factors added together, there is lower temperature drop across JT valve which is negligible when compared to the pressure drop.

Cryogenic cooling system for cooling Hydrogen gas was designed and patented for the Boeing Corporation in the same period. But this used oxygen as a primary fluid to precool Hydrogen below its inversion temperature, before throttling it to low temperature using JT valve (Hersey, 1988). This also confirms the above findings for the underperformance of the system.
Hence the method of implementing JT effect for cooling the gases didn’t meet the desired cooling objectives and therefore the second alternative was not considered for further investigation. This method did not meet the objectives because of higher concentration of hydrogen in the flue gas.

The next method experimented was the design of cooler using various refrigeration cycles. It is explained in the next section.
INVESTIGATION OF CHILLER DESIGN

The second method that is discussed in this section involves approaches and experiments on redesigning the chiller with different refrigeration cycles and with different refrigerants. This was in an attempt to maximise its efficiency. The chiller was designed using both single stage Vapour compression cycle and cascade refrigeration cycle. The various approaches taken for this method is discussed here.

Implementation of Single Stage Cycle

Referring back to figure 4 (figure re shown below) the objective of the chiller (E-105) that was used by Qi Tian in her thesis was to reduce the temperature of the feed gas from -25 °C (at 55 bar) to -55 °C (at 55 bar). In her thesis, Qi Tian found that the cooler was consuming 49MW which is high for a 500MW power plant it was designed for.

Multiple approaches were considered to optimise the cooler to reduce the power consumption. The first approach was to design a new chiller and simulate it with different refrigerants to compute the energy requirement. A number of commercially used refrigerants like ammonia, propane and ethane were investigated.
The refrigerants were selected through their operating temperature capability and availability. Out of the refrigerants, it was optimal to use ammonia, propane and ethane as they were more suitable to be used in the operating range (-25°C to -55°C).

A single stage vapour compression cycle was implemented using UNISIM and is depicted in figure 14. An expansion valve was used and a compressor with an isentropic efficiency of 70% was used to set the whole system. Stream 6 is the input feed gas stream at 55 bar and stream 5 is the output stream which was at -55 °C which is the condition as obtained from Qi Tian’s research.

The inputs for this UNISIM model were the feed stream conditions, composition of the stream 1 to 4. The operating pressure of streams 4 and 1 were taken as the saturation pressure for the refrigerant at -70 °C and they were also inputs to the system. The compressor pressure ratio was modified according to the temperature across condenser and repeated iterations were done to reduce the overall energy input. The same approach was followed for all the three refrigerants and the results and the overall energy requirement were estimated. The results are shown in the graph below.
It can be seen that the energy consumption through the single stage cycle designed is very high when compared to target value which we are focusing here. Hence this option didn’t perform as expected and therefore we go ahead with implementation of cascade cycles.
Implementation of Cascade Refrigeration Cycle

In this approach, multistage cascade refrigeration was modelled as a replacement for the chiller and its performance was analysed. The first step of designing the cascade refrigeration cycle is to identify the refrigerants that are to be used in the cycle. Sarkar et. al came up with methods to analyse the suitability of a refrigerant pair for a specific application. They researched on eight natural refrigerants and came up with suitability of a pair based on best system performance and volumetric cooling. The result of their research is shown in figure 16 below (Sarkar, et al., 2013).

![Figure 16: Selection of refrigerants for maximum COP (Sarkar, et al., 2013)](image)

For our application we require the evaporator to be at -70º C and the condenser operating at room temperature of 30º C. Based on this it was finalised that the best refrigerant pair to operate with for the application discussed here would be Ammonia – Ethane pair.

The system was modelled as follows. It can be seen that the evaporator of the top cycle acts as the condenser for the bottom cycle. Both the individual cycles were designed similarly to the vapour compression cycle that was discussed earlier. The following were the inputs to the system.

1. The input and the output condition of the feed stream
2. The refrigerant used in bottom cycle was ethane and the top cycle was ammonia
3. 50% valve opening
4. Polytrophic efficiencies of the compressors were set at 80%
5. Saturated gas at compressor inlet and saturated liquid at compressor inlet- No superheating and sub-cooling.
The overall system modelled in UNISIM is shown below in figure 17.

Figure 17: Cooling Through Cascade Refrigeration

The compressors were operated at different operating conditions and the cooling load requirement was estimated. At higher pressures the cooling load was lower and at lower pressures the cooling load was higher. Repeated iterations were done on the pressure at the exit of compressor and the minimum power consumption that we were able to achieve was 47 MW. Usage of cooling water in the condenser to reduce the energy requirement was also considered. The system implementing the idea is shown below in figure 18.

The water inlet temperature was set at 273K and the outlet temperature is set at 293K. The irregularity in this system was the temperature cross as it was handling multiphase (as discussed in the literature review section). Feed water (stream 12) at 0°C was given as input to the system and the output water (stream 11) temperature was set at 30°C and at atmospheric pressure. This gave a water consumption of 530 kg/s which is very high and would require a huge amount of energy to get it at 0°C. This method is also high energy consuming and we hence we had to come up with other energy efficient method of cooling.
The energy consumption of this method is lower when compared to the single stage cycle. This is in accordance with literature review that cascade cycle is more efficient than single stage vapour compression cycle. But the energy consumption is still high not satisfying the objectives.
INVESTIGATION OF USING SUB-COOLED CO\(_2\) AS COOLING SOURCE

After a number of attempts and not getting the desired results, the initial work done by Qi-Tian was re analysed. During the analysis it was observed that stream 10 (figure 19) exiting heat exchanger E-103 was at -20°C (55bar) and unused. Also while attempting the JT method it was observed that CO\(_2\) had higher JT effect when compared to other gases. The JT method failed because we had excess hydrogen in it. It has to be observed that, stream 10 contains very less hydrogen and it contains 99.97% carbon dioxide.

Putting all these ideas together, it was attempted to expand the CO\(_2\) and using it to further cool down the feed stream before it was entering the chiller. The idea was put in an UNISIM program and it is shown below.

![Figure 19: Subcooled CO\(_2\) as a cooling source](image)

It has to be noted that the system is similar, up-to the heat exchanger E-104, to initial research done by Qi Tian. Stream 6, which was entering into the cooler as per the initial design, was at -25°C. It had to be cooled down by -30 °C by the chiller to bring it to the desired temperature. Hence the energy consumption for that process was as high as 50MW.

According to this layout, the stream 10 exiting the heat exchanger E-103 was further expanded to 5bar with 50% valve opening. The valve opening was set at this value after repeated experiments for minimum losses. After expansion (stream 1), it was found that the
CO₂ stream was at -58°C which was a lower temperature. The full stream conditions are attached as Appendix A.

Hence this stream was used to further bring down the temperature of feed gas through heat exchanger E-105. The output conditions of the CO₂ from this heat exchanger was set as completely vapour. This signifies there is only phase change and hence therefore no sensible heat addition. This helped us to overcome the problem of temperature cross.

It was observed that, after implementing the heat recovery methods, the temperature before the chiller was brought down to -45.78 °C. Hence the energy required for further cooling down this stream to the desired temperature (-55°C) was observed to be 11.78 MW.

This was a significant energy reduction and it was also the proposed energy consumption in the initial research work done by Surovtseva et. al. Hence this meets the objective of the thesis. It has to be noted that, E105 is the new heat exchanger that was added in this system. It is also to be noted that, this heat exchanger is handling multiple phases. Hence designing the heat exchanger is discussed in the upcoming section.
Heat Exchanger design

After literature review on auxiliary devices and performance improvements it was decided that shell and tube heat exchanger is most suitable for this application because of the following reasons.

1. Can operate at high pressure conditions
2. Less fouling Concerns
3. Ease of maintenance
4. Lower pressure drop across the heat exchanger
5. Capable of handling phase changes
6. Lower cost with all the above benefits

After modelling the system in UNISIM as shown in figure 19, the operating conditions of the heat exchanger was noted down and the values are shown below.

![Figure 20: Boundary Conditions of Heat Exchanger](image-url)
Shell Side Calculations

It can be seen that, the shell side completely involves only phase change (x: 0.2648 to 1) and doesn’t involve any temperature change. Hence boiling is occurring at the shell side. Hence the scenario can be modelled into boiling over a row of tubes. In this case the heat flow is due to direct motion of fluid and buoyancy effects. Hence this condition can be modelled as an external forced convection boiling.

Assuming a low velocity flow, for a liquid which is moving at a velocity V in crossflow over a cylinder of diameter D, the maximum heat flux $q''_{max}$ is given by Lienhard and Eichhorn as follows:

$$q''_{max} = \frac{1}{\pi} \left[ 1 + \left( \frac{4}{W_{ed}} \right)^{1/3} \right]$$

(Incropera, et al., 2011)

Where:

- $\rho_v$, is the density of the vapour phase of the fluid flowing.
- $h_{fg}$, is the latent heat of vaporisation at those pressure and temperature conditions.
- $W_{ed}$, is the Weber number which is a ratio of inertia to surface tension forces and has the form,

$$W_{ed} = \frac{\rho_e V^2 D}{\sigma}$$

(Incropera, et al., 2011)

Where, $\sigma$ is the surface tension of the liquid phase.

In this case, the diameter of the tubes is assumed to be 3inches (0.0762 m) and velocity of fluid inside the shell is set at 1 m/s. The other fluid properties in shell and tube were taken from the UNISIM conditions table and it is summarised below.
Table 4: Properties in Shell Side

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
<th>Unit</th>
</tr>
</thead>
<tbody>
<tr>
<td>hfg</td>
<td>701.9</td>
<td>kJ/kg</td>
</tr>
<tr>
<td>ρvap</td>
<td>12.97</td>
<td>kg/m³</td>
</tr>
<tr>
<td>σ</td>
<td>0.02252</td>
<td>N/m</td>
</tr>
</tbody>
</table>

Based on these values the heat flux was estimated to be 4200 kW/m².

**Tube Side Calculations**

The heat flux of shell side can be equated to heat flux in the tube side assuming there is no heat loss in the system. The heat flux can be related to the tube side conditions as follows.

\[ q''_{\text{max}} = U \Delta T_{lm} \]

(Incropera, et al., 2011)

Where, \( U \) is the overall heat transfer coefficient and \( \Delta T_{lm} \) is the log-mean temperature difference which is given by the stream conditions of shell and tube,

\[
\Delta T_{lm} = \frac{[(T_{\text{in}})_{\text{tube}} - (T_{\text{sat}})_{\text{shell}}] - [(T_{\text{out}})_{\text{tube}} - (T_{\text{sat}})_{\text{shell}}]}{\ln \left[ \frac{[(T_{\text{in}})_{\text{tube}} - (T_{\text{sat}})_{\text{shell}}]}{[(T_{\text{out}})_{\text{tube}} - (T_{\text{sat}})_{\text{shell}}]} \right]}
\]

(Incropera, et al., 2011)

Using the above relations, the overall heat transfer coefficient for the system was estimated to be 194 kW/m² K. This can be used for computing the dimensions of heat exchanger using NTU method.

**NTU Method**

The NTU method of heat exchanger design was used to compute the dimensions of the heat exchanger, basically the heat transfer area from which the number of tubes can be estimated.

The effectiveness of the heat exchanger is assumed to be 80% while designing as the text specifies the value of \( q_{\text{max}} \) can deviate by 20% (Incropera, et al., 2011). The heat capacity value for hot fluid and cold fluid were calculated based on the mass flow rate and specific heat capacity of the fluids. Based on this the ratio of heat capacities (\( C_r \)) was estimated

\[ C_{\text{tube}} = m_{\text{tube}} c_{\text{tube}} \]

\[ C_{\text{shell}} = m_{\text{shell}} c_{\text{shell}} \]
The heat capacities were calculated based on the fluid properties that were obtained through UNISIM and their values are shown below.

\[ C_{\text{tube}} = 403806 \frac{J}{K} = C_{\text{max}} \]

\[ C_{\text{shell}} = 273.41 \frac{J}{K} = C_{\text{min}} \]

The ratio of heat capacities, \( C_r \), is given by

\[ C_r = \frac{C_{\text{min}}}{C_{\text{max}}} = 0.00068 \]

Since this value is close to zero, the NTU relation for the heat exchanger is given by

\[ NTU = -\ln(1 - \varepsilon) \]

(Incropera, et al., 2011)

Where, \( \varepsilon \) is the effectiveness which was assumed to be 0.8. Hence the value of NTU for 80% effectiveness is 1.61.

NTU can be further used to calculate the heat transfer area as follows,

\[ NTU = \frac{UA}{C_{\text{min}}} \]

(Incropera, et al., 2011)

The only unknown in this relation is Area of heat transfer, \( A \). This can be estimated and used to calculate the length (\( L \)) of the tube as follows.

\[ A = \pi DL \times N \]

Where ‘\( N \)’ is the number of tubes in the heat exchanger. It was assumed to be 7 and hence the length of each tube was estimated to be 1.36 m.

The shell and tube heat exchanger with 7 tubes of diameter 3 inches each and length 1.36m (heat transfer area 2.3 m\(^2\)) should be used for this application.
CONCLUSION

The purpose of this work was to develop an alternative cooling system for requirements for CO₂ capture from IGCC flue gas. Multiple integrated cooling systems were investigated, incorporating a heat recovery system which utilises the cold energy exiting the system. The initial research done by Surovtseva et. al. (Surovtseva, et al., 2011) estimated a cooling load of 13MW, but when the system was designed and modelled in UNISIM it was found to be around 50MW.

The first arrangement was based on a conventional cryogenic refrigeration cycle and utilised the JT effect. It was found that at operating pressures, the JT effect is insufficient to provide any added value to the cooling system and the energy requirement was very high in the range of 100MW. The high concentration of hydrogen in flue gas and lower JT effect of CO₂ were attributed for this low performance.

This method was followed by attempts on optimising the design of the final stage chiller used for this application. The chiller was operated using different refrigerants and different cycles, but still the energy required for cooling was very high in the range of 60 MW.

Finally another alternative involving sub-cooling the CO₂ exiting the system taking advantage of JT effect and using it to cool the feed gas was attempted. Multiple approaches were tried and finally the energy requirement for cooling for this process was 11.7 MW, which is close to the energy value of 13MW proposed in the original research (Surovtseva, et al., 2011). The heat exchanger for this process was selected to be a shell and tube heat exchanger capable of handling multiple phases and then it was sized appropriately for this application.

This improved the energy performance of the low temperature Cryogenic and Hydrate methods for pre-combustion CO₂ capture from IGCC flue gases confirming its economic feasibility. Overall, this is another step towards carbon capture and technology.
BIBLIOGRAPHY


## APPENDIX A

Full Stream Conditions of the CO$_2$ supercooling system

<table>
<thead>
<tr>
<th>Streams</th>
<th>T</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
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<th>10</th>
<th>11</th>
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</thead>
<tbody>
<tr>
<td>Vapour Fraction</td>
<td>1.0000</td>
<td>1.0000</td>
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<td>1.0000</td>
<td>0.6785</td>
<td>0.9572</td>
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<td>0.0000</td>
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<tr>
<td>Temperature (°C)</td>
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<td>22.00</td>
<td>-11.28</td>
<td>-15.51</td>
<td>-55.00</td>
<td>-25.50</td>
<td>-25.00</td>
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<tr>
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<tr>
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