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## Simulation and cost comparison of CO<sub>2</sub> liquefaction

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### Abstract

Liquefaction of CO<sub>2</sub> is an intermediate step for storage or ship transport. Two processes are suggested. The traditional method is based on external refrigeration and the other is an integrated refrigeration process. In the external refrigeration process, traditional refrigeration based on ammonia was selected. In the internal refrigeration process, liquefaction is achieved by compression, cooling and expansion of the CO<sub>2</sub>. Simulation models in Aspen HYSYS have been developed for different alternatives. A process based on ammonia refrigeration was calculated to be most cost optimum. There are however still possibilities for improvements especially for the internal refrigeration process.

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### 1. Introduction and process description

#### 1.1. Literature on alternative processes for CO<sub>2</sub> liquefaction

In a liquefaction process with an external refrigerant, CO<sub>2</sub> is compressed directly to the transport or intermediate storage pressure. Then it is liquefied using an external refrigeration cycle. Ammonia or light hydrocarbons can be used as refrigerants [1,2]. It has been claimed that using ammonia is optimum due to less power consumption [2].

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Alternative refrigerant based processes including cascades with two refrigerants are evaluated by Alabdulkarem et al. [2]. CO<sub>2</sub> can be used as a refrigerant in both an external refrigeration process and as a part of the liquefaction operation in an open process. Alabdulkarem et al. also stated that there are many studies available on CO<sub>2</sub> compression, but few studies on CO<sub>2</sub> liquefaction.

The second liquefaction method, which employs compression followed by cooling and expansion, was suggested by Aspelund et al. [1]. In this process CO<sub>2</sub> is compressed to a pressure higher than the pressure required for transport or intermediate storage. The compression can be performed with several compressor stages, and after each compression stage CO<sub>2</sub> is cooled down by ambient air or water. Once CO<sub>2</sub> is liquefied, it is expanded to meet the product specifications. Similar processes have been suggested and evaluated by e.g. Lee et al. [3] and Duan et al. [4]. Different process simulation programs including Aspen HYSYS, Aspen Plus, GT-PRO and EBSILON have been used in such evaluations.

According to Aspelund et al. [1] and Lee et al. [3] the process which employs external refrigeration cycles is not optimum for processing large amounts of CO<sub>2</sub>. Its main drawbacks are the higher cost of external refrigerants and the application of multiple heat exchangers. A study was conducted on energy and cost comparison of the liquefaction method without external refrigeration cycles and an alternative process with multistage expansion and multi-stream heat exchangers. The study calculated that the proposed alternative process reduced the energy consumption by 8 % and the total cost by 5.5 % compared to the existing processes [3].

Seo et al. [5,6] have performed economical evaluations of CO<sub>2</sub> liquefaction processes for ship-based transport after carbon capture. They evaluated four processes, one closed liquefaction process based on ammonia, and three open processes called Linde Hampson, Linde Dual-pressure and precooled Linde Hampson. The closed process showed the lowest life cycle cost while the precooled Linde Hampson process had only a slightly higher life cycle cost. For the external refrigeration process, several refrigerants were evaluated, and ammonia was found to be the most energy efficient.

In Seo et al. [6] the lifecycle cost was also calculated dependent on different parameters. Especially the lifecycle cost was calculated for the two product pressures 600 kPa and 1500 kPa. They calculated that the difference in liquefaction cost between an external and internal based process was larger for low pressure (600 kPa) than for a medium pressure (1500 kPa). It was claimed that the open process was not suitable for low pressure and temperature conditions. Decarre et al. [7] calculated that 1500 kPa was a more optimum pressure than 700 kPa for a ship based CO<sub>2</sub> transportation system. On the other hand, Lee et al. [3] specified 6.5 kPa and -52 °C as product condition in their evaluation of CO<sub>2</sub> liquefaction processes.

In general, there is no agreement in literature whether a liquefaction process based on external refrigeration or a process based on internal refrigeration is the most cost optimum. In this work, some process alternatives are compared making use of simplified process simulation and a detailed factor cost estimation method to search for the most economical process.

## *1.2. Principles for the different CO<sub>2</sub> liquefaction processes*

The principle of liquefaction after compression using a traditional refrigerant circuit is shown in Fig. 1. The refrigeration medium can be assumed to be ammonia in this process. The cooling is called external because the refrigerant is not in contact with the main (CO<sub>2</sub>) gas. The first separator separates condensed components like water before the compression. After compression and cooling, liquid which is mainly water is removed. The CO<sub>2</sub> is condensed in the heat exchanger with evaporating ammonia as the cooling medium. The evaporated ammonia is then compressed, cooled and expanded in a traditional refrigeration circuit.

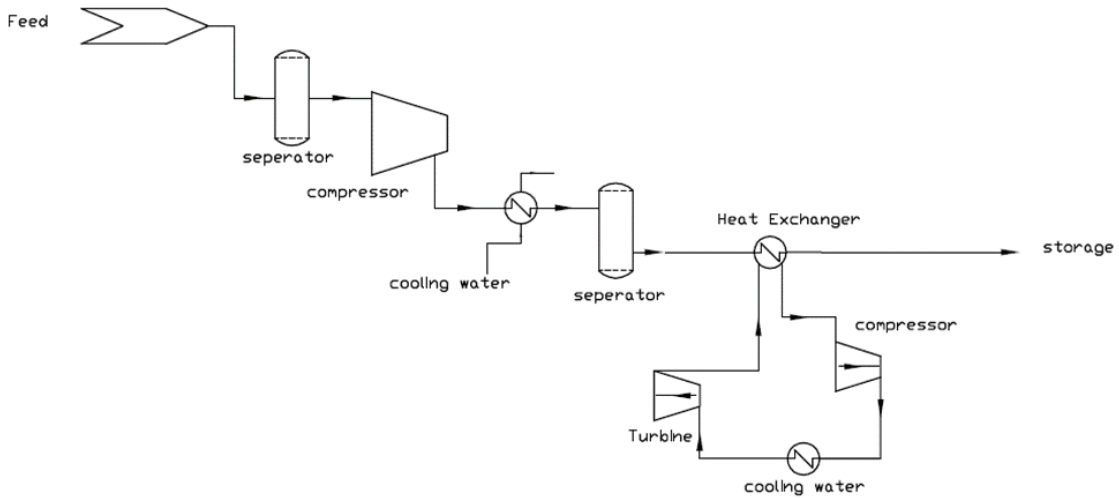


Fig. 1. Principle of CO<sub>2</sub> liquefaction based on external refrigeration

The energy consumption in a refrigeration process can be reduced by having more refrigeration circuits at different temperature levels. In Fig. 2 there are two refrigeration circuits where the first is operated at a higher (not so low) temperature in the evaporator compared to the second circuit. Because the first compression circuit does not need to achieve as low temperature, the compression ratio can be lower and the work per cooling energy unit will be lower.

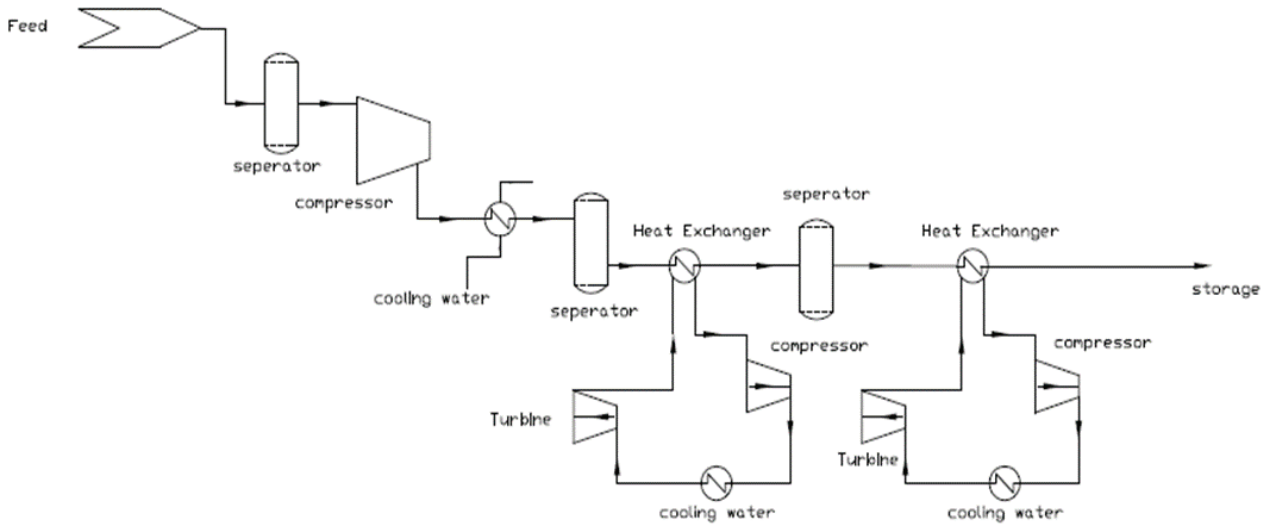


Fig. 2. Process flow diagram of the improved ammonia cooling process with two refrigeration circuits

The principle of the internal cooling process is shown in Fig. 3. The CO<sub>2</sub> is compressed to a high pressure and then cooled and expanded to the delivery pressure. The non-liquefied CO<sub>2</sub> is recirculated to the feed and recompressed.

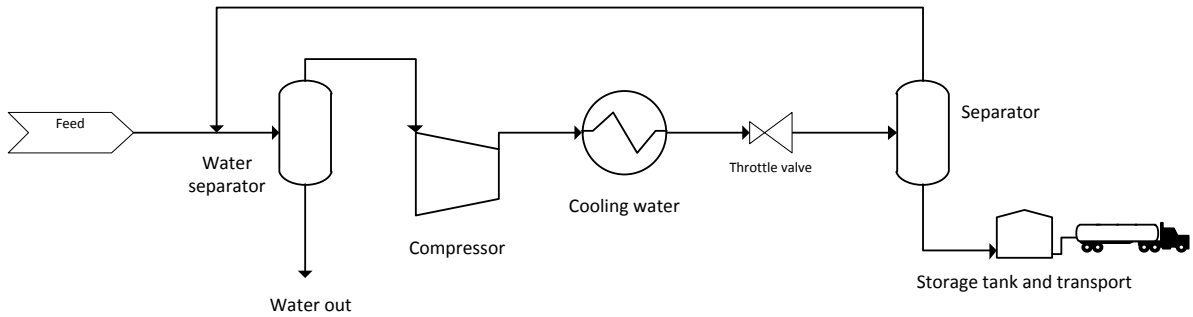


Fig. 3. Principle of the internal cooling process

The energy consumption can be reduced by performing the compression and depressurization in several steps. One alternative is shown in Fig. 4. The power consumption is reduced compared to the simple process because all the recompressed gas is not compressed from the lowest pressure.

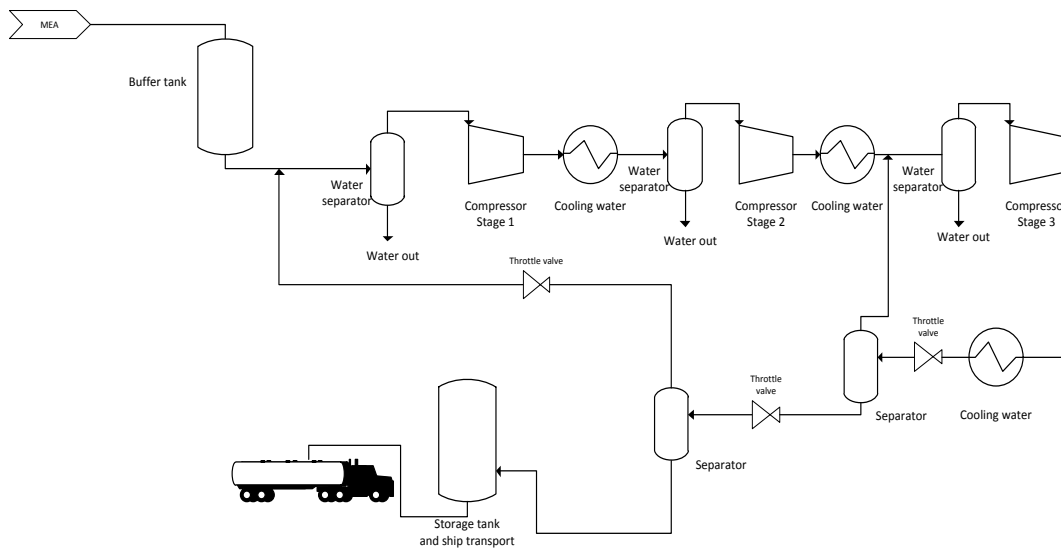


Fig. 4. Process flow diagram of the internal cooling process with several compressor stages

## 2. Simulations

### 2.1. Simulation of ammonia based liquefaction processes

A standard process as in Fig. 1 was simulated using Aspen HYSYS version 8.0. The standard Peng-Robinson equation of state [8] in Aspen HYSYS was selected to calculate equilibrium and thermodynamic properties. When specifying standard Peng-Robinson in Aspen HYSYS, the Costald method is used to estimate liquid densities. Adiabatic efficiencies of 0.85 were specified for the compressors and 0.9 for the expanders in all the simulations. The simulation flowsheet is shown in Fig. 5. Specifications for the standard case simulation are given in Table 1.

The inlet conditions are typical from an amine based CO<sub>2</sub> capture process. Impurities except from water are not included in the inlet streams. In case of a post-combustion process, the level of impurities except for water is expected to be low [9]. The pressures in the ammonia circuit are dependent on the specified temperatures at saturation conditions and have been calculated by Aspen HYSYS to be 29 kPa for the evaporating conditions and 721 kPa for the condenser conditions.

Table 1. Input specifications for Aspen HYSYS standard ammonia (base case) refrigeration process

| Parameter                           | Value         |
|-------------------------------------|---------------|
| Inlet gas temperature               | 20 °C         |
| Inlet gas pressure                  | 200 kPa       |
| Inlet gas flow                      | 125 ton/h     |
| Inlet gas water content             | 2.38 % (mass) |
| Compressor outlet pressure          | 800 kPa       |
| Ammonia evaporation temperature     | -55 °C        |
| Ammonia condensing temperature      | 15 °C         |
| CO <sub>2</sub> product temperature | -50 °C        |
| CO <sub>2</sub> product pressure    | 700 kPa       |

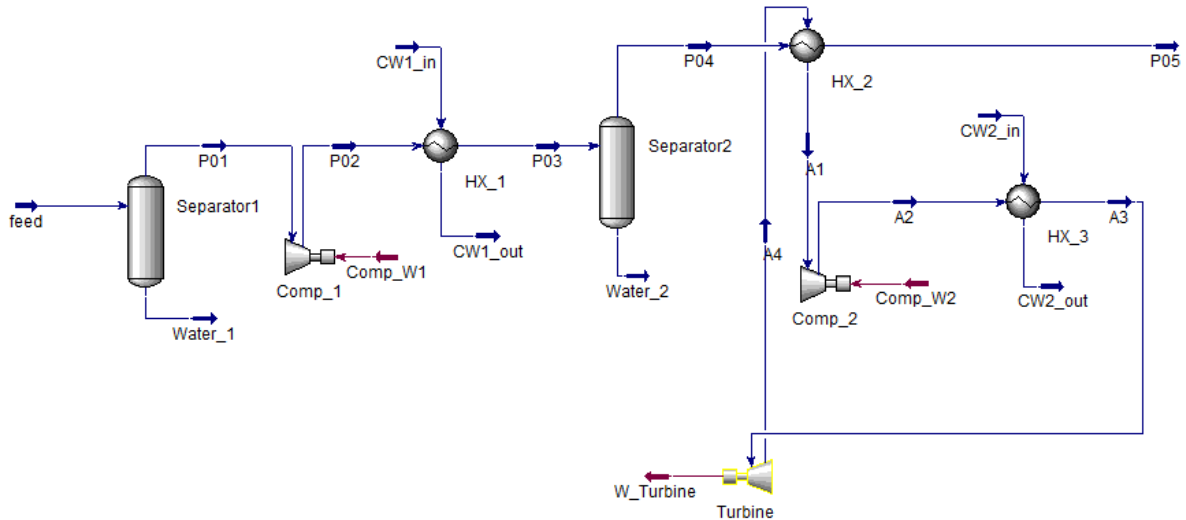


Fig. 5. Aspen HYSYS flowsheet for the standard ammonia (base case) refrigeration process

In the improved ammonia refrigeration process based on the process in Fig. 2, an additional refrigeration process was added. This had an evaporating pressure of -4 °C, which gives an evaporating pressure of 364 kPa. The pressure drop is assumed to be 50 kPa in each heat exchanger. The flowsheet in Aspen HYSYS is shown in Fig. 6.

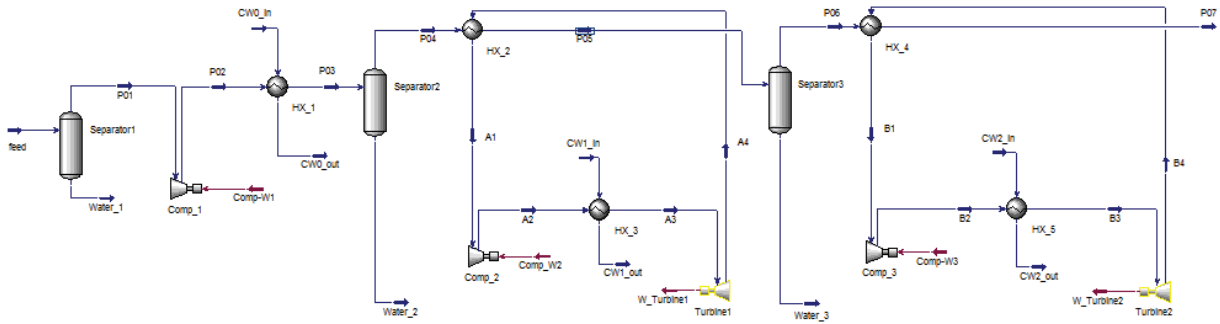


Fig. 6. Aspen HYSYS flowsheet for ammonia cooling process, improved case

2.2. Simulation of internal cooling liquefaction processes

A simple internal cooling process as in Fig. 3 was simulated in Aspen HYSYS and the simulation flowsheet of the simple process is shown in Fig. 7. Specifications for the simple case simulation are given in Table 2. The inlet conditions are the same as in Table 1. No intercooling between the stages was assumed in the compression. In practice, a compressor with such a pressure increase would have intercooling.

Table 2. Input specifications for Aspen HYSYS ammonia refrigeration process

| Parameter  | Value    |
|--|----------|
| Pressure after compression                       | 7000 kPa |
| Pressure after expansion                         | 700 kPa  |
| Temperature after cooling with water             | 15 °C    |
| Temperature in recirculating gas after reheating | -5 °C    |

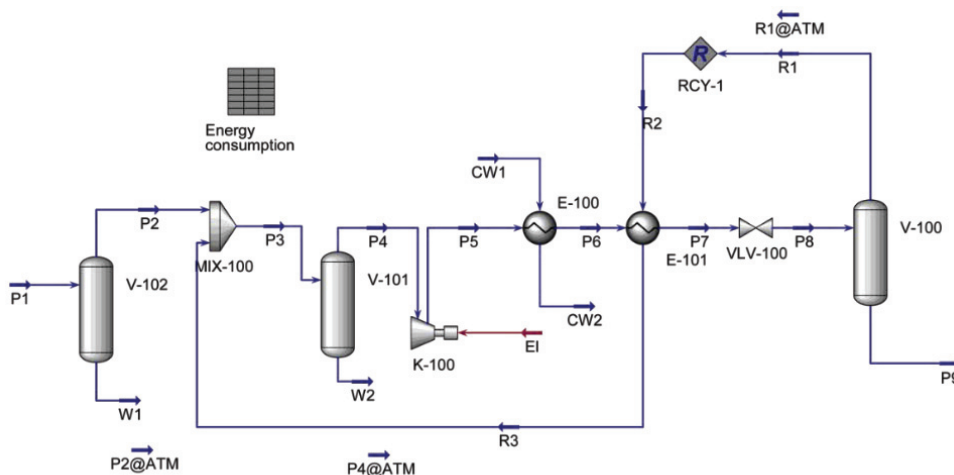


Fig. 7. Aspen HYSYS flowsheet for internal cooling process, base case

An improved process based on the process in Fig. 4 is shown in the flowsheet in Fig. 8. The pressures after each compressor are 700, 1200, 3800 and 7000 kPa. There is no further intercooling in the compressors. The intermediate pressure after first expansion is 3800 kPa and after the last 700 kPa (the product pressure). The pressure drop in each of the heat exchangers is 50 kPa. Other specifications are given in the Master project [10].

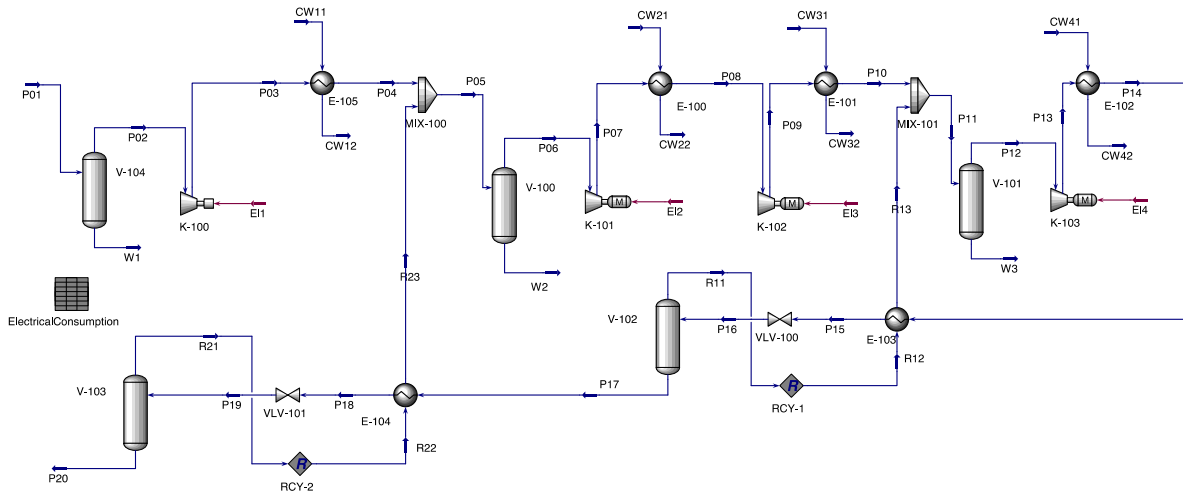


Fig. 8. Aspen HYSYS flowsheet for internal cooling process, improved case

The main results from the Aspen HYSYS simulations are presented in Table 3. It can be seen from Table 3 that the simple refrigeration process has a much lower energy consumption compared to the simple base case internal refrigeration process. It can also be seen that the difference in energy consumption is very low for the improved processes.

Table 3. Main simulation results from the alternative processes

| Process configuration                             | Net Duty<br>[kW] | Investment<br>[mill. EURO] | Operating cost<br>[mill. EURO/yr] |
|---|------------------|----------------------------|-----------------------------------|
| Standard refrigeration process (Fig. 5)           | 10040            | 22.3                       | 3.96                              |
| Multistage refrigeration process (Fig. 6)         | 10030            | 23.1                       | 3.95                              |
| Simple Internal refrigeration process (Fig. 7)    | 17900            | 27.9                       | 6.75                              |
| Multistage internal refrigeration process (Fig.8) | 10870            | 27.3                       | 4.09                              |

The duties in the compressors and the turbines and also the refrigeration heat exchangers are presented in Table 4 for the external ammonia process and in Table 5 for the internal refrigeration processes.

Table 4. Duties in the different units for the external (ammonia) refrigeration processes

| Process units    | Standard  | 2 stages  |
|------------------|-----------|-----------|
|                  | Duty [kW] | Duty [kW] |
| Comp_1           | 3559      | 3739      |
| Comp_2           | 6953      | 41        |
| Comp_3           |           | 6705      |
| Turbine_1        | 468       | 2         |
| Turbine_2        |           | 451       |
| HX_2             | 13727     | 461       |
| HX_4             |           | 13238     |
| Total (net) duty | 10044     | 10032     |

Table 5. Duties in the different units for the internal refrigeration processes

| Process units    | Simple    | Multistage |
|------------------|-----------|------------|
|                  | Duty [kW] | Duty [kW]  |
| K-100            | 17918     | 3170       |
| K-101            |           | 1941       |
| K-102            |           | 4009       |
| K-103            |           | 1752       |
| Total (net) duty | 17918     | 10872      |

The coefficient of performance (COP) defined as cooling duty from the refrigeration circuit divided by the difference between the compressor duty and the turbine duty was calculated to 2.0 for the standard case using ammonia refrigeration. When two refrigeration loops are used, the COP was increased in the first. However, the total efficiency improves only slightly, because most of the cooling is performed in the circuit with the lowest COP. There is also an additional pressure drop in the alternative process with two refrigeration circuits.

### 3. Dimensioning, cost estimation and optimization

#### 3.1. Equipment dimensioning

The main equipment units have been dimensioned by traditional methods. To calculate the heat transfer area in the heat exchangers, duties and temperature conditions from the simulations were used. The overall heat transfer number in the heat exchangers were estimated to be 2500 W/(m<sup>2</sup>·K) for the ammonia condensers, 2000 W/(m<sup>2</sup>·K) for the CO<sub>2</sub> condensers, 250 W/(m<sup>2</sup>·K) for the gas/water coolers and 400 W/(m<sup>2</sup>·K) for the other heat exchangers without phase change. To calculate the diameter of the separators, the vertical gas velocity in separators was specified to 5 m/s. The steel thickness was specified to 0.01 m. Other specifications are given in the project report [10].

#### 3.2. Operating cost

The electricity cost was specified to 0.05 EURO/kWh. Other operating cost factors like cooling water and maintenance were neglected. The cost of cooling water is assumed to be small compared to the electricity cost. Normally, the maintenance cost is estimated as a percentage of the investment. Because the investment is dominated



by the compressors, it can be assumed that the estimated maintenance cost would be close to proportional to the power consumption like the electricity cost.

### 3.3. Estimation of investment

Investment cost was calculated using a detailed factor method. The total investment is the sum of installed cost of each equipment unit. Installed cost for each equipment unit is the cost of each equipment unit in carbon steel times an installation factor. The installation factor is a function of type of equipment, equipment size, material and site description (soil, existing buildings, building types etc.). Details can be found in the Master project [10].

Estimates on equipment cost have been found by an internet calculator [11] based on the textbook of Peters et al. [12]. The cost has been adjusted for currency and year. An exchange rate of 8.5 has been used to convert from NOK to EURO. The cost estimate is a 2014 estimate at a generic location (Rotterdam). With a 20 % contingency the estimates will have a typical accuracy of +/- 40%.

### 3.4. Results of cost calculations

The main results of the cost estimation calculations are presented in Table 3. The process with the lowest investment (and also the lowest total cost) is the simple process based on external refrigeration. The process with lowest operating cost is the process based on two external refrigeration circuits. The improvement with two refrigeration circuits is however small.

The improved process with internal refrigeration has an operating cost close to the processes based on external refrigeration. The investment cost is however larger. When comparing the simple and improved process with internal refrigeration, the improved process has both a lower investment and a lower operating cost. One reason for the high cost of the simple process with internal refrigeration is that it is not specified with intercooling.

The cost data for each equipment unit in carbon steel (CS), the installation factor and the total (installed) equipment cost is presented Table 6 for the external ammonia process and in Table 7 for the internal refrigeration processes.

Table 6. Cost data for all the process units for the external (ammonia) refrigeration processes [mill. EURO]

| Equipment unit | Unit Cost (CS) | <u>Standard</u> |               | Unit Cost (CS) | <u>Two-stage</u> |               |
|----------------|----------------|-----------------|---------------|----------------|------------------|---------------|
|                |                | Inst. Factor    | Total Eq.Cost |                | Inst. Factor     | Total Eq.Cost |
| Separator1     | 0.03           | 8.98            | 0.28          | 0.03           | 8.98             | 0.28          |
| Comp_1         | 2.08           | 3.48            | 7.22          | 2.17           | 3.48             | 7.57          |
| HX_1           | 0.04           | 8.98            | 0.35          | 0.04           | 8.98             | 0.35          |
| Separator2     | 0.02           | 8.98            | 0.15          | 0.02           | 8.98             | 0.15          |
| HX_2           | 0.02           | 8.98            | 0.42          | 0.01           | 8.98             | 0.11          |
| Turbine1       | 0.13           | 5.61            | 0.73          | 0.003          | 12.88            | 0.04          |
| HX_3           | 0.02           | 8.98            | 0.15          | 0.009          | 14.01            | 0.13          |
| Comp_2         | 3.74           | 3.48            | 13.01         | 0.04           | 8.13             | 0.30          |
| Separator3     |                |                 |               | 0.02           | 8.98             | 0.14          |
| HX_4           |                |                 |               | 0.03           | 8.98             | 0.46          |
| Turbine2       |                |                 |               | 0.13           | 5.61             | 0.72          |
| HX_5           |                |                 |               | 0.02           | 8.98             | 0.15          |
| Comp_3         |                |                 |               | 3.65           | 3.48             | 12.70         |
| <u>Total</u>   | <u>6.07</u>    |                 | <u>22.31</u>  | <u>6.06</u>    |                  | <u>23.09</u>  |

Table 7. Cost data for all the process units for the internal refrigeration processes [mill. EURO]

| Equipment unit       | UnitCost(CS) | Simple       |                | Unit Cost (CS) | Multi-stage  |                |
|----------------------|--------------|--------------|----------------|----------------|--------------|----------------|
|                      |              | Inst. Factor | Total Eq. Cost |                | Inst. Factor | Total. Eq.Cost |
| Separator V-100      | 0.01         | 8.98         | 0.13           | 0.01           | 14.0         | 0.11           |
| Separator V-101      | 0.02         | 8.98         | 0.17           | 0.01           | 14.0         | 0.11           |
| Separator V-102      |              |              |                | 0.01           | 14.0         | 0.11           |
| Separator V-103      |              |              |                | 0.01           | 14.0         | 0.11           |
| Compressor K-100     | 7.8          | 3.48         | 27.12          | 1.86           | 3.48         | 6.47           |
| Compressor K-101     |              |              |                | 1.17           | 4.29         | 5.02           |
| Compressor K-102     |              |              |                | 2.32           | 3.48         | 8.08           |
| Compressor K-103     |              |              |                | 1.06           | 4.29         | 4.56           |
| Heat Exchanger E-105 |              |              |                | 0.04           | 8.98         | 0.35           |
| Heat Exchanger E-100 | 0.03         | 8.98         | 0.19           | 0.04           | 8.98         | 0.95           |
| Heat Exchanger E-101 | 0.01         | 14.0         | 0.14           | 0.04           | 8.98         | 0.68           |
| Heat Exchanger E-102 |              |              |                | 0.05           | 8.98         | 0.42           |
| Heat Exchanger E-103 |              |              |                | 0.01           | 14.0         | 0.08           |
| Heat Exchanger E-104 |              |              |                | 0.01           | 14.0         | 0.12           |
| <u>Total</u>         | <u>7.86</u>  |              | <u>27.74</u>   | <u>6.62</u>    |              | <u>27.27</u>   |

#### 4. Discussion

The processes based on traditional ammonia refrigeration have been calculated to be both energy and cost optimum. However, the improved process with internal refrigeration was not far from being energy optimum. The simple process based on internal refrigeration had a very high energy consumption. The process was however not very realistic because it was not assumed any intercooling in the compressor. The calculations indicate that it is the process with internal refrigeration which has the largest potential for improvements.

The power consumption for liquefaction has been calculated in this work to values between 10.0 and 17.9 MW for 125 ton CO<sub>2</sub>/h. Alabdulkarem et al. [2] calculated values for different options to values between 6.1 and 8.65 MW for liquefaction of 72.4 ton CO<sub>2</sub>/h to 600 kPa. The lowest values are very close, 80 and 84 kWh/ton CO<sub>2</sub> respectively.

Some literature references [1,3] claim that the liquefaction processes based on ammonia is not optimum. Other literature references [2] claim that the ammonia based liquefaction process is both most energy and cost optimum, and Seo et al. [5,6] claim that a closed process based on ammonia has the lowest cost, but a special open process (precooled Linde Hampson) has only slightly higher life cycle cost.

The most important factor when comparing the cost of the processes is the power consumption. Also in Lee et al. [3] and in Seo et al. [6] the capital and operating cost was dominated by the compressors. The operating cost is in our calculations proportional to the power consumption. But also the investment is very dependent on the power consumption because the cost of compressors is the dominating investment. The results of this work indicate that looking for the most cost optimum solution is close to looking for the most energy optimum solution.

The possibility to replace some of the expansion valves with turbines is one suggestion for reducing the power consumption based on internal refrigeration. Intercooling between compressor stages is also a possibility to increase the efficiency in the process. Lee et al. [3] calculated cost optimum pressure ratios between 2 and 4 in a CO<sub>2</sub> liquefaction process.

An efficiency of 0.85 has been used for the compressors and 0.9 for the turbines. Centrifugal compressors have been assumed. For the compressors, assuming the same efficiency in both processes should make the comparisons between the process alternatives reasonable. Even though there is more experience with ammonia compressors, the challenges with CO<sub>2</sub> and ammonia compressors are regarded to be similar. For the turbines, only the suggested external refrigeration process make use of turbines, so a high estimated efficiency would favor external refrigeration.

However, a possible improvement for the internal refrigeration processes is the use of turbines. In that case, an increased estimated turbine efficiency should not influence much on the comparison between the process alternatives.

The optimum pressure after liquefaction is not obvious. In this work 700 kPa is specified to achieve low investment in storage tanks due to lower wall thickness. Seo et al. [6] claim that an internal process is unsuitable for low product pressures (600 kPa). In this work, a product pressure of 700 kPa is used, and the difference between an improved internal refrigeration and an ammonia based process was small also for this low pressure.

More research is necessary to find the cost optimum process for CO<sub>2</sub> liquefaction. A reasonable approach is to first search for processes with a low energy consumption, and then cost estimate these alternatives. Simulation models e.g. in Aspen HYSYS should be useful for further development.

## 5. Conclusions

Process flowsheets for the liquefaction of 1 million tons/yr CO<sub>2</sub> have been simulated in the process simulation program Aspen HYSYS. Improved cases with several refrigeration stages and several compression stages were calculated for both external refrigeration and integrated refrigeration. Equipment dimensioning, equipment cost estimation and energy cost estimation were performed for the different process alternatives for comparisons.

The most expensive process units from both a capital and an operating cost point of view, were the compressors. Because of that, the energy optimum process was also the cost optimum process. The improved alternatives had energy consumptions close to each other. Among the evaluated cases, the most cost optimum process was a process based on external refrigeration. The investment was estimated to 23 million EURO and the operating cost was estimated to 4 EURO/ton. The investment was estimated to be 4 million EURO higher and the operating cost was estimated to be only 0.13 EURO/ton higher for the internal refrigeration process.

There are still possibilities for improvements especially for the process based on internal refrigeration. The developed Aspen HYSYS models are regarded to be useful in search of finding a cost optimum liquefaction process.

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