Design and analysis of the natural gas liquefaction optimization process- Energy Storage of
Cryogenic Carbon Capture (CCC-ES)

Farhad Fazlollahi, Alex Bown, Edris Ebrahimzadeh and Larry L Baxter

Chemical Engineering Department, Brigham Young University, Provo, UT 84602, USA.

Corresponding author: larry_baxter@byu.edu

Abstract- The cryogenic carbon capture (CCC) process provides energy- and cost-efficient carbon capture and can be configured to provide an energy storage system using an open loop natural gas (NG) refrigeration system. This system stores energy during non-peak times by liquefying and storing natural gas to be used as a refrigerant. This investigation compares four different natural gas liquefaction processes simulated by Aspen HYSYS as incorporated as part of the CCC-ES process. In these processes, LNG vaporizes in the CCC process and the cold vapors return through the LNG heat exchangers exchanging sensible heat with the incoming flows. Aside from this difference, this investigation uses process designs similar to traditional LNG processes. The simulations meaningfully compare these alternative liquefaction options, eliminating differences in assumptions and process details inherent in comparing processes simulated by different authors or different codes. The comparisons include costs and energy performance with individually optimized processes, each operating at three operating conditions: energy storage, energy recovery, and balanced operation. Given similar quality turbomachinery, efficient heat exchangers in particular reduce energy input requirements and maximize energy savings and capital costs, including heat exchangers used to cool compressed gases.

Keywords: Cryogenic carbon capture, Liquefaction of natural gas, Aspen HYSYS, Optimization

Introduction

Energy storage represents one of the most critical needs in our current energy infrastructure and this need will increase as intermittent and alternative energy production increase. Energy storage (ES) is the conversion of energy from one form that is difficult or expensive to store to a different form that stores more conveniently or economically and, typically at a later time, the conversion back to the original or to another useful form. Energy storage differs from energy sources in that the roundtrip conversion involved in energy storage at best conserves the useful energy and in practice always loses some energy or availability (if it did not, it would be an energy source). Nearly always, interest in energy storage is driven by capacity, convenience, or economic considerations that mitigate the inherent energy loss of the process. The most desirable characteristics of these energy storage systems include: sufficient capacity to affect grid dispatch schedules; rapid response to changes in demand; high round-trip efficiency; and low cost.

This discussion highlights an energy storage process enabled by Cryogenic Carbon Capture™ (CCC). CCC provides cost-effective and energy-efficient CO₂ separation from flue gases compared to the alternative technologies [1-4]. This system consists of two major subsystems: cryogenic carbon capture, and energy storage via natural gas liquefaction. The natural gas liquefaction is integrated into CCC as a refrigerant to cool the flue gas. The CCC process is as follows:

Flue gas that would otherwise be exhausted to the stack enters the CCC process, dries, and cools. After this preliminary cooling, it flows through a recuperative heat exchanger, cooling to near its CO₂ frost point. Other pollutants, such as Hg, As, NO₂, HCl, and SO₂, condense or de-sublimate as the flue gas cools allowing CCC-ES to exceed the target removal of most criteria pollutants,

mercury, and hazardous gases. Further cooling in patented de-sublimating heat exchangers removes CO₂ as a solid and leaves a light gas stream. A solids handling system pressurizes the CO₂ to at least the triple point and at most the final CO₂ product delivery pressure. A heat exchanger recuperates energy from the sensible heat of the light gas and from the cold solid CO₂ as it warms, melts, and further warms to room temperature (Figure 1-3).

The CCC process and the LNG-based cooling loop deliberately operate independently of one another, with essentially no heat integration aside from the LNG delivery to CCC and its return. The CCC process often couples to a power plant or some other CO₂-producing process that preferably operates at constant or very slowly varying load. The energy storage process, by contrast, responds rapidly to changes in demand, intermittent renewable energy sources, and other transients with widely varying time constants. Both processes involve substantial internal heat integration, but heat integration between them would destroy the ability of the LNG cooling process to respond rapidly to load changes without affecting the CO₂ capture process.

The CO₂ removal process requires a constant source of refrigerant (Balanced case). The LNG used as a refrigerant to drive the system can be stored and later used in with the LNG process turned off. During off peak hours, the energy-storing (ES) version of the process generates more refrigerant in the compressors than is needed for the process and the excess refrigerant is stored in an insulated vessel as a liquid at the low-temperature, modest-pressure point in the cycle. During peak demand, the previously stored energy in the form of the stored, cold, liquid refrigerant provides energy recovery (ER) by replacing refrigerant that must be generated for CO₂ capture with the stored refrigerant, eliminating nearly all of the energy demand required by cryogenic carbon capture for as long as the stored refrigerant lasts.

The distinction between the two cooling loops is elaborated in more detail here. After separating CO₂ from flue gas, the pure CO₂ solid product is well below its melting point, the latter being approximately -55 °C depending very slightly on pressure. This pure CO₂ stream melts at essentially constant temperature, which melting involves a significant amount of the energy required in the CCC process. This is illustrated in the diagram above as Coolant Loop I or Inner Loop. A single-component cooling loop efficiently transfers the heat of CO₂ fusion from the melting point (near -56 °C) to lower temperatures by condensing at the CO₂ melting point and at an elevated pressure and vaporizing at a lower temperature and pressure, in this case at the lowest temperature in the loop. Since the CO₂ is essentially pure at this stage, this CO₂ melting/refrigerant condensation process occurs isothermally on both sides of the heat exchanger, minimizing losses. The lowest temperature at which CO₂ forms a liquid phase at any pressure is the triple point, which is -56.6 °C (at a pressure of 5.2 bar). Therefore, the inner-loop refrigerant must condense at this temperature or higher and at some reasonable pressure to provide the heat to melt the solid CO₂. It is possible to provide this heat exchange with sensible heating or with a mixed refrigerant, but neither of these options can occur at a single temperature. They would therefore create an increasing temperature difference in the heat exchanger that would lead to larger inefficiencies.

Cooling Loop II, or the outer cooling loop, transfers more heat than the inner loop in general, and unlike Cooling Loop I, this heat transfer occurs over a broad temperature. The baseline CCC systems uses several refrigerants in this loop to optimally match the temperature profiles on both sides of the heat exchangers, that is, to maintain a constant and small temperature difference throughout the process.

During normal operation, that is, when energy is being neither stored nor recovered, there is no NG flow to the process and no power output from the NG turbine. The outer cooling loop uses NG as a refrigerant as indicated. The overall cooling loop flow is highlighted with thick, red lines. During energy storage, NG enters the outer loop compressor from an external supply, increasing the total amount that flows through the compressor compared to normal operations and hence increasing the compressor power consumption. The excess NG flows into the LNG storage at V1 with the normal amount of NG continuing through the process, increasing the LNG level in the storage vessel. The vessel exit is closed to the process loop, allowing the NG to accumulate. This stores the extra compressor energy in the form of LNG at cryogenic temperature and modest pressure. When the stored energy is used, the external NG supply valve closes and the external loop compressor load can decrease potentially to zero. Some or all of the cooling is provided by the stored NG, which flows from the vessel through the process and eventually to the NG turbine, where it is combusted. The hot gases from the NG turbine vent into the boiler, providing NG combined cycle efficiency without having to build an additional Rankine cycle. During this phase, the power required for the largest compressor decreases, potentially to zero, and the turbine generates additional power.

This investigation considers four refrigeration processes, each operating at three conditions. The conditions include:

- Energy storing, where LNG production increases by 40% to use excess energy available on the grid,
- Balanced, where LNG production equals LNG demand from the power plant to run the CCC process, and

- Energy recovery, where LNG production decreased by 70% to decrease the parasitic loss and hence place more power on the grid when demand is low.

The theoretical energy demands for any carbon capture process are quite high – in all cases much larger than the sum of all other parasitic loads combined for commercial coal boilers and similar power plants. The CCC-ES process shifts its energy demand to periods of low power costs and high power excess capacity. This time shifting or demand response enables a form of energy storage at very large scale, highly efficient, and very cost effective. This energy storage system has several practical requirements one of which is a refrigerant that can be stored at both the cold and warm ends of the cycle. Storing cold refrigerant in liquid form is neither difficult nor expensive, but storing warm gaseous refrigerants in quantities needed for this process is not practical. Therefore, the energy storing version of CCC uses natural gas as the refrigerant. Natural gas storage as a liquid is a well-established technology, and warm natural gas can either be placed back on the pipeline or burned in a gas turbine to generate more power.

A natural gas liquefaction plant is energy and cost intensive. Therefore, one of the major issues in the liquefaction plant is to reduce energy input requirements and total cost [5]. Several processes have been reported for natural gas liquefaction, but few of them are in use (onshore plants) and many do not have industrial references. There are mainly three types: cascade liquefaction process [6, 7]; mixed refrigerant liquefaction process [8-10]; and expander liquefaction process [11, 12]. The comparison among these liquefaction processes has been conducted by experts on energy input requirements, exergy analysis, liquefaction rate and economic performance. Some selected mixed refrigerant process for the lowest energy input requirements [13, 14], while some proposed the expander cycle [12]. Nevertheless, the recent research has indicated that nitrogen expansion [15] is the most adaptive process considering

energy input requirements, economic performance, safety, sensitivity to motion and operability. Shell has simulated the cascade, mixed-refrigerant and N₂ expander cycle and analyzed their advantages and disadvantages [16]. Expander cycles have low footprints and high inherent safety as no storage of hydrocarbon refrigerant is required. The major disadvantage is its relatively high power consumption. Alabdulkarem et al. [10] optimized the mixed refrigerant cycle with propane precooling and found a new composition of mixed refrigerant that can reduce the unit product liquefaction power consumption. Shen et al. [17] proposed a process to liquefy natural gas by utilizing the gas pipeline pressure energy, but did not make a detailed analysis and optimization. He and Ju [18] also evaluated and analyzed the exergy losses of different equipment in the process and showed that their liquefaction process is suitable to utilize the gas pipeline pressure due to its low unit energy input requirements, simplicity and flexibility.

It is well established that heat exchanger and compression cause exergy loss that can be minimized by minimizing heat exchanger temperature differences and using staged, inter-cooled compressors [19]. Process optimizations generally focus on balanced, steady flow rate systems. This investigation differs from others in that the objective is to minimize exergy losses and costs in a process experiencing continual load changes as it shifts through the cycle of energy storing to balanced operation to energy recovery. This investigation analyzes four different liquefaction models in Aspen HYSYS and optimizes them using the Visual Basic for Applications (VBA) optimizer. These processes are: propane-precooled mixed refrigerant (C3-MR) cycle, a modified dual mixed refrigerant (MDMR) cycle and two single mixed refrigerant (SMR) cycles. Results show that for SMR models energy input requirements have been reduced to 0.06 kW/kg NG for Balanced, 0.008 kW/kg NG for ER and 0.07 kW/kg NG for ES which are the minimum compared to other models. SMR models have the highest heat exchanger's efficiencies of above

90% for Balanced, highest efficiencies for ER. For ES, MDMR has the highest efficiency. C3-MR has the exergy loss of 0.0046 kW/kg NG for ER, 0.0356 kW/kg NG for Balanced and 0.0393 for ES which are the least compared to other models. MDMR is the most expensive model for 71,146,000 dollars capita cost whilst C3-MR has the highest electricity cost of 3389.7 (\$/h) and 121.2 (\$/h) for water.

The model simulations include flow and thermal transients and the consequent responses of all processing steps. The transient flows are a non-trivial contribution in the energy performance and their analysis and inclusion is a distinguishing characteristic of these simulations. The Aspen Energy Sensitivity model further optimized the best models (SMR cycles). The discussion includes comparisons with available literature results. However, the comparisons of these four processes discussed in this investigation use the same assumptions, the same thermodynamics and process models, and are optimized using the same techniques as one another and therefore provide a more reliable comparison to one another than is possible in comparing to independent literature results. This analysis also includes comparisons during balanced operation and during intentionally imbalanced operation, unlike most literature investigations.

2. Process design

2.1. Feed gas parameters

The simulations discussed in this paper require several specifications as summarized in Table 1. A feed gas flow rate of 21200 kgmol/h represents the amount of natural gas required for the steady operation of CCC on a 550 MW power plant. During energy storage and recovery the rates change to 29680 kgmol/h and 6360 kgmol/h, respectively. These flow rates represent a 40% increase and a 70% decrease in coolant flow. The assumption here is that the amount of cooling

provided to the CCC process at the power plant is constant (base-loaded plant) while the demand on power for the LNG production process could change +40% to – 70% based on grid variations. In reality, the off-peak demand commonly lasts much longer than the peak demand, so the increase in LNG flow would be much smaller than even these numbers reflect compared to a potential large decrease in flow. All other parameters mentioned in Table 2 remain constant. The liquefaction rate is 100% for all four models after the main heat exchanger, but to satisfy our purposes in the CCC process, the LNG partially vaporizes (27%) after the main heat exchanger.

2.2. Liquefaction process

On the basis of the characteristics of a full-scale LNG plant, three sets of typical liquefaction processes were designed in this paper for analysis and comparison. Four different models have been simulated using Aspen HYSYS. Two different single mixed refrigerant cycles (SMR), one Modified Dual Mixed Refrigerant (MDMR) and one Propane-pre-cooled mixed refrigerant (C3-MR) cycle are simulated and optimized to choose the best model with regard to energy input requirements, exergy loss, cost and heat exchanger efficiencies.

Single Mixed Refrigerant cycle (SMR)

The SMR is the simplest NG liquefaction process. The Single MR Cycle uses only one MR loop for pre-cooling, liquefaction, and sub-cooling (Figure 4). The cycle compresses and cools the refrigerant until liquid is formed, and then vaporizes the refrigerant in the LNG exchanger. The refrigerant cools -condenses the natural gas, NG, to liquefied natural gas, LNG. The refrigerant composition is optimized such that the temperature profiles in the heat exchangers are as close and parallel as possible.

The simulation flow sheet of the SMR process (Figure 4) uses an eight-stage compressor with inter-stage cooling. Phase separators separate the mixed refrigerant into gas and liquid phases in each cooler. Pumps compress the liquid after the phase separator. A single LNG heat exchanger liquefies and cools the natural gas to the required LNG storage condition of about -119.4 °C and 11.45 bar pressure. Subsequent expansion prepares the LNG for the CCC process. Table 3 summarizes the optimized mixed refrigerant composition and key parameters, where ER, B and ES represent energy storing, balanced, and energy recovery respectively, that is, conditions when LNG is accumulating for future use by the CCC process, when LNG demand from the CCC process is balanced with its production rate, and when stored LNG levels are dropping.

Modified Dual Mixed Refrigerant (MDMR)

In the modified Dual MR cycle, a high boiling point Warm Mixed Refrigerant (WMR) containing methane, ethane and propane replaces the pre-cooling propane refrigerant in the SMR process. This significantly reduces the propane inventory in the process. The MDMR process achieves efficiencies comparable to C3-MR (discussed later) and has been used successfully in land-based LNG [6]. The MDMR design balances refrigeration loads across the two similar refrigerant compressors, reducing overall power requirements, increasing the applicable range for the most robust centrifugal compressor designs and reducing spare parts costs. The WMR forms a single liquid phase after compression, cools in the main heat exchangers pre-cooler, and flashes to a single pressure level to provide pre-cooling refrigeration. The process can therefore be configured and optimized to meet the project requirements. Figure 5 includes the simulation flow sheet of the MDMR process. This process adds a second loop to the SMR design, which increases the cost relative to the SMR even though the two loops require smaller compressors than the SMR. This additional loop controls the flow rate and temperature, resulting in the most

efficient heat exchanger among these processes. Other options to avoid the methane loop are not practical like using ethane instead of methane. The minimum temperature approach is unrealistically less than 1 °C. The process can therefore be configured and optimized to meet the project requirements. Table 4 and Table 5 indicated the optimized mixed refrigerant conditions and key parameters for the warm and the cold loops, respectively.

Propane-pre-cooled mixed refrigerant cycle (C3-MR)

The C3-MR process produces over 80% of the world's LNG. This process combines a propane-based refrigeration loop for the initial cooling to around -30 °C with a mixed refrigerant for cooling to the final conditions (Figure 6). Natural gas liquefies at high pressures (20-50 bar) and condensation temperatures at or near the critical temperature (about -83 °C). The propane also pre-cools the compressed, mixed refrigerant. After pre-cooling, the partially condensed mixed refrigerant separates into liquid and vapor phases. As the mixed refrigerant vaporizes and flows downward the shell side of the main heat exchanger, it cools and condenses the natural gas. The vaporized mixed refrigerant returns to the compressor to complete the loop. The single-component pre-cooling fluid provides an efficient, easy to control pre-cooling step. The mixed refrigerant permits phase change over a temperature range that matches the range of condensation temperatures of the NG, leading to high efficiency. Consequently, the C3-MR cycle minimizes the number of equipment items and leads to easier operation, and high reliability. Table 6 and Table 7 provide optimized mixed refrigerant conditions and propane precooled conditions, respectively.

2.3. Phase equilibrium equations

Phase equilibrium provides the quantitative basis for much of the process simulation. The Peng-Robinson equation is used in this study. The PR equation is a cubic equation of state:

$$P = \frac{RT}{v-b} - \frac{a}{v(v+b) + b(v-b)}$$

Where:

$$a = \sum \sum z_i z_j (a_i a_j)^{0.5} (1 - k_{ij})$$

$$b = \sum z_i b_j$$

The PR equation can also be expressed in the form of a compression compressibility factor, in which form its classification as a cubic equation of state is more evident (cubic in compressibility):

$$Z^3 - (1-B)Z^2 + (A-3B^2-2B)Z - (AB-B^2-B^3) = 0$$

Where:

$$Z = \frac{PV}{RT}$$

$$A = \frac{aP}{(RT)^2}$$

$$B = \frac{bP}{RT}$$

3. Process optimization

The C3-MR, MDMR, and a version of the SMR system called SMR 2 models developed in Aspen HYSYS provided energy input requirements, cost and exergy utilization rate, and the three factors taken as figures of merit for natural gas liquefaction. This investigation minimized energy input requirements by changing mixed refrigerant composition, pressure, temperature, and flow rate, subject to several constraints discussed later. The exergy and cost of these optimized processes were then compared. The work done per unit of LNG production formed the objective function:

$$f(X) = \min\left(\frac{W_{net}}{q_{LNG}}\right)$$

Where:

$$W_{net} = \sum W_{compressors} - \sum W_{expanders}$$

q_{LNG} represents the mass flow of the produced LNG. The constraints for this optimization include:

- 1) A minimum internal temperature approach (MITA) >1 °C
- 2) No condensates in Compressors (100% vapor flow)
- 3) A single liquid phase in turbines and pumps (100% liquid flow)
- 4) The sum of mixed refrigerant mole/mass fraction is 1

4. Result and analysis

The optimization indicates that SMR 2 consumes the least energy in all three operation modes (energy recover, balanced, and energy storing). The Aspen energy sensitivity analysis using a genetic algorithm provided a marginally improved set of conditions and refrigerant compositions using the same objective function. This new set of conditions is called SMR 1 (Table 7). As indicated in Figure 7, SMR 1 demands the least energy during energy recovery (ER) and balanced (B) cases while SMR 2 demands the least energy during energy storage (ES).

Using all of these constraints and key parameters, it was concluded that SMR 2 is the best model for energy saving. Using the same objective function and constraints, SMR was then optimized using Aspen plus Energy Sensitivity and genetic algorithm and was renamed to SMR 1. The resulting optimized energy usage was less than the original optimization method. . Hence, the only difference between SMR 1 and SMR 2 is the optimization method and all other conditions are kept constant. As a result, SMR 1 was concluded to be the most energy efficient model to our general objective function. The energy input requirements results are shown in Figure 7. As shown, SMR 1 has the least energy input requirements for ER and Balanced cases. For ES case, SMR 2 is the best among four models. C3-MR is the worst model for energy input requirements in both Balanced and ES cases. Optimized mixed refrigerant conditions and key parameters for SMR 1 are given as follows:

Additional figures of merit beyond energy efficiency for these processes include exergy loss, heat exchanger efficiency, and both capital and operating costs. Each of these is discussed in the sections below.

4.1. Exergy theory and Exergy analysis of equipment

Exergy is closely related to availability and efficiency and measures the irreversible losses in a process, typically in the forms of losing capacity to do work. For a system operating at the same temperature as the heat rejection temperature, exergy and Gibbs energy are identical. If the system is operating at other temperatures, the definition of exergy is very similar to Gibbs energy, namely

$$E(T, P) = (H_1(T) - H_0(T)) - T_0(S_1(T, P) - S_0(T, P))$$

where H_0 and S_0 are the enthalpy and entropy at the equilibrium state, normally considered as the ambient temperature and pressure. This represents the maximum amount of useful work that can be produced (or the minimum work that needs to be supplied) as a system changes from its current state to an equilibrium state. The exergy consumed by each component in a process indicates the extent to which that component can be optimized for efficiency.

The equipment, including the heat exchanger, compressor, expander, water cooler, pumps and valves, causes irreversibility of the process leading to exergy losses. Li et al. [15] have introduced the exergy losses equations of the equipment, as is shown in Table 9. where ΔE_x represents the exergy losses of the equipment, E_{in} is the inlet exergy of the equipment, E_{out} the outlet exergy of the equipment, T_0 the ambient temperature, S_{in} the inlet entropy of the equipment, S_{out} the outlet entropy of the equipment, H_{in} the inlet enthalpy of the equipment, H_{out} the outlet enthalpy of the equipment, W_c the power consumption of the compressor, W_e the exporting work of the expander, Q the exchanged heat flow of the water cooler, ΔT the water temperature difference of the cooler, $T_{w_{in}}$ the temperature of water into the water cooler, $T_{w_{out}}$ the temperature of water out of the water cooler.

Table 10 summarizes the exergy changes associated with these processes. In a normal liquefaction process, the exergy losses of the water cooler are very large because the outlet temperature of the compressor is high, therefore it loses exergy large amounts of exergy when the gas is cooled down to a low temperature.

For C3-MR, the compression ratios for compressors are the highest (2.6) and contribute to highest exergy losses. The compressors contribute small exergy losses [18, 19] in all four processes because compression ratios are generally small. This can be improved by high efficiency, multi-stage, intercooled compression. The valves contribute less than 0.1% to the total exergy loss for SMR 1, MDMR and SMR 2. For C3-MR they contribute almost 5.5% of the total exergy losses. Pump and expander exergy losses are negligible for all four models and three cases. Turbine exergy losses for all models except C3-MR are less than 6 percent.

Regarding heat exchangers, it should be mentioned that all four models' exergy losses are lower than the ones reported in literatures [18, 19, 21]. It confirms high efficiencies for these heat exchangers and as it will be shown, temperature profiles are also consistent with this point.

Figure 8 illustrates the overall exergy loss for all four models. The MDMR model has the highest exergy loss for all three cases, consistent with its highest energy input requirements. In all cases, exergy loss is greatest during energy storage, consistent with the higher total energy demand, and lowest – approaching zero – during energy recovery, again consistent with a very low energy demand.

4.2. Heat exchangers' efficiency

Heat exchanger efficiency conceptually describes a change in availability, or thermodynamic ability to do work. Specifically, heat exchanger efficiency is the Gibbs energy difference in the

streams entering and leaving a heat exchanger normalized by the largest possible difference in this Gibbs energy. The largest possible difference occurs if all the streams come to the same temperature. These analyses assume there are no heat losses or gains between the exchanger and the environment. This assumption means the steady-state enthalpy of the incoming stream equals that of the outgoing stream. Therefore, the Gibbs energy difference becomes a difference in the product of the temperature and entropy. The steady-state mass flow of the incoming streams also equals that of the exiting streams, so the efficiency depends on the specific entropies and temperatures. The mathematical description becomes

$$\begin{aligned}\eta &= 1 - \frac{\sum_{outlet} m_i \hat{g}_i - \sum_{inlet} m_i \hat{g}_i}{\sum_{eq} m_i \hat{g}_i - \sum_{inlet} m_i \hat{g}_i} = 1 - \frac{(\Delta Ts)_{obs}}{(\Delta Ts)_{eq}} \\ &= 1 - \frac{\sum_{outlet} T_i m_i \hat{s}_i - \sum_{inlet} T_i m_i \hat{s}_i}{\sum_{eq} T_i m_i \hat{s}_i - \sum_{inlet} T_i m_i \hat{s}_i}\end{aligned}$$

where $(\Delta Ts)_{obs}$ represents the net change in the product of temperature and entropy observed in the streams, $(\Delta Ts)_{eq}$ represents the entropy change that would occur if the streams came to thermal equilibrium, $\sum_{outlet} T_i m_i \hat{s}_i$ represents the sum of the product of the temperature, mass flow rate and specific entropy summed over all outlet streams, $\sum_{inlet} T_i m_i \hat{s}_i$ represents a similar sum over all inlet streams, and $\sum_{eq} T_i m_i \hat{s}_i$ represents this product summed over outgoing streams at the equilibrium temperature, that is, the temperature that results from all streams reaching thermal equilibrium. This efficiency is unity if the inlet and outlet streams transfer heat with no temperature difference (ideal heat exchanger) and is zero if all outlet streams come to the same equilibrium temperature, which would be the poorest possible heat exchanger performance from an efficiency or entropy point of view.

Few definitions of heat exchanger efficiency exist and none is commonly accepted. Heat exchanger effectiveness, by contrast, indicates how rapidly or completely heat transfers from one

body to another and many authors define it. Occasionally, though rarely, authors use the term efficiency when they mean effectiveness. Efficiency provides a convenient measure of energy losses in process equipment and a useful and objective figure of merit for heat exchanger design and operation. It differs from exergy or availability, both of which use a heat rejection temperature. The heat rejection temperature provides a poor reference point for efficiency. Such efficiencies confusing depend strong on fluid inlet temperature rather than on design features dictate temperature differences between the inlet and outlet. A high-temperature system would exhibit a higher efficiency than the low-temperature system even if they perform identically in terms of heat exchange. The efficiencies defined by the equations above do not suffer from this problem.

Figure 9 summarizes the heat exchanger efficiencies for all four models. SMR 1 and SMR 2 have the highest efficiencies. For the balanced case, SMR 1 is better but for energy storage and energy recovery cases, SMR 2 is better.

There are three heat exchangers involved in the C3-MR process (the third one is the main heat exchanger summarized in the graph) and their efficiencies are as follows:

4.3. Heat exchange load and temperature distribution

The main energy loss of a natural gas liquefaction process occurs in compressors and heat exchangers. The comparison of heating and cooling curves in MDMR, C3-MR, SMR 1, and SMR 2 illustrate the temperature and heat transfer profiles for the exchangers. Individual heating and cooling curves for ER, ES, and Balanced (Figure 10) indicate where the major inefficiencies for each exchanger occur as the distance between the cold and hot stream temperature at each point. The pinch analysis theory [22] is adopted in the LNG heat exchanger.

The temperature difference and heat exchange load in heat transfer contribute to the exergy loss, so large temperature difference and heat exchange load primarily contribute to exergy loss in heat exchangers. If the hot composite curve matches the cold curve well, the resulting exergy loss is small. The MDMR model has the lowest temperature difference hence it has the lowest heat exchanger exergy loss. The results for exergy loss and heat exchanger efficiency agree with the temperature profiles.

4.4. Costing

Capital and operating costs represent the final figure of merit investigated here. Capital and operating costs of all systems depend as much or more on market demands and raw materials costs as on equipment size and type. These market-based fluctuations cannot be accurately anticipated. The approach used here relies on a consistent method to predict costs and to compare them across designs, with full recognition that actual costs may vary substantially based on unforeseen market shifts. The largest equipment size required by any one operating regime determines equipment costs. The largest size generally corresponds to energy storage. Heat exchangers, compressors and expanders are the most expensive units. Since HYSYS does not include a multi-stage compressor, several single-stage compressors with inter-stage cooling provide cost estimation. A single stage compressor with high pressure ratio is both more costly and less efficient. Total capital costs for C3-MR are the lowest (Figure 11) due to lower-pressure-ratio compressors. The SMR 1, SMR 2 and MDMR processes divide compression into a series of single-stage compressors. Operating costs in terms of electricity and water costs follow (Figure 12 and Figure 13).

Conclusions

Several natural gas liquefaction processes that provide coolant for the energy-storing version of cryogenic carbon captureTM differ in their overall energy demand, exergy loss, heat exchanger efficiencies, capital costs, and operating costs. The processes analyzed and optimized in detail included propane and mixed-refrigerant systems (C3MR), a modified dual mixed refrigerant system (MDMR), and two single mixed refrigerant systems (SMR 1 and SMR 2). Process simulations for three operation modes provide a basis for comparison:

- Energy storing, where LNG production increases to 40% more than is required by the CCC process, as would be used when excess energy is available on the grid,
- Balanced, where LNG production equals LNG demand from the power plant to run the CCC process, and
- Energy recovery, where LNG production decreases by 70% to decrease the parasitic loss and hence place more power on the grid when demand is low.

The optimal LNG energy demand ranges from 0.2 GJ_e/ton of LNG delivered to the CCC process and up during balanced operation. The SMR process has the lowest demand. During energy storage, energy demand increases significantly as excess LNG is produced. During energy recovery, energy demand decreases as stored LNG supplements the LNG produced from this process. The exergy analytical results show that the compressors and water coolers primarily contribute to total exergy loss. Measures to reduce the exergy loss are also explored. The SMR 1 process has the lowest energy demand and utilities' costs. The C3-MR process had lowest capital but highest operating costs. The SMR models have the highest heat exchanger efficiencies.

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Figure 1 Energy storage version of CCC process under normal (neither storing nor recovering energy) operation.

Figure 2 CCC process during energy storage operations. NG from the supply is pressurized and cooled prior to storage in the LNG storage at nominally -119.4 °C and 11.45 bar pressure. NG refrigerant follows the solid red/thick process lines.

Figure 5 a) Flow sheet of MDMR model b) HYSYS simulation of MDMR

a)

b)

Figure 6 a) Flow sheet of C3-MR model b) HYSYS simulation of C3-MR

Figure 7 Energy input requirements for four models

Figure 8 Exergy loss comparison for four models

Figure 9 Heat exchangers' efficiencies for four models

a)

b)

c)

d)

Figure 10 Temperature profiles for a) C3-MR; b) MDMR; c) SMR 2 and d) SMR 1, each operating at the three process conditions.

Figure 11 Total capital cost for four models

Figure 12 Electricity cost for each model

Figure 13 Water cost for each model

Table 1 Process parameters for the four processes

Parameters	Value	Notes
Feed gas pressure	3700 kPa	
Feed gas temperature	21 °C	
Feed gas flow rate	21200 kgmol/h	
Feed gas mole fraction components	CH ₄	0.95
	C ₂ H ₆	0.03
	C ₃ H ₈	0.02
LNG storage pressure	1145 kPa	
LNG temperature before expansion	-94 °C	
Liquefaction rate before expansion	100%	
NG temperature after expansion	-119.4 °C	
Liquefaction rate after expansion	73%	
Pressure drop in heat exchanger	5 kPa	To simplify the process
Pressure drop in water cooler	1 kPa	
Temperature after water cooler	21 °C	
Ambient temperature	25 °C	
The adiabatic efficiency of compressor	90%	[20]
The adiabatic efficiency of turbine	92%	[20]
Pressure ratio of each compressor	1-3	

Water's temperature from CCC	1.0561 °C
From CCC's temperature	-98.5 °C
From CCC's flow rate	21200 kgmol/h
The minimum approach temperature of heat exchanger	1-3 °C

Table 2 Specifications of NG common to all four models.

Conditions	From pipeline	To CCC (final conditions)
Vapor / Phase Fraction	1	0.27
Temperature [C]	21	-119.4
Pressure [kPa]	3700	1145

Table 3 a) key parameters and b) Mixed refrigerant conditions for three cases.

a)

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES

Vapor / Phase Fraction	0.43	0.023	0.00	0.23	0.086	0.10
Temperature [C]	30.00	20.5	16.12	-87.99	-93.191	-92.05
Pressure [kPa]	3704.65	3704.65	3704.65	108.4	108.394	108.40

b)

Component	Mole fraction
Methane	0.0418
Ethane	0.8461
Propane	0.0017
n-Butane	0.0053
n-Pentane	0.1051

Table 4 a) key parameters and b) warm mixed refrigerant composition for MDMR.

a)

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES
Vapor / Phase Fraction	1	1	1	1	1	1

Temperature [C]	24.97	16.90	10.83	-37.48	-17.85	1.244
Pressure [kPa]	194.1	194.1	194.1	189.1	189.1	189.1

b)

Component	Mole fraction
Methane	0.335
Ethane	0.255
Propane	0.410

Table 5 a) key parameters and b) cold mixed refrigerant composition for MDMR.

a)

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES
Vapor / Phase Fraction	0.11	0.1028	0.1028	0.79	1	1
Temperature [C]	-95.86	-96.52	-96.52	-82.15	-18.34	15.72
Pressure [kPa]	107.9	107.9	107.9	102.9	102.9	102.9

b)

Component	Mole fraction
Methane	0.064
Ethane	0.830
Propane	0.106

Table 6 a) key parameters and b) mixed refrigerant composition for C3-MR

a)

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES
Vapor / Phase Fraction	1	0.84	0.90	0.08	0.00	0.00
Temperature [C]	30.07	11.02	14.98	-102.8	-94.54	-95.18
Pressure [kPa]	2561	2561	2561	995.7	1398	1326

b)

Component	Mole fraction
Methane	0.3330

Ethane	0.4340
Propane	0.2050
n-Butane	0.0224
n-Pentane	0.0056

Table 7 Propane-precooled conditions

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES
Vapor / Phase Fraction	0.99	0.99	0.90	0.16	0.16	0.15
Temperature [C]	-3.18	-2.10	-0.19	-2.81	-1.74	0.15
Pressure [kPa]	429.14	443.85	470.71	434.14	448.85	475.71

Table 8 a) key parameters b) mixed refrigerant composition for SMR 1

a)

	Entrance			Loop		
Conditions	ER	B	ES	ER	B	ES
Vapor / Phase	0.00	0.00	0.00	0.06	0.08	0.09

Fraction						
Temperature [C]	22.13	21	16.15	-96.04	-93.19	-92.99
Pressure [kPa]	4500	4500	4500	108	108	108

b)

Component	Mole fraction
Methane	0.0423
Ethane	0.8451
Propane	0.0000
n-Butane	0.02813
n-Pentane	0.08447

Table 9 Exergy change expressions for differing types of equipment

Equipment	Exergy loss equation
Compressor	$\Delta E_x = E_{in} - E_{out} + W_c$
Expander	$\Delta E_x = E_{in} - E_{out} - W_c$
LNG heat exchanger	$\Delta E_x = \sum E_{in} - \sum E_{out}$
Water cooler	$\Delta E_x = E_{in} - E_{out}$
Valve	$\Delta E_x = T_0 (S_{out} - S_{in})$

Table 10 The exergy analytical results as a percents of total exergy loss for major equipment

	C3-MR			MDMR			SMR 1			SMR 2		
	ER	B	ES	ER	B	ES	ER	B	ES	ER	B	ES
LNG	24.91%	9.26%	5.85%	5.25%	0.72%	0.51%	12.67%	0.91%	1.74%	9.32%	1.00%	1.62%
Valves	5.25%	0.40%	0.35%	0.01%	0.01%	0.00%	0.04%	0.01%	0.01%	0.03%	0.01%	0.01%
Compressors	41.02%	29.64%	27.44%	24.58%	24.05%	20.28%	14.66%	17.49%	17.26%	15.56%	16.84%	16.57%
Water												
Coolers	15.92%	58.20%	64.34%	66.35%	68.76%	72.78%	66.61%	79.75%	79.50%	69.91%	80.37%	80.32%
Turbines	12.87%	2.40%	1.96%	0.47%	1.06%	0.95%	6.01%	1.83%	1.47%	5.16%	1.76%	1.48%
Pumps	0.04%	0.12%	0.08%	0.00%	0.01%	0.01%	0.03%	0.03%	0.03%	0.02%	0.03%	0.02%
Expanders	0.00%	0.00%	0.00%	3.35%	5.40%	5.49%	0.00%	0.00%	0.00%	0.00%	0.00%	0.00%

Table 11 C3-MR heat exchangers' efficiencies

Heat exchanger	Efficiency (η) %		
	Energy Recovery	Balanced	Energy Storage
1 st heat exchanger	77.28%	13.27%	96.65%
2 nd heat exchanger	20.58%	35.21%	42.14%