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# Techno-economic analysis of biomethane liquefaction processes

Biogas Utilization Opportunities in Ostrobothnia Region

Biokaasun hyödyntämismahdollisuudet Pohjanmaalla

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## Abstract

Heavy vehicle road transport produces about 40% of the transport sector's emissions annually in Finland. To achieve meaningful  $CO_2$  emission reductions, it is clear that heavy-duty transport will soon have to move towards alternative fuels as part of climate-sustainable transport. In this effort, biogas offers an environmentally friendly and 100 % renewable alternative, provided that there is a sufficient supply of high-quality and cost-competitive biogas and that the gas distribution is wide enough. For biogas to be a realistic alternative for heavy-duty vehicle fuel, in particular, the production of liquefied biogas and the network of liquefied gas filling stations needs to be increased.

This report investigates the feasibility of a new regional infrastructure for liquefied biogas in the Vaasa region. The key to the successful implementation of this kind of nanoscale liquefaction project is to find the optimal balance between technical, operational, and economic characteristics. The report provides a brief technical description of alternative liquefaction processes suitable for nanoscale (<10 tons per day) plants as well as examples of existing nanoscale liquefaction plants based on these technologies. The available commercial solutions for each technology are also reviewed. Hereafter, the various liquefaction processes are studied from an economic perspective. The life cycle costing (LCC) method was used to analyze the trade-offs between investment cost and future operating costs. To further compare the different processes, the levelized per-MWh costs of liquefaction were defined for each process. Finally, sensitivity analyses were performed to provide a broader view of the economic assessment results and address uncertainties related to investment costs and future expenditure flows. The case study was based on existing biogas production capacity in Ostrobothnia.

# Tiivistelmä

Raskas tieliikenne tuottaa Suomessa vuosittain noin 40 % liikennesektorin päästöistä. Hiilidioksidipäästövähennyksien saavuttamiseksi on selvää, että raskaan tieliikenteen on pian siirryttävä kohti vaihtoehtoisia käyttövoimia osana ilmastollisesti kestävää liikennettä. Biokaasu tarjoaa raskaalle liikenteelle ympäristöystävällisen ja täysin uusiutuvan vaihtoehtoisen polttoaineen edellyttäen, että korkeatasoista, kestävästi tuotettua ja kustannuksiltaan kilpailukykyistä biokaasua on saatavilla riittävän paljon ja kaasun jakelu on riittävän laajaa. Jotta biokaasun käyttö olisi realistinen vaihtoehto raskaan tieliikenteen käyttövoimaksi, on erityisesti nesteytetyn biokaasun tuotantoa ja kaasutankkausasemaverkostoa kasvatettava merkittävästi.

Tässä tutkimuksessa selvitetään uuden alueellisen nesteytetyn biokaasun infrastruktuurin toteutettavuutta Vaasan seudulle. Avain tällaisen nanomittakaavan nesteytyshankkeen onnistuneeseen toteuttamiseen on löytää optimaalinen tasapaino prosessin teknisten, operatiivisten ja taloudellisten ominaisuuksien välillä. Raportti tarjoaa lyhyen teknisen kuvauksen vaihtoehtoisista nanomittakaavan (<10 tonnia päivässä) laitoksiin soveltuvista nesteytysprosesseista sekä esimerkkejä jo olemassa olevista nanomittakaavan nesteytyslaitoksista. Myös saatavilla olevat kaupalliset ratkaisut kullekin tekniikalle kartoitettiin. Tämän jälkeen eri nesteytysprosesseja tutkittiin talouden näkökulmasta. Investointikustannusten ja aikaisten kokonaiskäyttökustannusten analysointiin elinkaaren käytettiin elinkaarikustannusmenetelmää. Lisäksi eri prosesseille laskettiin keskenään vertailukelpoiset megawattituntiperusteiset tuotantohinnat. Taloudellisen arvioinnin tuloksia laajennettiin lopuksi herkkyysanalyyseilla. Tapaustutkimus perustui Pohjanmaan olemassa olevaan biokaasun tuotantokapasiteettiin.

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# **1** Introduction

The role of natural gas as a fuel for heavy transport is growing with the technology development and new gas infrastructure. This also opens up the possibility for the biogas market to grow, especially in the form of liquefied biogas (LBG) [Gustafsson et al. 2020], also referred to as liquefied biomethane (LBM), or bio-LNG. A key advantage of LBM over compressed biogas (CBG) is its high energy density. LBM is three times more energy-dense, and spaceefficient, than CBG at 200 bar [Pellegrini et al. 2018]. This entails two significant advantages: The high energy density of LBM makes distribution efficient and economical compared to its gaseous counterpart – especially for the limited quantities, which are typical for biogas production plants [Capra et al. 2019]. More efficient transportation allows for a larger geographical marketing area. Besides, the high energy density makes LBM suitable for heavyduty vehicles, and even shipping [Gustafsson et al. 2020], thereby opening up new markets.

To be able to use biogas as transport fuel, it must first go through cleaning and upgrading processes (Fig. 1). Biogas cleaning is usually considered to be the first step in biogas treatment. Cleaning means the removal of minor unwanted components of biogas, such as H<sub>2</sub>O, hydrogen sulfide (H<sub>2</sub>S), siloxanes and halogenated compounds. The second treatment - upgrading - aims to increase the energy content of biogas by removing CO<sub>2</sub> [Bailón Allegue & Hinge 2012; Adnan et al. 2019]. The final product, biomethane, consists of nearly pure CH<sub>4</sub> (95–99 %) [Adnan et al. 2019]. After the cleaning and upgrading processes, the biomethane can be transformed into LBM. The focus of this work is on various biomethane liquefaction processes.



Figure 1. The major steps from raw biogas towards liquefied biomethane. Study boundary in red circle.

The two main ways to produce LBM are: (1) Conventional upgrading technology connected with a small-scale liquefaction plant, and (2) cryogenic upgrading technologies, where the purified gas is obtained directly at low temperatures [El Ghazzi & Tenkrat 2019; Pellegrini et al. 2018].

The most common way to produce LBM is to upgrade the raw biogas with conventional technologies, and then liquefy  $CH_4$  using small-scale liquefaction technology [El Ghazzi & Tenkrat 2019]. To prevent dry ice formation and corrosion in the downstream liquefaction

stage, the upgraded biogas stream prior to liquefaction has to satisfy the technical specifications reported in Table 1 [Capra et al. 2019].

Table 1. Quanty requirements for biomethane inqueraction [Capita et al. 2019].						
Component	Limit value	Issue				
CO <sub>2</sub>	50 ppm	Solidification on cold surfaces				
$H_2S$	1-4 ppm	Corrosion				
H <sub>2</sub> O	0.1–1 ppm	Ice formation on cold surfaces				

**Table 1.** Quality requirements for biomethane liquefaction [Capra et al. 2019].

If the upgrading process does not reach these requirements, an additional polishing step is needed before liquefaction [Johansson 2008].

Knowledge of liquefaction processes and refrigeration cycles is mature since it has been implemented in LNG plants for decades [Hashemi et al. 2019]. The three main processes commercialized for natural gas liquefaction purposes are 1) cascade liquefaction processes, 2) mixed refrigerant liquefaction processes, and 3) expander liquefaction processes [Ancona et al. 2017; Cao et al. 2016; Hashemi et al. 2019].

The liquefaction process of biomethane is, in principle, similar to that of natural gas [Capra et al. 2019]. The two main differences are:

- 1. Fluid composition: Raw natural gas is a hydrocarbon gas mixture consisting primarily of methane (CH<sub>4</sub>), but commonly containing varying amounts of other higher hydrocarbons such as ethane ( $C_2H_6$ ), propane ( $C_3H_8$ ), butanes ( $C_4H_{10}$ ) and pentanes. For this reason, the condensation of natural gas occurs at varying temperatures, while biomethane almost pure methane condenses at a nearly constant temperature.
- 2. Plant size. The capacity of existing large natural gas liquefaction plants ranges from one to almost 8 million tons per year [Tractebel Engineering 2015]. The size of biomethane liquefaction plants is significantly smaller, from 0.001 and 0.01 Mt/year [Capra et al. 2019]. Therefore, we need to introduce another scale for gas liquefaction [Cryonorm 2020a]:
  - Small-scale <500 tons per day (182,500 TPA)
  - Micro-scale <75 tons per day (27,000 TPA)
  - Nanoscale <10 tons per day (3,650 TPA)

The three main liquefaction processes mentioned above differ, e.g., in terms of complexity, liquefaction capacity and performance [Cao et al. 2016]. The most energy-efficient liquefaction process is the cascade system, operating with different pure refrigerants in three refrigeration cycles. As a result, the cascade process requires a high equipment count and capital cost, and is therefore not suitable for small-scale applications [Nguyen et al. 2017]. The mixed refrigerant process, consisting of a single cooling cycle with a mixture of different refrigerants, is more straightforward and has lower capital costs than the cascade system [Yin et al 2008]. The expansion processes are compact and simple, but they are generally less efficient than mixed refrigerant processes [Yin et al. 2008; Nguyen et al. 2017].

A major challenge for the LBM liquefaction process is the scale. In fact, the most appropriate method for small-scale liquefaction plants may differ significantly from those used in large-scale applications, as these techniques are neither practical nor economical when applied to small plants [Baccioli et al. 2018]. The specific power consumption per unit of LBM plays an important role, but other factors such as the size and compactness and the ease of operation and maintenance, are also crucial [Nguyen et al. 2017].

This paper presents a technical analysis of liquefaction processes suitable for nanoscale (<10 tons per day) applications. Viable technologies for this type of application could be limited to the following categories:

- 1. N2 expander layouts built on different configurations of reverse Brayton cycle
- 2. Rankine cycle with mixed refrigerant processes
- 3. Linde cycle
- 4. Stirling refrigeration
- 5. Cryogenic liquid vaporization

A brief technical description of each technology is presented, as well as examples of existing nanoscale liquefaction plants based on these technologies. Moreover, available commercial solutions of each technology are reviewed. Additionally, cryogenic upgrading technology, where the purified gas is obtained directly at low temperatures, is briefly discussed. Finally, the study presents a comparative economic analysis of different liquefaction processes.

# 2 Liquefaction technologies

## 2.1 Nitrogen expander processes (Reverse Brayton cycle)

 $N_2$  expander layouts are built on different configurations (e.g., single or dual expansion process, with or without a pre-cooling cycle) of reverse Brayton cycles. The process works by compressing the gaseous refrigerant and then cooling and expanding it to produce temperatures low enough to liquefy the feed gas [Tractebel Engineering 2015]. The most common working fluid is nitrogen. Reverse Brayton cycles are typically proposed with one or two expanders [Capra et al. 2019].

## 2.1.1 Single N<sub>2</sub> expander liquefaction process

The single nitrogen expander is the simplest configuration among expander-based technologies [Khan et al. 2017]. In this process,  $N_2$  provides the required refrigeration for the entire temperature range of the process, including the pre-cooling, liquefaction, and sub-cooling sections [Roberts et al. 2015]. The operating principle is shown in Fig. 2a: The pure  $N_2$  refrigerant is compressed and following this cooled in the after-cooler and the main heat exchanger, and then expands to low pressure and low temperature. The  $N_2$  working fluid maintains a gaseous state throughout the process. [Roberts et al. 2015].



Figure 2. Single-expander N<sub>2</sub> Brayton cycles [Roberts et al. 2015].

To make the single-expander  $N_2$  process more efficient, a portion of the high-pressure  $N_2$  can be taken prior to the expander, further cooled and liquefied, and expanded through a Joule-Thomson valve. In this configuration (Fig. 2b), some of the refrigeration for the sub-cooling step comes from vaporizing liquid  $N_2$  [Roberts et al. 2015].

The obvious disadvantage of the single  $N_2$  expander process lies in the expansion of the entire working fluid to the lowest temperature, although most is required at higher temperatures. This introduces large temperature differences between the refrigerant and the feed gas, causing high compression energy requirements [Khan et al. 2017]. This shortcoming can be remedied by implementing additional levels of expansion, allowing more dedicated refrigeration to each of the temperature ranges [Roberts et al. 2015].

## 2.1.2 Dual N<sub>2</sub> expander liquefaction process

In the dual  $N_2$  expander process, the introduction of the second expander allows splitting the working fluid and causing the expansion of only required part to the lowest pressure, thus saving compression energy [Khan et al. 2017]. Fig. 3 describes one such process: A warm expander provides refrigeration at pre-cooling and liquefaction steps, while the second – a cold expander – provides sub-cooling refrigeration [Roberts et al. 2015].



Figure 3. Dual N<sub>2</sub> expander liquefaction process [Roberts et al. 2015].

The  $N_2$  expansion liquefaction process is considered a suitable process for a small-scale liquefaction plants due to its fast start-up, simplicity and convenient maintenance [He & Ju 2014]. However, although nitrogen is an effective refrigerant in cryogenic applications, its efficiency at higher temperature levels of the liquefaction process is poor. Therefore, many nitrogen cycles include a pre-cooling unit that provides refrigeration duty at these higher temperature levels [Kohler et al. 2014]. Adding a pre-cooling cycle, e.g., with propane, CO<sub>2</sub>, or ammonia as a refrigerant to the process could reduce power consumption by 15–35 % [He & Ju 2014; Khan et al. 2017; Kohler et al. 2014; Zhang et al. 2020a]. Two variations are described in the following sections.

## 2.1.3 N<sub>2</sub>-CO<sub>2</sub> single expander liquefaction process

The N<sub>2</sub>-CO<sub>2</sub> single expander process is an improvement over the single N<sub>2</sub> expander process discussed above. In this process, two separate refrigerant loops are used: CO<sub>2</sub> for pre-cooling and N<sub>2</sub> for liquefaction and sub-cooling. The use of CO<sub>2</sub> pre-cooling improves system efficiency by reducing the specific compression power demand by 33 % [Khan et al. 2017]. However, this efficiency increase must be weighed against the increase in cost and complexity [Tractebel Engineering 2015] as well as the potential impact on operability and reliability.

## 2.1.4 N<sub>2</sub>-CO<sub>2</sub> dual expander liquefaction process

The  $N_2$ -CO<sub>2</sub> dual expander is an advancement over the dual  $N_2$  expander process. Both technologies employ the two-stage expansion of nitrogen at different pressure levels. CO<sub>2</sub> precooling lowers the compression power requirements by 17 % compared to dual  $N_2$  expander process [Khan et al. 2017]. Again, an increase in efficiency must be weighed against the cost and complexity increase.

The various configurations of expansion-based layouts based on inverted Brayton cycles are compact, simple, and inherently safe if inert fluid, such as nitrogen, is used as the refrigerant medium [Roberts et al. 2015; Tybirk et al. 2018]. Besides, the ease of operation of the N<sub>2</sub> cycle is an important factor. The N<sub>2</sub> expander cycle is straightforward for operating staff to understand, manage and troubleshoot, as the process requires less monitoring and control points and minimal operator intervention compared to, e.g., mixed refrigerant (MR) processes [Pak 2013].The working fluid is in gaseous phase, which avoids instabilities and maldistribution issues in the heat exchangers [Nguyen et al. 2018]. In addition, N<sub>2</sub> as a cooling medium can be produced on site directly from the air, which eliminates the import and storage of hydrocarbon refrigerants. The down side is that they are generally less efficient than mixed refrigerant processes [Nguyen et al. 2017], whose cooling capacity is based on a mixture of hydrocarbons and nitrogen.

One of the world's first LBM plants, Lidköping biogas plant in Sweden, is based on dual  $N_2$  expander liquefaction technology.

# 2.1.5 Existing small scale liquefaction plants based on $N_2$ expander technology: Lidköping biogas plant, Sweden

Lidköping's biogas plant is a close collaboration between Gasum AB and FordonsGas Sverige AB. The collaboration consists of Gasum operating the biogas plant and being responsible for biogas production and upgrading. The biogas is upgraded in accordance with the Swedish standard for biogas as a vehicle fuel (97 % CH<sub>4</sub>) in a water scrubber. After upgrading, FordonsGas Sverige AB – a part of the Air Liquide Group – takes and liquefies the biomethane into LBM for regional distribution. A small portion of biomethane is compressed for local distribution. The amount of biomethane produced is 65 GWh/year [Gasum 2020].

A majority (88 %) of the bio-CH<sub>4</sub> produced is liquefied in the condensation plant (Fig. 4, red rectangle). The technology is supplied by Air Liquide, and the design capacity of the liquefaction plant is 13 TPD [Air Liquide 2013]. To liquefy the biomethane, most of the remaining CO<sub>2</sub> is first removed by pressure temperature swing absorption (PTSA) (CO<sub>2</sub><10 ppm). After this extra polishing step, the treated bio-CH<sub>4</sub> is cooled down using a 2-stage Brayton cycle and then condensed in a plate-fin heat exchanger (Fig. 5) [Rouaud 2017]. The technology allows liquefaction in the span of -140 °C (at 4 barg) to -161 °C (at atmospheric pressure) [Lidköping Biogas 2020]. The LBM produced is then sent to a cryogenic 115 m<sup>3</sup>, 20 meters tall LBM storage tank. The distributer, FordonsGas Sverige AB, fills 50 m<sup>3</sup> tanker

trucks every second day and transports the gas to filling stations, e.g., in Göteborg [Lidköping Biogas 2020].



**Figure 4.** Air Liquide biomethane liquefaction plant (13 TPD) in Lidköping, Sweden [Air Liquide 2013].





### 2.2 Rankine cycle with mixed refrigerant processes

#### 2.2.1 Single mixed refrigerant process

In mixed refrigerant processes, the refrigerant is a mixture of several compounds, mainly hydrocarbons with low boiling points and nitrogen [Mokhatab et al. 2014]. The evaporation process takes place over a temperature glide rather than at a single temperature point as with

refrigerants of pure components. It is therefore possible to tune the refrigerant composition so that its evaporation curve matches the cooling curve of the feed gas from ambient to cryogenic temperatures [Nguyen et al. 2018]. Refrigeration is always being provided at the warmest possible temperature, resulting in better thermal efficiency.

In the single mixed refrigerant (SMR) process, the feed gas is pre-cooled, liquefied and subcooled in a single cryogenic three-flow heat exchanger. Figure 6 shows the schematic diagram of the SMR liquefaction process.



Figure 6. Process flowsheet of the SMR liquefaction process [Khan et al. 2017].

The refrigeration process follows the reverse Rankine cycle: compression-coolingcondensation-expansion-evaporation [Mokhatab et al. 2014]. The working fluid is compressed in the vapor phase from the evaporation to the condensation pressure (1 to 2), cooled and condensed to subcooled liquid (2 to 3), and then throttled through an adiabatic valve device (Joule-Thomson valve) (4 to 5). It is then redirected to a heat exchanger where it is fully evaporated to provide the refrigeration effect (5 to 1) [Capra et al. 2019].

SMR is widely used in cryogenic processes for small-scale LNG applications due to its compactness and small footprint [Qyyum et al. 2018]. The refrigerant process with phase changes reduces equipment and piping size compared to  $N_2$  loop [Wärtsilä 2020a]. The power consumption of this process is also lower than the  $N_2$  expander cycle one [Ancona et al. 2020]. The efficiency advantage of the mixed-refrigerant process is two-folded. The refrigerant composition and temperature glide can be tuned to thermally match the feed gas composition. In addition, the working fluid is mostly in two-phase conditions, and latent heat can be exploited throughout most of the process [Nguyen et al. 2018].

### 2.2.2 Dual or pre-cooled mixed refrigerant processes

In large-scale applications, numerous versions of cascades of Reverse Rankine cycles, such as the propane pre-cooled mixed refrigerant ( $C_3MR$ ) cycle, or dual-stage cooling cycles, are widely employed. Figure 7 shows the basic schematic of  $C_3MR$  process. The process includes two refrigeration cycles. The first cycle consists of a propane refrigeration cycle, and the other one is a mixed refrigerant (MR) refrigeration cycle. The propane pre-cooling refrigeration cycle cools the feed gas and the second refrigerant down to -30–40°C range. The MR refrigeration

cycle ensures liquefaction and sub-cooling to -162°C in multi-stream heat exchangers [He et al. 2018, Nguyen et al. 2017].



Figure 7. Schematic of C<sub>3</sub>MR liquefaction process [He et al. 2018].

The existence of a propane pre-cooling cycle can eliminate the large temperature difference at the warm end of the heat exchanger [He et al. 2018]. Therefore, such setups can achieve higher system efficiencies than layouts with only one single refrigerant [Nguyen et al. 2017]. The downsides of this layout are the higher complexity of the process and the high equipment count and capital cost compared to simple configurations.

# 2.2.3 Existing small scale liquefaction plants based on Mixed Refrigerant technology: EGE Biogas, Norway

The EGE biogas plant is located in Nes, Romerike, an agricultural region close to Oslo. The plant, operated by Cambi AS on behalf of the plant owner EGE Energigjenvinningsetaten, produces biomethane from food waste to be used as biofuel for public transport in Oslo. The production capacity of the EGE Biogas plant is around 14,000 Nm<sup>3</sup> bio-CH<sub>4</sub> per day. [Wärtsilä 2020b]

The biomethane liquefaction plant (Fig. 8) with a capacity of 11 TPD, was delivered by Wärtsilä in 2013. The pretreatment at EGE consists of two stages; water scrubbing and  $CO_2$  polishing [Sherrard 2014].



Figure 8. The EGE biogas liquefaction plant (modified from Sherrard 2014).

The installation at EGE is based on Wärtsilä's NewMR (mixed refrigerant) technology (Fig. 9). A glycol pre-cooling system is used to cool  $(-10^{\circ}C)$  both the clean gas and the mixed refrigerant to improve energy efficiency and to ensure stable operation conditions independent of ambient variations.



Figure 9. A schematic of Wärtsilä MR Liquefaction Plant [Siggberg 2017].

The MR side operates according to the following principles: The compressed MR is cooled to a partially liquefied state, and then split in the separator into a liquid fraction and a vapor fraction before being transferred to a cryogenic heat exchanger (Cold Box). In the Cold Box, part of the mixed refrigerant stream is used to cool the MR itself. The clean pre-cooled bio-CH<sub>4</sub> transfers its heat to the cooled two-phase MR (gas and liquid) in a counter flow until it condenses, and exits as LBM from the bottom part of the Cold Box at a temperature of -155°C to -162°C. Before being stored in a 180 m<sup>3</sup> storage tank, LBM is throttled to a lower pressure according to the storage tank pressure. [Siggberg 2017]

The pre-cooler and MR compressor are equipped with a variable frequency driver, also giving efficient operation at part load [Siggberg 2017]. According to Wärtsilä, the plant is designed to be fully automatic with uncrewed operations requiring only electricity as an energy source. Modular, containerized plug and play engineering results in low investment costs and low operational expenses thanks to simple unmanned operation and low power consumption. The technology can be scaled up to a capacity of 80 tons per day [Wärtsilä 2016].

In 2017, Wärtsilä provided similar technology for the Nordics' largest LBM facility (25 TPD) located in Skogn, Norway. Furthermore, Wärtsilä has been awarded a contract for a LBM plant in Asker, Norway, with a capacity of 20 tons per day. The plant is expected to become in commercial operation during 2020. Moreover, a new LBM plant in Linköping, Sweden, will be supplied by Wärtsilä. The plant is scheduled to start up during 2020. Initially, it is projected to produce volumes of 60 GWh per year (12 TPD), and gradually ramping up to a full capacity of 85–90 GWh/year (17–18 TPD).

The literature shows that the mixed-refrigerant and expander-based processes dominate the small-scale LNG industry in Nordic countries. Other technologies suitable for micro-scale production include Linde cycle, Stirling refrigeration and liquid nitrogen vaporization. These liquefaction techniques are described in the following Sections.

## 2.3 Linde cycle

## 2.3.1 Technology overview

In an open Linde cycle, the gas being liquefied, i.e., CH<sub>4</sub> itself, is used as the working fluid, and a throttling process is used to liquefy the gas. The principle flow diagram is shown in Figure 10. The CH<sub>4</sub> is compressed from ambient conditions to a pressure up to 200–300 bar [Tybirk et al. 2017]. This high compression ratio may require several compression and cooling steps (3 in this example) [Zare 2016]. The high-pressure gas then passes through the cryogenic heat exchanger, where it is pre-cooled by the return stream of cold, low-pressure gas. Finally, the cold high-pressure gas is expanded through a Joule–Thomson valve to the desired pressure level, typically 2–3 bar [Tybirk et al. 2017]. At the exit of the valve, the flow is in the two-phase (liquid–vapor) region [Windmeier & Barron 2013]. The liquid phase is collected in the liquid receiver. Uncondensed gas is recirculated and mixed with the feed gas to replace the condensed product and returned to the compressors to complete a new trip through the cycle.



Figure 10. An open Linde cycle, modified from [Zare 2016].

# 2.3.2 Existing small scale liquefaction solutions based on Linde technology: Galileo Cryobox®

The Galileo Cryobox® liquefaction technology is based on the multi-stage gas compression and expansion process described above. The high-pressure thermodynamic cycle of the Cryobox® converts feed gas (CH<sub>4</sub>) into a liquid state by cooling its temperature below -153°C. The process includes an automatic boil-off recovery system eliminating the venting [Galileo Technologies 2017]. The Cryobox® is available in two sizes (15 and 4.8 TPD). Several trailer size modules (Fig. 11) can be connected in parallel if the capacity requirement is higher [Tybirk et al. 2017].

Cryobox® is delivered for outdoor operation with no need for building. The compact, preassembled modular design allows easy transport and fast on-site mounting minimizing construction costs. The entire process is run by a single simple compressor in the Cryobox®, with a minimum of rotating parts to maintain. The operation runs completely unmanned and includes 24/7 remote monitoring via Galileo Technologies SCADA System. [Galileo Technologies 2017; Tybirk et al. 2017].



Figure 11. Cryobox® LNG production station [Galileo Technologies 2017].

Besides producing LNG/LBM, Cryobox® provides compressed gas as needed. In this way, both fuels are simultaneously available, for example, CBG for light-duty vehicles and LBG for high heavy-duty applications [Galileo Technologies 2017].

Galileo Technologies debuted the Cryobox® as a multi-unit, dual fuel (CNG and LNG) production plant serving Buquebus, the oceanic ferry company in Argentina. In 2015, Galileo Cryobox® technology was delivered to Terra Energy for flaring mitigation in North Dakota's Bakken shale region, converting the wellhead gas into more than 13 tons of LNG per day.

## 2.4 Stirling refrigeration

## 2.4.1 Technology overview

The cooling power of a Stirling-type cooler is created by the reverse Stirling cycle, i.e. compression and expansion of the working fluid in a closed cycle by mechanical pistons [Stirling Cryogenics 2016]. The basic type of Stirling cooler is illustrated in Figure 12.



Figure 12. Schematic diagram of a Stirling-type cooler.

The refrigeration cycle consists of two constant volume processes and two isothermal processes [Capra et al. 2017]:

- 1. An isothermal compression at ambient temperature, with heat transfer to the surroundings
- 2. Regenerative cooling at constant volume where heat is transferred to a solid regenerator
- 3. Isothermal expansion to provide the useful cooling power
- 4. Regenerative heating at constant volume, with heat transfer from the solid regenerator to the working fluid.

At the end of the fourth step, the state of the cooler is the same as in the beginning, and the cycle is repeated. The feed gas flows through the cold heat exchanger, where energy is extracted and the gas will liquefy. The typical working fluid in Stirling coolers is Helium (He).

## 2.4.2 Existing small scale liquefaction solutions based on Stirling technology: StirLNG

There are a limited number of Stirling refrigeration applications in the field of methane liquefaction. One such is shown in Fig. 13: StirLNG manufactured by Stirling Cryogenics. These small and compact cryocoolers are modified to produce micro-scale LNG/bio-LNG,

typically 200–15,000 kg/day. The biggest drawback is that only small capacity units are available; achieving larger sizes requires the use of multiple units in parallel, preventing economies of scale. In addition, in long-term continuous use, they may be less maintainable than the more common reverse Brayton and Rankine cycles [Capra et al. 2017].



Figure 13. Stirling Cryogenics LNG systems [Stirling Cryogenics 2016].

Integration of the Stirling Cryogenerator into the process is straightforward. Since the cycle to produce cooling power is completely separate, there is only need for in- and outlet lines to be connected, supplying the flow of gas to be liquefied [Stirling Cryogenics 2020b].

An integrated biomethane conditioning and micro-liquefaction plant relying on Stirling-technology was introduced in 2018 in Foggia, Italy (Fig. 14).



Figure 14. Micro-scale LNG production [Stirling Cryogenics 2019].

More references of Stirling cryogenerators can be found in boil-off gas (BOG) re-liquefying applications, e.g. on an LNG bunker barge in Jacksonville, USA. To manage the BOG, six StirLNG-4 units with a total capacity of 5.4 TPD are placed on top of the LNG tank. The StirLNG-4 units will liquefy the methane gas that flows back from the containership's tank when being filled with LNG [Stirling Cryogenics 2020c].

## 2.5 Cryogenic liquid vaporization

In  $LN_2$  vaporization, the cooling duty is provided by liquid nitrogen ( $LN_2$ ), which is produced outside the biomethane production plant.  $LN_2$  vaporization is functionally the simplest and less capital-intensive option; it only requires installing a heat exchanger and a liquid nitrogen tank [Capra et al. 2017]. A simplified diagram of the combination of gas liquefaction and  $LN_2$ evaporation system is shown in Figure 15, and Figure 16 presents a possible layout of a 12 TPD liquefaction plant.



Figure 15. LN<sub>2</sub> vaporization [Capra et al. 2017].



Figure 16. Cryotec's LNG E-LIN plant layout, capacity 12 TPD [Cryotec 2014].

 $LN_2$  vaporization is suitable for plants with low capacity and availability of liquid nitrogen [Cryotec 2014]. The liquid nitrogen consumption equals 2.5 kg  $LN_2$  per kg biomethane [SIAD 2018a]. The advantages of  $LN_2$  vaporization are:

- High level of safety using an inert gas as cooling factor
- No need for liquid hydrocarbons storage systems for refrigerant mixture
- Simple plant management process and control

Technologies described in Chapters 2.1-2.5 are suitable for the further processing of CH<sub>4</sub> produced by conventional upgrading technologies. Cryogenic upgrading represents another approach, in which the purified gas is obtained directly at low temperatures.

## 2.6 Cryogenic biogas upgrading and liquefaction

In cryogenic upgrading,  $CO_2$  and other unwanted components are separated from the gas flow through condensation. The process is performed in a series of successive temperature reductions, and  $CO_2$  and other impurities are steadily removed from the gas flow as per their boiling points [Kapoor et al. 2019]. The final product, almost pure bio-CH<sub>4</sub>, is then liquefied. An advantage of this process is that the purified gas is obtained directly at low temperatures, which reduces the cooling requirement in LBM production [Hashemi et al. 2019]. Another advantage of using cryogenic upgrading technology is that  $CO_2$  is obtained as a clean liquid by-product to be used in further applications [Pellegrini et al. 2018]. Figure 17 shows an integrated cryogenic upgrading and liquefaction system developed by Cryo Pur.

- 1. The incoming raw biogas is first treated with activated carbon filters to remove  $H_2S$ . Biogas is then cooled in two steps (-40°C and -75°C) to remove water. VOCs and siloxanes are removed together with water.
- 2. The dry, pretreated biogas is further cooled to -120°C and carbon dioxide is recovered to ensure that the biomethane reaches the required purity for liquefaction. During this step, pure CO<sub>2</sub> is recovered in liquid form, forming a valuable by-product.
- 3. The almost pure (>99 %) bio-CH<sub>4</sub> is then compressed and liquefied, and finally stored in a cryogenic vessel. [Cryo Pur 2020]





Cryo Pur technology is being adapted for biogas projects with flows ranging from 200 Nm<sup>3</sup>/h to 2,000 Nm<sup>3</sup>/h raw biogas [Cryo Pur 2020]. Electricity consumption for the whole process (pretreatment–upgrading–liquefaction) is 1.4–1.77 kWh/kg LBM [Capra et al. 2019].

The first commercial Cryo Pur plant, with a capacity of 3 TPD, was built in 2017 at Greenville Energy's site in Northern Ireland. In 2019, Cryo Pur launched the design of a 7.5 TPD unit for project in France.

Cryogenic upgrading may be a viable option especially for new plants with no existing upgrading facility in place.

# **3** Technical aspects for nanoscale LBM production

## 3.1 Key project objectives

The key project objectives for nanoscale LBM production are simple design, compactness, ease of operation and low costs. The technical characteristics that should be taken into account when choosing the most appropriate technology are:

- Process efficiency
- Process safety
- Operability and maintainability
- Easy optimization in changing process conditions
- Refrigerant availability
- Technology maturity
- Space requirements
- Scalability / modular design approach

## 3.2 Commercially available liquefaction technologies for nanoscale LBM production

Typically, biogas is produced in small quantities locally, so LBM production requires microor nanoscale liquefaction plants. A list of commercially available liquefaction technologies for applications <10 TPD production are reported in Table 2, with details of technology providers, main features of the process, and the main technical specifications.

Technology	Technology provider / Commercial	Production Capacity	Specific energy consumption	Footprint	Remarks	Ref.
	name					
Brayton cycle	Air Liquide / Turbo-Brayton cryogenic systems • TBF-175 • TBF-350 Turbo-Brayton with double expanders	4.8 TPD 12 TPD 13 TPD	0.93 kWh/kg 0.78 kWh/kg 0.9 kWh/kg	9.5x1.7x3m Weight 15 t 11x1.7x3m Weight 17 t	<ul> <li>N<sub>2</sub> Brayton cycle, non-hydrocarbon refrigerant improving safety</li> <li>Delivered mounted on a chassis, ready-to-run package.</li> <li>Low utilities: only water and electricity.</li> <li>Can also be designed with specific architectures (containerized system, air-cooled system)</li> <li>Suited to both atmospheric and pressurized storage</li> <li>Simple and understandable process, easy to use and maintain.</li> <li>An experienced Air Liquide industry organization to provide after-sales services.</li> </ul>	Air Liquide 2020; Air Liquide Nordic 2013; Rouaud 2017
Rankine cycle	Wärtsilä / NewMR	5.5 TPD – up to 80 TPD Standard sizes: 10, 17 and 25 TPD	>0.70 kWh/kg	15 x 15 m, includes access and maintenance space	<ul> <li>Single Mixed Refrigerant (SMR) with water-glycol pre-cooler.</li> <li>Closed system with zero refrigerant loss. The MR fluid requires careful handling, but the refrigerant is contained within the process thanks to the fully closed loop</li> <li>Compact and modularized, plug-and-play design</li> <li>Simple energy supply, only electric power needed</li> <li>Advanced technology requires skilled personnel (training and support available)</li> <li>Robust and reliable technology</li> <li>Designed for unmanned operation</li> </ul>	Wärtsilä 2016; Wärtsilä 2020a; Tybirk et al. 2018

**Table 2.** A list of commercially available nanoscale liquefaction solutions, and the key technical specifications.

Technology	Technology	Production	Specific energy	Footprint	Remarks	Ref.
	provider / Commercial name	Capacity	consumption			
Linde cycle	Ecospray Technologies / ECO-µLNG	1–20 TPD Possibility of size customization	0.7-0.9 kWh/kg (at 3 barg)	10.0 x 2.5 x 2.5 m	<ul> <li>Linde cycle with pre-cooling</li> <li>No use of cooling media (N<sub>2</sub> or MR)</li> <li>Pre-assembled on skids for fast installation</li> <li>Adaptable for all needs</li> </ul>	Ecospray 2019
	Galileo Technologies / Cryobox Cryobox-Bio (integrated biogas upgrading and liquefaction station)	15 TPD 4.8 TPD	0.7 kWh/kg (0.66–0.85 kWh/kg, depending on inlet pressure)	14.1 x 3.0 x 1.95 m	<ul> <li>Linde cycle with pre-cooling</li> <li>Dual-mode capability: LBG and CBG simultaneously available.</li> <li>Delivered for outdoor operation with no need for building. Simple transport and on-site mounting.</li> <li>Modular, plug and play design: The installation requires only concrete pads and connection to gas source, electricity, and compressed air</li> <li>Scalability: installation can grow according to demand changes.</li> <li>Boil-off free (automatic boil-off recovery system)</li> <li>Short start time</li> <li>Fully automatized process with distant surveillance</li> </ul>	Galileo 2017; Verdek 2018; Himmel- strup 2019
Stirling cryocooler	Stirling Cryogenics / StirLNG-16	2-5 TPD, depending on feed gas pressure (0-20 barg)	0.43–1.45 kWh/kg (depending on inlet pressure, e.g. at 2 barg 1.03 kWh/kg)	5.90 x 2.35 x 2.20 m Weight 6,000 kg	<ul> <li>Plug and play design: All equipment installed, aligned and pre-wired on a skid. The only required connections are process lines, cooling water and power.</li> <li>Robust, stand-alone system, easy to operate, low amount of operator involvement</li> <li>Explosion proof classification ATEX 2 or 1</li> </ul>	Stirling Cryogenics 2020a; Tybirk et al. 2018
	Cryonorm / Cryocooler	5 TPD			<ul> <li>Containerized design, easy to install</li> <li>Standard design for 5 TPD, LBM storage volumes flexible and to be discussed with the customer.</li> </ul>	Cryonorm 2020b

Technology	Technology provider / Commercial	Production Capacity	Specific energy consumption	Footprint	Remarks	Ref.
	name					
LN <sub>2</sub> vaporization	SIAD Macchine Impianti / Smart LIN-LNG	2 TPD – up to 25 TPD	0.07 kWh/kg		<ul> <li>A micro capacity plant that uses liquid nitrogen as a refrigerant. LN<sub>2</sub> is brought to the plant with road tankers.</li> <li>LN<sub>2</sub> storage tanks for 3 days endurance required</li> <li>High level of safety using an inert gas as cooling factor</li> <li>Simple plant management process and control</li> </ul>	SIAD, 2018a, 2018b
	Cryotec /					
	LNG-E-LIN	Standard capacity 12 or 24 TPD Tailor-made solutions provided to suit specific capacity needs.	Very low energy consumption		<ul> <li>Process with liquid nitrogen evaporation for liquefaction.</li> <li>Process is designed in modular units – scalability.</li> <li>Suitable for plants with low capacity and availability of liquid nitrogen</li> <li>High level of safety using an inert gas as cooling factor</li> <li>Simple plant management process and control</li> </ul>	Cryotec 2014

# **4 Economic aspects**

This section provides an economic analysis of the liquefaction processes presented in Chapters 2.1 to 2.5. An economic analysis of cryogenic upgrading is excluded because this study focuses on liquefaction technologies that are to be combined with an existing upgrading facility. The specific capital costs are harmonized first, followed by the operating costs. Operating expenses (O&M) in this chapter include energy costs and maintenance costs. Cost data were collected from academic literature and publicly available technical reports. All costs mentioned in this paper are indexed to EUR<sub>2019</sub> currency.

## 4.1 Capital costs

Capital investment includes the main equipment (refrigeration compressors and drivers, cryogenic heat exchangers, power and control systems), auxiliary equipment, installation, and indirect costs (engineering, freight charges, taxes and insurance). Table 3 summarizes the available investment cost data for liquefaction capacities <15 TPD. Most studies considered only the liquefaction system, excluding, e.g., feed gas pre-treatment, cryogenic storage system, and the owner's costs, such as feasibility studies, commercial contracts, negotiations with financiers, and approval bodies for permitting.

In addition to the total capital costs, specific capital costs are presented, which facilitates comparison between different technologies. The specific capital costs ( $\epsilon$ /TPA) are calculated as total capital costs divided by the plant capacity. A graphical summary of the specific capital costs collected and calculated in this study is shown in Figure 18. The average specific capital costs for liquefaction plants with production capacities <15 TPD ranged from 440–1350  $\epsilon$ /TPA. The LN<sub>2</sub> vaporization and SMR processes cost was at the lower end of the price range, while the Linde cycle represented the upper end of the price range. The average specific costs of N<sub>2</sub> expander based technologies, pre-cooled MR process and Stirling refrigeration ranged from 760–1090  $\epsilon$ /TPA.



Figure 18. Specific capital costs for liquefaction capacities <15 TPD.

# Academic literature

	Plant	Reverse Br	ayton cycle	Rankine	cycle with MR	Linde	Stirling	Liquid	
	capacity /	Single N <sub>2</sub>	Dual N <sub>2</sub>		Precooled or	cycle	refrigeration	(LN <sub>2</sub> )	Ref.
Cost	notes	expander	expander	SMR	dual MR			vaporization	
Liquefaction system - Compressors - Turboexpander - Cold box		0.127							
Control and instrumentation	3 TPD	0.096							Fan et al. 2009
Auxiliary devices		0.146							
Building projects		0.048							
Total capital investment, M€		0.417							
Specific capital cost, €/TPA		571							
Total Installed cost (TIC), M€ - Major equipment cost - Other direct, installation and indirect		1.283		0.767		1.383		0.592	C 1 2010
Auxiliary equipment (20% of TIC) ME	4.6 TPD	0.257		0 153		0.277		0.118	Capra et al. 2019
Total canital investment ME		1 54		0.155		1.66		0.110	
Specific capital cost, €/TPA		957		571		1031		441	
Specific capital cost, €/TPA	6.5-10				822				Gong et al. 2012
Specific capital cost, €/TPA	8.5 TPD				1000				Hönig et al. 2019
<ul> <li>Cold box, screw expanders, compressors</li> <li>Installation and indirect costs</li> <li>Contingencies and fees</li> <li>Total capital investment, M€</li> <li>Specific capital cost. €/TPA</li> </ul>	10 TPD		0.465 0.277 0.134 <b>0.875</b> <b>350</b>						Pasini et al. 2019
Total capital investment, M€ Specific capital cost, €/TPA	10 TPD		3.986 1203						Palizdar et al. 2019
Total capital investment, M€ Specific capital cost, €/TPA	13.2 TPD		6.718 1454						Gustafsson et al. 2020

## Academic literature

	Plant	Reverse B	rayton cycle	Rankine cycle with MR		Linde	Stirling	Liquid	
	capacity /					cycle	refrigeration	Nitrogen	Ref.
	notes	Single $N_2$	Dual $N_2$		Precooled or			$(LN_2)$	
Cost		expander	expander	SMR	dual MR			vaporization	
Direct Cost (DC)									
- Total equipment cost (TEC), M€			2.998	2.406		5.786			
- Buildings (10% of TEC), M€			0.300	0.241		0.579			
- Auxiliary equipment (15% of TEC), M€	15 500		0.450	0.361		0.868			
Indirect Cost (IC)	15 IPD Movable								
- Insurance, freight and tax (1.5% of DC)	liquefaction		0.056	0.045		0.108			Lee et al. 2020
- Engineering cost (3.5% of DC)	plant		0.131	0.105		0.253			
- Overhead cost (15% of DC)			0.562	0.451		1.085			
Contingency (10% of DC+IC)			0.450	0.361		0.868			
Total capital investment, M€			4.947	3.970		9.547			
Specific capital cost, €/TPA			951	763		1836			

# **Technical reports**

		Reverse Br	ayton cycle	Rankine cy	cle with MR	Linde	Stirling	Liquid	
		0' 1 N			Precooled	cycle	refrigeration	Nitrogen (LN <sub>2</sub> )	
Cost	Plant capacity	Single N <sub>2</sub> expander	Dual $N_2$ expander	SMR	or dual MR			vaporization	Ref.
Investment estimate MF		enpuircei	enpuncer			2.8			
- Delivery containerized	4.8 TPD					2.0			
- No civil works included	Cryobox								
Specific capital cost, €/TPA						1687			Himmelstrup
Investment estimate, M€						4.5-5.0			2019
- Delivery containerized	15 TPD								
- No civil works included	Cryobox								
Specific capital cost, €/TPA						867-964			
Investment cost, M€	1 5 TPD						0.580		
- Excluding LNG/LBG storage and	2xStirLNG4								Dioguardi
handling	liquefier								2013
Specific capital cost, €/TPA							1105		
Investment cost, M€									
- Gas conditioning									
- Stirling Liquefaction units StirLNG-4									
- Water chiller							1.950		Stirling
- Piping, fittings, wiring, insulation,	5 TPD						1.850		Cryogenics
cabling									2020d
- Electrical systems									
- Necessary software & accessories									
- LBG storage tank not included									
Specific capital cost, €/TPA							1072		

For comparison, Songhurst [2018] analyzed the published data of 25 large- and mid-scale LNG-projects constructed during 2014–2018. In his report, specific capital costs for mid-scale liquefaction plants – with production capacities in the 0.5 to 2 MTPA (million tons per annum) range – varied from 513  $\notin$ /TPA to 1115  $\notin$ /TPA. The SMR process cost was at the lower end of the price range, while the pre-cooled MR represented the upper end of the price range. The specific costs of N<sub>2</sub> expansion based technologies ranged from 700 to 825  $\notin$ /TPA.

## 4.2 Operating expenses

The operating expenses of a liquefaction plant consist of feed gas cost, electricity cost, labor cost, maintenance cost, water cycle cost, and refrigerant cost. The costs of cooling water and refrigerant represent only a small part of the total operating cost and are ignored in this study. The exception is the  $LN_2$  vaporization cycle, where the cost of liquid nitrogen is significant and therefore added to the operating costs. The cost of feed gas is also not considered in this study. Thus, the operating expenses in this study include two parts: energy costs, and operating and maintenance (O&M) costs.

## 4.2.1 Energy costs

Liquefaction processes are energy-intensive processes due to their cryogenic conditions. Indeed, energy costs are typically the highest operating cost. A summary of the specific energy consumption ( $kWh/kg_{LBM}$ ) associated with various liquefaction technologies is presented in Table 4 and Figure 19. The energy consumption of liquefaction is closely related to the cooling curves of the processes (Zhang et al. 2020).

Table 4.	Specific power	consumption	associated	with	various	liquefaction	technologies.	Liquefaction
capacity	<15 TPD.							

	Technical reports		Academic literature	
Process	Specific power consumption (kWh/kg <sub>LBM)</sub>	Ref.	Specific power consumption (kWh/kg <sub>LBM)</sub>	Ref.
Single N <sub>2</sub> expander	0.78-0.93	[1]	0.75-1.20	[8],[9]
Dual N <sub>2</sub> expander	0.90	[2]	0.52-1.08	[9],[10],[11],[12]
SMR			0.57-0.85	[8],[13],[14]
Precooled MR	>0.70	[3]	0.32-0.82	[13],[15],[16]
Linde cycle	0.66-0.90	[4],[5]	0.88-1.16	[8],[10]
Stirling refrigeration	0.43-1.45	[6]		
LN <sub>2</sub> vaporization	0.007	[7]	0.003	[8]

Data from: [1] Air Liquide 2020, [2] Rouaud 2017, [3] Wärtsilä 2016, [4] Ecospray 2019, [5] Himmelstrup 2019, [6] Stirling Cryogenics 2020a, [7] SIAD 2018b, [8] Capra et al. 2019, [9] Nguyen et al. 2018, [10] Lee et al. 2020, [11] Palizdar et al. 2019, [12] Pasini et al. 2019, [13] Khan et al. 2013, [14] Morosanu et al. 2018, [15] Gong et al. 2012, [16] Hönig et al. 2019



**Figure 19.** Specific power consumption associated with various liquefaction technologies. Liquefaction capacity <15 TPD. Mean values with blue squares, high and low estimates with line segments.



Figure 20 presents the specific energy consumption per kWh<sub>LBM</sub> produced.

**Figure 20.** Specific power consumption in kWh/kWh<sub>LBM</sub>. Liquefaction capacity <15 TPD. Mean values with green diamonds, high and low estimates with line segments.

Based on the above, it can be concluded that even identical processes with approximately the same capacity can have large variations in specific energy consumption. According to, e.g. Zhang et al. [2020], the explanation is the different process parameters, including:

- Inlet gas temperature and pressure
- Feed gas composition
- Liquefaction rate
- Compressor and expander efficiency
- LBM storage pressure
- Ambient temperature

## 4.2.2 Operation and maintenance cost

O&M costs composes of operating work, maintenance work, and maintenance material costs. O&M costs in economic studies of liquefaction technologies [Gustafsson et al. 2020; Lee et al. 2020, Nagy et al. 2017; Pasini et al. 2019; Rehman et al. 2020] range from 2 % to 4 % of investment costs. In this study, O&M costs were assumed to be 2.5 % of the total investment.

## 5 Economic analyses of different liquefaction technologies for 5 TPD plant

The optimal process design is a compromise between investment cost and operating cost. In this section, the economic performances of different liquefaction cycles are investigated based on the life cycle cost (LCC) model. It must be noticed that only liquefaction costs are included in the analysis; even though anaerobic digestion and gas cleaning and upgrading account for a significant fraction of the overall costs of the LBM production chain, they are not analyzed in this work.

The case study is based on existing biogas production capacity in Ostrobothnia. A capacity of 5 TPD represents half of the total available capacity.

#### 5.1 Methodology

LCC is a useful tool for analyzing trade-offs between the investment cost and future operating cost to get minimum total costs during the project's lifetime. In this work, LCC is defined as the sum of the total capital investment (CAPEX) and the present value of future energy and O&M costs (Eq. 1). As a matter of simplification, any incremental investment or decomposition costs at the end of the project over the project's life are not considered in this study.

$$LCC = CAPEX + \sum_{t=1}^{n} \frac{c_{el} + 0\&M}{(1+r)^{t}}$$
 (Eq. 1)

In the LCC calculations, the net present values of future costs were considered with a discount rate (r) of 5 % [SIGMA 2016]. This is expected to cover the financing costs and the general price level increase. The service life (t) of the plant was set at 20 years. CAPEX was calculated by multiplying the average specific capital costs for each process (see Chapter 4.1) by the plant capacity (TPA). The future operating expenses include energy costs (Cel) and operating and maintenance costs (O&M). The energy costs for each process were calculated by multiplying the average specific energy consumption (see Chapter 4.2.1) by the plant capacity and the electricity price. Energy prices were assumed to increase 2 % per year, starting from 0.0862 €/kWh based on Eurostat statistics on electricity prices for non-household customers in Finland in first half of 2020 [Eurostat 2020]. For all processes, O&M costs were assumed 2.5 % of the CAPEX. For the LN<sub>2</sub> vaporization process, also the cost of liquid nitrogen was included in the operating expenses. The bulk cost of liquid nitrogen was assumed to be 100 €/ton, based on discussions with Finnish LN<sub>2</sub> suppliers. The price estimates obtained from domestic liquid nitrogen suppliers ranged between 80  $\in$  and 120  $\in$ , including transportation. An annual price increase of 2 % was also applied for LN<sub>2</sub>. Table 5 summarizes the parameters and assumptions used in the calculations.

 Table 5. Assumptions for LCC analyses.

Parameter	Value
Plant operation time	8280 h/year
Liquefaction capacity of the plant	5 TPD
Plant lifetime	20 years
Discount rate	5 %
Operation and maintenance cost	2.5 % of CAPEX
Price of electricity (first half of 2020)	0.0862 €/kWh
Cost of liquid nitrogen	100 €/ton

To further compare the different processes, the levelized costs of liquefaction (LCOL) were defined for each process in  $\epsilon$ /MWh<sub>LBM</sub>. The levelized costs of liquefaction also represents the average revenue per unit of LBM produced that would be required to cover the initial investment and the operating expenses over the expected lifetime of the project. In Eq. 2, L<sub>cap</sub> stands for annual liquefaction capacity in MWh<sub>LBM</sub>. The lower heating value of LBM is 50 GJ/t (13.9 MWh/t).

Levelized cost of liquefaction = 
$$\frac{CAPEX + \sum_{t=1}^{n} \frac{C_{el} + 0\&M}{(1+r)^{t}}}{\sum_{t=1}^{n} \frac{L_{cap}}{(1+r)^{t}}}$$
(Eq. 2)

### 5.2 Results

The total costs over the entire 20 years life cycle of the liquefaction plant and the cost breakdown for the different liquefaction technologies are shown in Figure 21. The initial capital costs for 5 TPD plant vary between 0.76 M $\in$  and 2.70 M $\in$ , representing 11–50 % of the total life-cycle cost. The processes arranged by the initial capital cost, from lowest to highest, are LN<sub>2</sub> vaporization, SMR, single N<sub>2</sub> expander, dual (or pre-cooled) MR, dual N<sub>2</sub> expander, Stirling refrigeration, and Linde.

Electricity cost accounts for the majority of total operating expenses, except for  $LN_2$  vaporization, for which the price of liquid nitrogen accounts for the vast majority of operating costs. The net present value of total operating cost ranges from 1.7 to 6.0 M $\in$  over the plant's 20-year service life.

The processes arranged by the total life-cycle cost, from lowest to highest, are SMR, dual (or pre-cooled) MR, single  $N_2$  expander, dual  $N_2$  expander, Stirling refrigeration, Linde, and  $LN_2$  vaporization. From an economic perspective, the Rankine cycle with mixed refrigerant appears the most advantageous option for the nanoscale liquefaction process, followed by nitrogen expander processes based on the reversed-Brayton cycle. The higher total life-cycle costs of Linde refrigeration is particularly driven by high investment costs. Liquid nitrogen vaporization is the least capital intensive, but its life-cycle costs depend largely on the price at which liquid nitrogen is available.



Figure 21. Total life-cycle costs (M€), discount rate 5 % and plant lifetime 20 years.

The levelized cost of liquefaction, illustrated in Fig. 22, represents the per-MWh cost of constructing and operating a 5 TPD liquefaction plant over its assumed financial life and activity level. The levelized liquefaction cost equals  $10.4 \notin MWh_{LBM}$  (0.14  $\notin /kg$ ) and 11.9  $\notin /MWh_{LBM}$  (0.17  $\notin /kg$ ) for the most profitable options SMR and pre-cooled MR, respectively. For N<sub>2</sub> expander processes, the levelized liquefaction cost is  $13.1-13.4 \notin /MWh_{LBM}$  (0.18  $\notin /kg$ ) depending on the process configuration. For Stirling refrigeration, the LCOL is 15.60  $\notin /MWh$ , and for Linde system the LCOL exceeds 17  $\notin /MWh$ . In the case of LN<sub>2</sub> vaporization, the LCOC exceeds 24  $\notin /MWh$ .



**Figure 22.** Levelized cost of liquefaction ( $\notin$ /MWh<sub>LBM</sub> and  $\notin$ /kg<sub>LBM</sub>), discount rate 5 % and plant lifetime 20 years.

## 5.3 Sensitivity analyses

To provide a broader view of the results of the economic assessment and to address uncertainties related to investment costs and future expenditure flows, a sensitivity analysis was performed against two types of variables:

- High/low investment cost
- Discount rate

### 5.3.1 High/low investment cost scenarios

The available capital cost data for <15 TPD liquefaction plants were limited to data from eight academic studies and three technical reports. Most studies considered only the liquefaction system, excluding, e.g., pre-treatment, storage system, civil works, and the owner's costs. Also, differences in the availability of existing infrastructure, safety standards, and labor costs for installation may vary. Therefore, a sensitivity analysis considering low and high CAPEX was performed. In the analysis, -20 % - +50 % variability around the initial estimate was applied. The sensitivity of levelized liquefaction costs to the plant capital cost is shown in Fig. 23.

Even with an increase of the CAPEX of mixed refrigerant processes by 50 %, these options remain less expensive in terms of  $\epsilon$ /MWh<sub>LBM</sub> than the most capital-intensive alternative Linde with 20 % CAPEX reduction, and the highly OPEX intensive LN<sub>2</sub> vaporization. In high CAPEX scenario, the LCOL for N<sub>2</sub> expander based applications ranges from 15.5–16.5  $\epsilon$ /MWh<sub>LBM</sub>. For Stirling the LCOL in high CAPEX scenario is 19  $\epsilon$ /MWh, and for Linde the LCOL exceed 20  $\epsilon$ /MWh<sub>LBM</sub> in high CAPEX scenario.



**Figure 23.** Sensitivity of biomethane liquefaction cost to CAPEX; -20 % - +50 % variability around the initial estimate.

### 5.3.2 The effect of discount rate

In the base case, a discount rate of 5 % was applied, representing the minimum return that an investor expects to achieve to cover the cost of finances and the general price level increase. In the sensitivity analysis, a risk premium of 5 % and 10 % was added, leading to the required return (=discount rate) of 10 % and 15 %. For CAPEX, the initial estimate was applied. The results of the sensitivity analysis are shown in Figure 24. The revenue per unit of LBM produced that would be required to cover the initial investment, the operating expenses, and the investment risk over the expected lifetime of an MR project is  $11.7-16.4 \text{ €/MWh}_{LBM}$  (0.16–0.23 €/kg<sub>LBM</sub>), depending on technology and investor's risk tolerance. For N<sub>2</sub> expander processes the required revenue per unit of LBM is  $14.6-18.4 \text{ €/MWh}_{LBM}$  (0.20–0.26 €/kg<sub>LBM</sub>), for Linde system 20.0–24.0 €/MWh<sub>LBM</sub> (0.28–0.33 €/kg<sub>LBM</sub>), for Stirling  $17.8-21 \text{ €/MWh}_{LBM}$  (0.25–0.29 €/kg<sub>LBM</sub>) and for LN<sub>2</sub> vaporization 25.2–26.0 €/MWh<sub>LBM</sub> (0.35–0.36 €/kg<sub>LBM</sub>) depending on technology, and investor's risk tolerance.



Figure 24. Sensitivity of biomethane liquefaction cost to the discount rate.

## **6** Summary

All liquefaction techniques have their advantages and disadvantages. The key to the successful optimization of a nanoscale liquefaction project is to find an appropriate balance between technical, operational, and economic characteristics. From a technical and operational perspective, the main findings in this study were:

- The N<sub>2</sub> expansion cycle has the advantages of a simple and understandable process that is easy to use and maintain, as the process requires less monitoring and control points and minimal operator intervention compared to, e.g., MR processes. The working fluid is in gaseous phase, which prevents maldistribution issues in heat exchangers. Moreover, nitrogen is an unreacted refrigerant that provides a high level of safety. Liquid hydrocarbon storage systems are also not required for the refrigerant mixture. The most significant disadvantages of N<sub>2</sub> expander processes are the lower energy efficiency compared to MR processes and relatively high space requirements.
- Mixed refrigerant processes require less power consumption than N<sub>2</sub> expansion, Linde, or Stirling processes. The MR processes' remarkable efficiency is attributed to the possibility of tuning the refrigerant composition so that its evaporation curve matches the cooling curve of the feed gas from ambient to cryogenic temperatures. The compact SMR process with phase changes reduces the piping size compared to the N<sub>2</sub> loop. Even higher system efficiencies can be achieved by adopting a pre-cooling cycle. The disadvantages of such setups are the higher complexity of the process and the high equipment count and capital cost compared to simple configurations. Robust and reliable, mature technology.
- The advantages of the Linde cycle includes simple set-up, no need for cooling media (N<sub>2</sub> or MR), easy maintenance, fast start-up, and boil-off free operation. Furthermore, besides producing LBM, the system can provide compressed gas as needed. In this way, both fuels are simultaneously available. The disadvantage of J-T systems is the high compression energy requirement and corresponding low efficiencies. The efficiency can be improved by implementing a pre-cooling cycle in the system.
- Stirling refrigeration is a robust, stand-alone system, easy to operate, and requires a low amount of operator involvement. The major drawback is that only small capacity units are available. Achieving larger sizes requires the use of multiple units in parallel, preventing economies of scale. On the other hand, the modular design enables a high partial-load capability. So far, there are a limited number of Stirling refrigeration applications in the field of methane liquefaction.
- LN<sub>2</sub> vaporization is functionally the most straightforward and least capital-intensive option, as the liquefaction process uses a considerably small amount of equipment and does not require high-cost turbomachinery. It also offers a high level of safety due to the use of inert gas as a refrigerant. The consumption of liquid nitrogen equals 2.5 kg LN<sub>2</sub> per kg

biomethane, so its life-cycle cost depends largely on the price at which liquid nitrogen is available. Best suited for plants with very low capacities and availability of liquid nitrogen.

The goal of financial optimization is to minimize total cost. For example, minimizing energy consumption by using more or larger heat exchangers or additional cooling cycles increases the complexity of the process, leading to higher capital cost, and thus, does not necessarily result in the minimum total life-cycle cost. Correspondingly, minimizing investment costs does not necessarily lead to the lowest life cycle costs, as was the case for  $LN_2$  vaporization. This study used an LCC method as a tool for analyzing trade-offs between the investment cost and future operating costs to get minimum total costs during the project's lifetime. A brief comparative summary of the costs of the different liquefaction technologies is presented in Table 6. Operating costs and life-cycle costs are presented relative to the most economically advantageous option, SMR.

	Single	Dual	SMR	Dual or	Linde	Stirling	LN <sub>2</sub>
	$N_2$	$N_2$		precooled	cycle	refrig.	vapor.
	expander	expander		MR			
Capital costs	Low to medium	Medium	Low	Medium	High	Medium	Low
Operating costs (relative to SMR)	1.35	1.2	1	1	1.4	1.4	3.5
Total LCC (relative to SMR)	1.25	1.3	1	1.15	1.65	1.5	2.4

 Table 6. Comparative summary of the costs of liquefaction technologies for 5 TPD plant.

Weighing up the technical, operational, and economic characteristics, at least the following objectives shall be kept in mind:

- Process simplicity
- Easy operation and maintenance
- Safety
- Low costs
- High reliability
- Scalability / modularity

The purpose of this survey was to provide an overview of the technical and economic performance of LNG processes suitable for small/nano-scale applications. In the actual procurement phase, the process parameters need to be defined in more detail for system optimization, in which case, for example, the energy consumption data may change. The conclusive cost analysis shall be made on the basis of binding offers.

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