

Production of liquid biogas, LBG, with cryogenic and conventional upgrading technology

- Description of systems and evaluations of energy balances

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Abstract

The aim of this master thesis is to evaluate energy balances for the production of liquid biogas, LBG. LBG can be produced in two main ways and these are by means; cryogenic upgrading technology and conventional technologies connected with a small-scale liquefaction plant. A third alternative is to inject biogas into the gas grid and then liquefy a part-flow at a pressure letdown station.

The cryogenic upgrading technology is based on differences in condensation temperature for different compounds, and by chilling biogas, impurities and CO₂ can be separated from CH₄. When using this technology also liquid CO₂, LCO₂, comes as a by-product, which could be used in external applications.

With one of the two main ways it takes between 0.8-1.8 kWh/Nm³ clean biogas to produce LBG. If the energy is expressed in primary energy, this energy consumption corresponds to 12-23 % of the energy content in the product. The net energy consumption is affected by the disposal of waste heat and use of LCO₂ in external processes while CH₄ losses have a small influence.

The production of LBG is more energy intensive than the production of compressed biogas but in some situations the product is more valuable since the biogas becomes available for more customers.

Keywords

Cryogenic technology, liquid biogas, LBG, energy balance, small-scale liquefaction, LCO₂, primary energy

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Executive summary

Biogas is produced when organic material is decomposed under anaerobic conditions, for example in a landfill or under controlled conditions in a digester chamber. The product mainly consists of carbon dioxide (CO₂) and the energy carrying compound methane (CH₄), usually with a higher part of the latter. The interest in using biogas as a vehicle fuel in Sweden is growing, but to be able to do so, the biogas needs to be purified and upgraded, which means that impurities and CO₂ are removed. There are a number of technologies available for this purpose and the most commonly used upgrading technologies in Sweden are water scrubbing and PSA. Upgraded biogas has a CH₄ content of approximately 97 % and before fuelling it is compressed to 200 bar.

One alternative to the conventional technologies is to upgrade biogas with *cryogenic technology*, which means that the gas is chilled and the differences in condensation temperature for different compounds are used to separate impurities and CO₂ from CH₄. CO₂ condense at -78.5 °C at atmospheric pressure. The technology can be used to upgrade raw biogas by chilling it to the condensation temperature for CO₂ or it can be further chilled to -161 °C (condensation temperature for CH₄ at atmospheric pressure) to produce liquid biogas, LBG. It is more energy intensive to chill the gas to -161 °C but in some situations it results in a more valuable product since LBG is more than 600 times space efficient compared to biogas in its gas phase at atmospheric pressure (around 3 times more space efficient compared to compressed biogas, 200 bar). This makes the biogas available for more customers since the produced LBG can be transported on road in vacuum insulated semi-trailers to remote fuel stations. On a multi-purpose fuel station it is then stored as LBG and fuelled as either LBG or CBG (compressed biogas, 200 bar).

LBG can also be produced using one of the conventional upgrading technologies connected with a small-scale liquefaction plant. This small-scale liquefaction plants are either a closed nitrogen cycle or a closed mixed refrigerant cycle. The first has a low efficiency but it is not as complex as the latter since it only use one refrigerant (nitrogen).

A third alternative is to inject the upgraded biogas into the gas grid and then liquefy it at a pressure letdown station. Here the pressure drop is used to produce electricity in a turbo expander and the electricity is then used to drive a compressor. A part flow of the gas stream can be liquefied without any addition of energy. This is an energy efficient way to produce LBG, but as much as 7-9 % of propane must be added to the biogas to meet the heating value of natural gas.

When using cryogenic upgrading technology or conventional upgrading technology connected with a small-scale liquefaction plant it takes approximately 0.8-1.8 kWh electricity (and heat)/Nm³ clean biogas to produce LBG. The most energy efficient ways are to use water scrubber, PSA or Coaab (including heat recovery), connected with a mixed refrigerant cycle or to use Scandinavian GtS's process (cryogenic technology).

By using the *primary energy* concept, where original data are converted to primary energy (the energy needed to produce one unit of useful energy, like electricity or heat), different technical solutions can be compared and evaluated in energy balances. The inserted *primary energy* to produce LBG is between 12-23 % of the energy content in the product.

An important aspect when evaluating biogas-upgrading technologies is CH₄ losses and these varies between *none* and 2 %. Because of the low numbers the CH₄ loss has a small impact on the net energy consumption. However, it results in a loss of income and also in an emission of a greenhouse gas, more than 20 times efficient than CO₂. One parameter that does affect the net energy consumption is the disposal of waste heat in external processes. This opportunity is however very site specific since a user of low value heat is not always available.

When using cryogenic upgrading technology clean, liquid CO₂, LCO₂, comes as a by-product. This LCO₂ could be used in external processes replacing fossil energy and bring in extra income to the biogas production plant. Two possible applications are cryogenic temperature control and fertilizing of greenhouses. The LCO₂ probably has to be sold directly to the user, and the possibility to get an income from this product is also very site specific. An interesting alternative could be to place a greenhouse close to a biogas production and upgrading plant. In this way the greenhouse could get an organic fertilizer from the digester chamber and heat and CO₂ from the upgrading process.

Sammanfattning

Biogas bildas när organiskt material bryts ner i syrefri miljö, till exempel i deponier och under kontrollerade former i en rötchammare. Biogas består i huvudsak av koldioxid, CO_2 , och energibäraren metan, CH_4 , vanligtvis med en större andel av den sistnämnda. I Sverige ökar intresset för att använda biogas som fordonbränsle. För att kunna göra det måste dock biogasen renas och uppgraderas vilket innebär att föroreningar och CO_2 separeras från CH_4 . Det finns ett antal tillgängliga tekniker för detta ändamål och i Sverige är de vanligaste teknikerna vattenskrubber och PSA. Den uppgraderade biogasen har en CH_4 koncentration på omkring 97 % och innan den tankas komprimeras gasen till 200 bar.

Ett alternativ till kommersiell teknik är att uppgradera biogasen med *kryoteknik*, vilket innebär att gasen kyls och olika kondenseringstemperaturer för olika ämnen gör att föroreningar och CO_2 kan separeras från CH_4 . CO_2 kondenserar vid -78.5°C vid atmosfärstryck och teknologin kan användas för att producera uppgraderad biogas, genom att kyla gasen till denna temperatur. Ett annat alternativ är att kyla gasen ytterligare, till -161°C (kondenseringstemperaturen för CH_4) för att producera flytande biogas, LBG. Det går åt mer energi för att kyla till -161°C men i vissa sammanhang resulterar det i en produkt som är mer värd, eftersom LBG tar mer än 600 gånger mindre plats än biogas i gasform vid atmosfärstryck (omkring 3 gånger mindre plats i jämförelse med komprimerad biogas, 200 bar). Detta gör att biogasen blir tillgänglig för fler kunder, eftersom biogasen kan transporteras med lastbil i vakuumisolerade trailers till avlägsna tankstationer. På en multifunktionstankstation förvaras sedan gasen som LBG medan den tankas som LBG eller CBG (komprimerad biogas, 200 bar).

LBG kan också produceras genom att använda en konventionell uppgraderingsteknik tillsammans med småskalig förvätskningsteknik. Förvätskningstekniken är antingen en sluten kvävecykel eller en sluten cykel med en blandning av kylmedium (*mixed refrigerant*). Den första cykeln har en låg effektivitet men den är inte lika komplex som den sistnämnda eftersom endast ett kylmedium används (kväve).

Ett tredje alternativ är att injicera biogasen på naturgasnätet för att sedan förvätska ett delflöde i en MR-station. Här används tryckfallet vid MR-stationen för att i en turboexpander producera elektricitet, vilken sedan driver en kompressor. Med denna teknik kan ett delflöde förvätskas utan tillsats av extra energi. Detta är ett energieffektivt sätt att producera LBG men för att kunna injicera biogasen i naturgasnätet måste dock 7-9 % gasol tillsättas.

När LBG produceras med kryogen uppgraderingsteknik eller med kommersiell uppgraderingsteknik tillsammans med ett förvätskningssteg går det åt mellan 0.8-1.8 kWh elektricitet (och värme)/ Nm^3 ren biogas. De mest energieffektiva sätten är att använda en vattenskrubber, PSA eller Coaab (inklusive värmeåtervinning) tillsammans med en *mixed refrigerant* process eller att använda Scandinavian GtSs process (kryogen uppgraderingsteknik).

Genom att använda *primärenergibegreppet* kan olika tekniska lösningar jämföras och utvärderas i energibalanser. Primärenergibegreppet innebär att originaldata omvandlas till *primärenergi*, vilket är den energi det går åt för att producera en enhet nyttig energi som till exempel elektricitet eller värme. Den insatta primärenergien för att producera LBG är mellan 12-23 % av energiinnehållet i produkten.

En viktig aspekt vid utvärdering av biogasuppgradering är CH_4 -förluster. Dessa varierar mellan *inga* och 2 %. På grund av de låga siffrorna har CH_4 -förluster liten inverkan på nettoenergiförbrukningen. Dock resulterar det i en inkomstförlust och också ett utsläpp av en växthusgas som är mer än 20 gånger så effektiv som CO_2 . En parameter som har stor påverkan på denna förbrukning är avsättningen för återvunnen värme i externa processer. Möjligheten till avsättning är dock väldigt platspecifik eftersom en användare av lågvärdig värme inte alltid är tillgänglig.

När kryogen uppgraderingsteknik används fås ren, flytande CO_2 , LCO_2 , som en biprodukt. Denna LCO_2 kan användas i externa processer och på så sätt ersätta fossil energi, men även inbringa en extra inkomst till biogasuppgraderingsanläggningen. Två möjliga användningsområden är kyltransporter och som gödningsmedel i växthus. Den producerade LCO_2 :n måste förmodligen säljas direkt till användaren och även denna avsättning är väldigt platspecifik. Ett intressant alternativ skulle kunna vara att placera ett växthus i anslutning till en biogasproduktions- och uppgraderingsanläggning. På detta sätt skulle växthuset kunna få en ekologisk gödsel från rötningsprocessen och både värme och CO_2 från uppgraderingsanläggningen.

Nomenclature and explanations

Bellow follows a description of used abbreviations and expressions. Any parenthesis behind the explanation defines the assumptions used in the report.

CBG	<u>C</u> ompressed <u>B</u> iogas (200 bar and 97-99 % CH ₄)
CH ₄	Methane
Clean biogas	Purified and upgraded biogas. In this report clean biogas is defined with a CH ₄ content of 100 %. However, the actual CH ₄ concentration in a product varies between 97-100 %, depending on technology, but to enable comparison all numbers on energy consumption are expressed per 100 % CH ₄ .
CNG	<u>C</u> ompressed <u>N</u> atural <u>G</u> as
CO ₂	Carbon dioxide
Conventional upgrading technology	Biogas purification and upgrading technologies that are commercially available and/or commonly used in Sweden.
Cryogenics	The study of very low temperatures (< -150°C)
d	Relative density; the gas density relative the air density
Dry ice	solid CO ₂
Dual fuel	A technology that is used to substitute diesel for biogas in a diesel engine, without losing any efficiency. Diesel is only used for the ignition and as much as 90 % can be replaced.
Flashing	Reduction in pressure, which makes dissolved components in the liquid to leave.
H	Heating value
H ₂ S	Hydrogen sulphide
Heating Value	A measure of the energy content in the gas. There are both an upper and a lower heating value. The upper is the energy released during combustion and the lower is the energy released during combustion, minus the energy that is released when water vapour in the exhaust gases is condensed. In Sweden the lower heating value is used to specify the energy content in fuels.
LBG	<u>L</u> iquid <u>B</u> iogas (100 % CH ₄) Other existing expressions are LMG (<u>L</u> iquid <u>M</u> ethane <u>G</u> as) and LBM (<u>L</u> iquid <u>B</u> iomethane)

LCO ₂	<u>L</u> iquid <u>C</u> O ₂
LFG	<u>L</u> andfill <u>G</u> as
LNG	<u>L</u> iquid <u>N</u> atural <u>G</u> as
MRC	<u>M</u> ixed <u>R</u> efrigerant <u>C</u> ycle
N ₂	Nitrogen
Nm ³	Normal cubic meter, gas volume at 1.013 bar and 0°C
Primary energy	The energy needed to produce one unit of useful energy, like electricity or heat. The primary energy factors for a Nordic electricity mix and heat produced from the burning of forest fuel in a boiler are, in this report, defined as 1.6 and 1.16 respectively.
PSA	<u>P</u> ressure <u>S</u> wing <u>A</u> dsorption
Raw biogas	Untreated biogas with a CH ₄ content between 35-70 %, depending on source (either landfill gas (35-65 % CH ₄) or digester gas (60-70 % CH ₄)).
Siloxanes	Organic silica compounds that occasionally occurs in landfill gas and digester gas from sewage sludge.
Wobbe Index, W	$W = H/\sqrt{d}$ A measure used to determine the interchangeability for different gases.

Energy content in different fuels (SGC, 2007):

Fuel:	Energy content (kWh):
1 Nm ³ biogas (97 % CH ₄)	9.67
1 Nm ³ LBG (100 % CH ₄)	9.97
1 Nm ³ natural gas*	11.0
1 litre petrol	9.06
1 litre diesel	9.8
1 litre E85	6.6

1 Nm³ biogas is equivalent to ca. 1.1 litres of petrol

* Average value of Danish natural gas 2005

Company presentation

Below follows a short description of the companies mentioned in this report.

Acrion Technologies/ Terracastus Technologies	Acrion is a small American company that develops technologies for the separation and purification of CO ₂ rich gases. Terracastus Technologies holds the license to produce LBG from Acrion's <i>CO₂ Wash</i> [®] process. www.acrion.com , www.terracastus.com
AGA	A Swedish supplier of industrial gases, including biogas and CO ₂ . AGA is a member of The Linde Group. www.aga.se
Air Liquide Advanced Technologies	An expert in industrial gases and specialized areas, like biogas. A supplier of a process for the production of LBG using membranes for the separation of CO ₂ . A member of the Air Liquide Group. www.dta.airliquide.com/en/welcome.html
Air Liquide (Sweden)	A Swedish expert in the industrial gas field and a supplier of CO ₂ . A member of the Air Liquide Group. www.airliquide.se/
BOC Gases	A supplier of industrial gases and gas equipment. A member of The Linde Group. www.boc-gases.com/
Carbotech	A German company that develops plants for gas purification and generation. A supplier of PSA technology. www.carbotech.info
Cryo AB	A manufacturer of cryogenic equipment for the storage, transportation and handling of liquefied gases. A member of The Linde Group. www.cryo.se
Cryostar	A company specialized in cryogenic equipment. A supplier of fuel stations using LBG as a feedstock and also a supplier of small-scale liquefaction technology. www.cryostar.com
Flotech	A New Zealand company that, among others, operates in Sweden. A supplier of water scrubber technology. www.flotech.com
Gasrec	A British producer of liquid methane fuel. They use gas generated by the decomposition of biomass. www.gasrec.co.uk

GTI/Gas Technology Institute	An American research and development organization in the energy field. A developer of small-scale liquefaction technology. www.gastechnology.org
Hamworthy	A company working in the oil and gas market. A supplier of a small-scale liquefaction technology. www.hamworthy.com
Hardstaff Group	A U.K. company working in the road transport industry and a developer of natural gas vehicle technology with their own patented dual fuel technology www.hardstaffgroup.co.uk
H-O Nilsson AB/ Ingersoll Rand Svenska AB	A supplier of refrigerating plants, including CO ₂ cooling systems. www.honilsson.se
INL/ Idaho National Laboratory	An American national laboratory that supports the Department of Energy. A developer of liquefaction technology used at pressure letdown stations. www.inl.gov
Malmberg Water	A Swedish company working with the fields of; biogas, heat/cold, water treatment, drilling and environmental management. A supplier of water scrubber technology. www.malmberg.se
Nexgen Fueling	A provider of equipment needed for LNG liquefaction, distribution, storage and vehicle fueling. A part of Chart Industries. www.nexgenfueling.com
Prometheus-Energy	An American company that produce, sell and distribute LNG produced from small sources. A developer and supplier of cryogenic upgrading technology. www.prometheus-energy.com
Purac/Läckeby Water Group	Purac is a part of the Läckeby Water Group. Läckeby Water Group is a Swedish company working with water and wastewater treatment and biogas production. www.lackebywater.se
SGtS/Scandinavian GtS	A developer of biogas projects and a supplier of cryogenic upgrading technology. www.scandinaviangts.com
SITA UK	A British recycling and waste management company. www.sita.co.uk

The Linde Group	A gases and engineering company working in the biogas field, among others. www.linde.com
Vanzetti Engineering	A manufacture of cryogenic equipment and a supplier of fuel stations using LBG as a feedstock. www.vanzettiengineering.com
Waste Management Inc.	An American Waste Management company. www.wm.com

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1 Introduction

Today, there is a big interest in using renewable vehicle fuels and one option is to use biogas. Biogas is produced when organic material is decomposed under anaerobic conditions and in this report biogas is referred to as landfill gas and digester gas. The main constituents are methane, CH_4 , and carbon dioxide, CO_2 , usually with a higher part of CH_4 (around 60-70 %).

To be able to use the raw biogas as a vehicle fuel it must be purified and upgraded, which means that impurities and CO_2 respectively, are removed. There are a number of available upgrading technologies and the two most commonly used in Sweden are water scrubbing and PSA. The produced biogas can be injected into the gas grid or compressed to around 200 bar for use as vehicle fuel. However, in Sweden the gas grid is limited to the southwest coast and the biogas production plants are often situated far away from the end users. Therefore, compressed biogas (CBG) is distributed on mobile CBG storages, which is a very inefficient way to deliver gas since a huge share of steel is transported in comparison to the amount of gas.

One solution to this problem is to use cryogenic technology, meaning that low-temperature processes ($<-150\text{ }^\circ\text{C}$) and equipment are used, to produce liquid biogas, LBG. LBG is more than 600 times space efficient compared to biogas at atmospheric pressure (around 3 times more space efficient compared to CBG).

There are two main ways to produce LBG and these are cryogenic upgrading technology and conventional upgrading technology connected with a small-scale liquefaction plant. In the cryogenic upgrading technology differences in condensation temperatures are used to separate impurities and CO_2 from CH_4 . Using this technology also results in a clean liquid CO_2 product, LCO_2 , that could be used in external applications.

In the following text the different technical solutions for the production of LBG will be presented and analysed. Focus lies upon energy consumption but other important factors are environmental aspects and technical solutions. The analysis is based on energy balances and the primary energy concept is used to enable comparisons between different energy carriers. Included in the energy balances are the recovery of waste heat, CH_4 losses and the use of LCO_2 in external processes.

1.1 Aim

The aim of this master thesis is to evaluate the energy balances for cryogenic and commercial upgrading technologies for the production of LBG. The evaluations are made in a system perspective and included in the system are purification, upgrading and liquefaction of biogas, distribution and fuelling.

1.2 Method

Cryogenic technology and the production of LBG is not yet an everyday occurrence but the technology is on the way to commercialisation. Since there is little experience of the technology there is not much written about it. There are however three reports from the Swedish Gas Centre (SGC) that deal with cryogenic technology and LNG as an option for Sweden. These reports have been a starting point in the work with this thesis, but all three have a more economical approach.

The early work was focused on finding relevant suppliers of cryogenic technology, working in the biogas field. In this search the SGC reports were of great use. The work was then transferred to data collection from the suppliers found in the first step. The data presented in this report thus mainly originates from these suppliers.

The data concerning the biogas introduction and more conventional technologies comes from different reports from acknowledged Swedish and International organisations, institutes and companies.

To facilitate the reading of the report abbreviations for commonly used expression are used throughout the text. The explanations of these abbreviations can be found after the summary.

1.3 Delimitations

The analyses are made in a Swedish perspective. In Sweden the interest in using biogas as a renewable vehicle fuel is growing and because of the limited gas network the production of LBG could be a good alternative to make biogas available for more end users. The results from analyses would differ if it were based on another country, with for example a well-extended gas grid or were biogas mainly is used for electricity generation.

The costs for different technologies are not included in the report. This was meant to be included from the beginning but since only a few suppliers were willing to hand over this kind of information, it was excluded.

The environmental aspects of using biogas as vehicle fuel, compared to fossil fuel, is not included in the report.

1.4 Method criticism

The data presented in this report is mainly based on suppliers, which sometimes makes it less reliable. No study visits have been possible since the identified commercial and pilot LBG production plants are situated outside Sweden. Study visits probably would have raised the reliability in supplier's data and the understanding in the technological issues.

1.5 Content

The report begins with an introduction to biogas (chapter 2) followed by a description of the upgrading technologies in use today (chapter 3). After these two background chapters the report will focus on liquid biogas, LBG, in three chapters regarding production, distribution and fuelling (chapter 4, 5, and 6). Then follows a short chapter where commercial LBG production plants are presented (chapter 7) followed by a chapter that treats the use of liquid CO₂, LCO₂ (chapter 8). This leads up to chapter 9; Analysis and Energy Balances. In this chapter the different technical solutions are analysed and energy balances are evaluated. The report is then completed with discussions and conclusions in chapter 10.

Three appendices are attached to the report and these concerns the cryogenic upgrading technologies (appendix 1), a technology using membranes for the production of LBG (appendix 2) and original data, data conversions and calculations (appendix 3).

2 Introduction to biogas

Biogas is the name of the gas produced when organic material is decomposed under anaerobic conditions. These processes take place naturally when the amount of organic material is sufficient and where oxygen does not enter, for example in wetlands. (Energimyndigheten, 2008) In this report biogas is referred to as digester gas, gas produced under controlled conditions in digester chambers, and landfill gas, gas spontaneously produced in landfills.

Digestion processes has been used since 1960 but then the main goal was to stabilize and thicken sewage sludge at sewage treatment plants and to treat polluted organic process wastewater. After the energy crisis in the 1970s, the interest in recovering energy from renewable sources grew and the biogas production was expanded to include industrial waste and manure. In the beginning of the 1990s the first co-digestion plants for the joint digestion of different substrates, like organic farm- and household waste, were built. Landfill gas has been collected since the 1980s. At first it was collected because of safety reasons, but today the reduction of greenhouse gas emissions is an important argument. (SBGF et al, 2008)

The main constituents of biogas are methane, CH₄, and carbon dioxide, CO₂, with small amounts of hydrogen sulphide, H₂S and water vapor. The gas can be used directly for production of heat and/or electricity or it can be further processed to natural gas quality for use as vehicle fuel or for injection into the gas grid. (Persson & Wellinger, 2006)

2.1 Biogas process

As mentioned above biogas is produced when microorganisms, in an anaerobic environment, decompose organic material. The energy-carrying compound is CH₄ and a higher CH₄ concentration means higher energy content.

There are two sources of biogas and these are landfills and digester chambers. In landfills, gas is produced spontaneously as long as there is decomposition of organic material. Since the process is not controlled or optimized, it results in a lower CH₄ content, around 50 %, in comparison to digester gas. The landfill gas is collected with permeable tubes by applying a slight under-pressure. Since Jan 2005 it is prohibited to landfill organic material, which will result in a decreasing biogas production. However, the decomposition process in a landfill is slow, so they will probably give gas for another 30 – 50 years. (SBGF et al, 2008)

Digester gas is produced under controlled situations in a digester chamber. Most of the gas is produced in sewage sludge treatment plants, but the production from co-digestion is steadily rising. In co-digestion plant different substrates like manure, slaughterhouse and industrial waste and sorted food waste from food industry, restaurants and households are digested together. (Energimyndigheten, 2008) The digester gas is produced in three main steps, by a number of different microorganisms:

1. Hydrolysis

In the hydrolysis, microorganisms use enzymes to break down complex organic material to more simple compounds like sugars and amino acids.

2. Fermentation

Through fermentation organisms form intermediate products like fatty acids, alcohols and hydrogen.

3. Methane Production

In this step a unique group of microorganisms, called methanogens, produce methane from acetic acid. These organisms have very specific requirements on their environment. They are sensitive to temperature fluctuations and pH, they grow slowly and they die when in contact with oxygen. (SBGF et al, 2008)

The duration in the digester chamber is between 15-30 days, depending on substrate and temperature (Energimyndigheten, 2008). The process is mesophilic (~37 °C) or thermophilic (50-55 °C), which is the temperature at which the CH₄ producing bacteria have growth rate peaks. This means that heat must be added to the process. A stirring device helps to keep a steady temperature, and at the same time it gives better contact between microorganisms and substrate and prevents stratification. (SBGF et al, 2008)

The gas is taken out from the top of the tank and the CH₄ content varies between 60-70 %, depending on substrate. After digestion, there is a rest of organic material, called digestate, which is pumped out to a storage tank. Depending on the substrate origin and pretreatment, this digestate can function as an excellent fertilizer. Further, it could be certified and used as an organic fertilizer. (SBGF et al, 2008) A flow diagram over a digestion process can be seen in Figure 1.

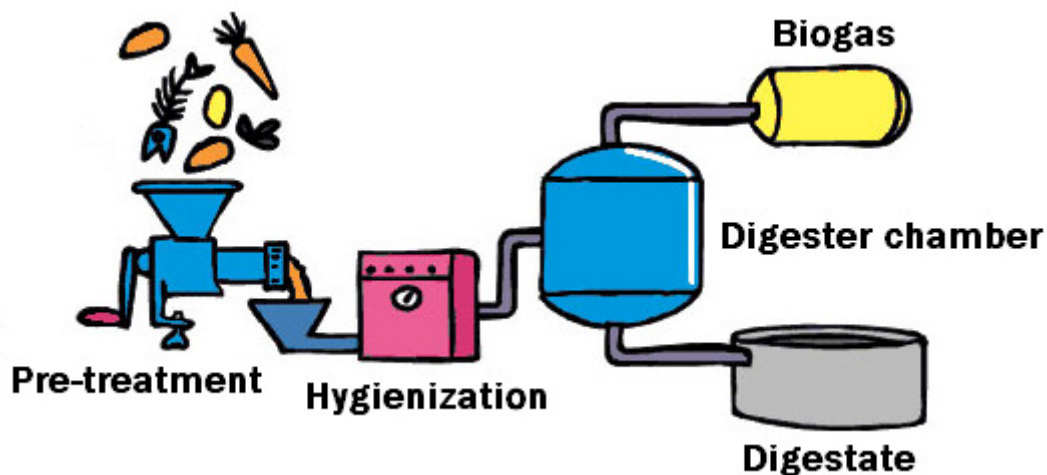


Figure 1 Flow diagram over the digestion process. (SGC, 2008a)

2.1.1 Pre-treatment

If the substrate has an animalistic origin the material must be hygienized to eliminate pathogenic bacteria. The material is heated to 70°C for at least one hour before it is injected into the digester chamber. (SBGF et al, 2008)

Some substrates, like food waste from households and restaurants, needs to be sorted and grinded into a fine homogenous material, called *slurry*, before it is injected into the gas chamber. Metals are removed from the waste with a magnet. (SBGF et al, 2008)

2.2 Composition

Typical properties and compositions of digester and landfill gas can be seen in Table 1 (The gas properties on the first three lines will be discussed in chapter 2.4.1). As mentioned above the main constituents of biogas is CH₄ and CO₂. The gas also contains traces of other substances like hydrogen sulphide, H₂S, ammonia, chlorinated compounds, siloxanes and dust particles and it is usually saturated with water vapour. Landfill gas can also contain traces of halogenated compounds, higher hydrocarbons and aromatic compounds (not presented in Table 1). (Persson & Wellinger, 2006)

Table 1 Typical properties and composition of landfill gas and digester gas. (SGC, 2007)

Character:	Unit:	Landfill gas:	Digester Gas:
Calorific value, lower	kWh/Nm ³	4.4	6.5
Density	kg/Nm ³	1.3	1.2
Wobbe index, upper	MJ/Nm ³	18	27
Methane	vol-%	45	65
Methane, range	vol-%	35-65	60-70
Long-chain hydrocarbons	vol-%	0	0
Hydrogen	vol-%	0-3	0
Carbon dioxide	vol-%	40	35
Nitrogen	vol-%	15	0.2
Nitrogen, range	vol-%	5-40	-
Oxygen	vol-%	1	0
Oxygen, range	vol-%	0-5	-
Hydrogen sulphide	ppm	< 100	< 500
Hydrogen sulphide, range	ppm	0-100	0-4000
Ammonia	ppm	5	100
Total chlorine as Cl ⁻	mg/Nm ³	20-200	0-5
Siloxane*	µg/g DW	12.9	-

* Average for samples on Swedish municipal sludge (Kaj et al., 2005)

The range in which different compounds lies within depends on many factors, like substrate origin and process design. Usually digester gas has a higher CH₄ content than landfill gas, due to the controlled and optimized process. A specific feature with landfill gas is that it usually has a high content of N₂. When the gas is sucked out from the landfill it makes air entering the empty space. The oxygen is consumed in the decomposing processes but N₂ follows with the gas.

Landfill gas also contains higher amounts of chlorinated compounds and siloxanes (organic silica compounds), while digester gas has a higher content of H₂S and ammonia. (SGC, 2007)

2.3 Biogas in Sweden

In 2006 there were 223 biogas production plants in Sweden, 60 of them were landfills and the rest digester chambers. The different types of biogas facilities, together with their average methane content and biogas production, can be seen in Table 2. Most of the digester chambers are sewage sludge treatment plants, which count for 138 plants of 163 totals. Interesting to notice is the high part of thermophilic processes and the higher average methane content for co-digestion plants. (Energimyndigheten, 2008)

Table 2 Number of different biogas production plants and their average methane content and biogas production, in Sweden year 2006. (Energimyndigheten, 2008)

Type:	Number of plants:	Number of mesophilic:	Number of thermophilic:	Methane content, Average (%):	Biogas production, (GWh):
Industrial waste	3	3	0	71.2	91.20
Farm-based	8	7	1	64.9	13.88
Co-digestion	14	6	6	67.4	183.90
Landfill	60	-	-	50	342.37
Sewage sludge	138	130	4	64.5	581.83
Total	223	146	11	-	1,213.18

The total biogas production for year 2006 was 1.2 TWh. It is expected to be higher for 2007 and 2008 since the co-digestion capacity has increased during these years. However, the landfill gas production has probably declined during the same time period due to the prohibition of depositing organic material, but not at the same rate. (Energimyndigheten, 2008) In Figure 2 the percental contribution from each biogas production plant type can be seen. Almost 50 % originates from sewage sludge treatment plants while 28 % was produced in landfills and 15 % in co-digestion chambers.

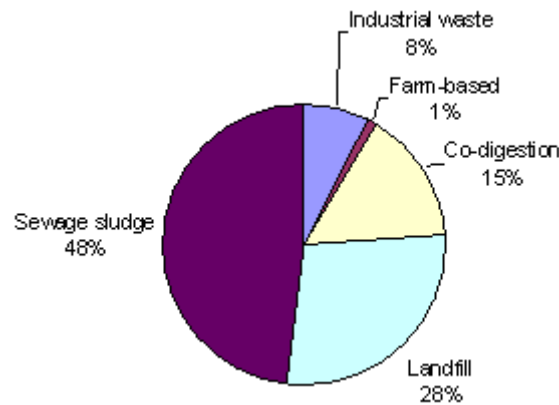


Figure 2 Percental distribution of biogas production from the different types of facilities, Sweden year 2006. (Energimyndigheten, 2008)

2.4 Biogas utilization

The main utilisation of biogas has been internal heat and/or power production. This is especially the case for landfill gas since it contains traces of many different contaminants and N₂ that is difficult to separate from CH₄. When used for heat and/or electricity production the gas does not need to be further processed. (SBGF et al, 2008) Figure 3 shows that more than 50 % of the biogas produced in Sweden 2006 was used for heat production, and 8 % for electricity generation. (Energimyndigheten, 2006)

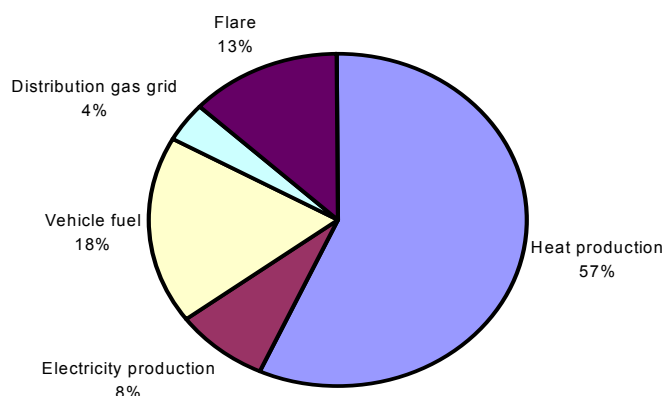


Figure 3 Percental distributions for the biogas utilization in Sweden year 2006. (Energimyndigheten, 2008)

A problem with heat production is that the need of heat varies with season, and during summer unused biogas is flared. In 2006 this amount was as much as 13 %. An alternative that becomes more common is to purify and upgrade biogas to natural gas quality for use in natural gas applications, such as vehicle fuel. In 2006 18 % of the produced biogas was used as vehicle fuel and 4 % was injected into the gas grid. Most of this gas was probably used as vehicle fuel in a later state. (Energimyndigheten, 2008) In this report it is assumed that the final use of the produced LBG is just as vehicle fuel.

2.4.1 Vehicle fuel

Biogas used as vehicle fuel must follow the “Swedish standard for biogas as vehicle fuel, SS 15 54 38”. This standard contains two types, A and B. There are only small variations between them and concerns vehicles without and with lambda regulation. (SGC, 2007) Since a fuel station must be able to supply gas to all kinds of vehicles, only the details for type A (the type with the most stringent requirements) is presented here, see Table 3.

Table 3 The Swedish Standard for biogas as vehicle fuel, SS 15 54 38, type A. (SGC, 2007)

Property:	Unit:	Value:
Wobbe index	MJ/Nm ³	44.7 – 46.4
Methane content	vol-%*	97 ± 1
Water dew point at the highest storage pressure (t = lowest average daily temperature on a monthly basis)	°C	t – 5
Water content, maximum	mg/Nm ³	32
Maximum carbon dioxide + oxygen + nitrogen gas content, of which oxygen, maximum	vol-% vol-%	4.0 1.0
Total sulphur content, maximum	mg/Nm ³	10
Total content of nitrogen compounds (excluding N ₂) counted as NH ₃ , max.	mg/Nm ³	20
Maximum size of particles	µm	1

The different parameters in the standard define the energy content in the gas and the maximum concentration of different trace substances. The wobbe index is a parameter that is used to determine the interchangeability for different fuel gases. This is an important

parameter since both biogas and natural gas are used as vehicle fuel, in the same applications. The CH₄ concentration determines the energy content in the gas since neither CO₂ nor N₂ (in landfill gas) contain any energy. (Näslund, 2003) The listed trace substances can cause corrosion, deposits and wear in compressors, storage equipment and engines if the concentrations are too high. (Persson & Wellinger, 2006)

To be able to live up to the Swedish standard raw biogas needs to be purified and upgraded. This will be treated in the following chapter. One parameter that is not represented in the standard is the siloxane content, however, the purified gas should not contain any of these compounds either. (Persson & Wellinger, 2006)

3 Purification and upgrading

Biogas used as vehicle fuel must first be purified and upgraded. Purification means that contaminants are removed from the gas stream while upgrading means that the energy content is raised through removal of CO₂. In the following chapter conventional purification and upgrading technologies will be briefly described.

3.1 Purification technologies

In the purification step damaging compounds like H₂O, H₂S and particles and, if present, siloxanes and halogenated compounds are removed. This could otherwise cause problems with corrosion, deposits and mechanical wear on the downstream equipment and engines. (Persson & Wellinger, 2006) Below follows a description of the most commonly used technologies for the removal of each component.

3.1.1 Hydrogen sulphide - H₂S

H₂S is formed when organic material containing sulphur is decomposed under anaerobic conditions. It is very corrosive on most metals and the reactivity increase with concentration and pressure, elevated temperature and in presence of water. Also, H₂S can cause problems with bad smell from the upgrading plant. (Persson, 2003)

H₂S can be removed in a catalytic oxidation reaction on activated carbon, forming elementary sulphur, S, and H₂O. By impregnating the carbon with potassium iodide or sulphuric acid the reaction rate can be increased. When saturated the activated carbon can be regenerated or replaced with new carbon. The technology is commonly used when a PSA system is used for the upgrading. (Persson & Wellinger, 2006)

Another alternative for H₂S removal is chemical absorption on a solid material containing a metal oxide. Commonly used metal oxides are iron hydroxide and oxide. Some materials can be regenerated while others need to be replaced when saturated. (Persson, 2003)

3.1.2 Water vapor – H₂O

Digester gas and landfill gas are usually saturated with water vapour. The concentration is increased with elevated temperature and at a temperature of 35°C the water content is around 5 %. Water forms corrosive acids in reaction with CO₂ and H₂S that can damage equipment if it is not removed. (Persson, 2003)

The most common technology for water removal is adsorption on the surface of a drying agent. This drying agent can be zeolites, silica gel, aluminum oxide or magnesium oxide. The drying agent is packed in two vessels and while one is in operating mode the other one is regenerated. (Persson, 2003)

3.1.3 Other components

Siloxanes

Siloxanes are organic silica compounds that occasionally occurs in landfill gas and digester gas produced from sewage sludge. During combustion it is converted to inorganic siliceous

compounds, forming a white powder. Deposits of this powder in the downstream equipment cause extensive damage by erosion and blockage. Siloxanes can be removed from the gas stream through absorption in a liquid mixture of hydrocarbons or with activated carbon. Spent activated carbon cannot be regenerated. (Persson & Wellinger, 2006)

Halogenated compounds

Halogenated compounds are often present in landfill gas. During combustion it forms a corrosive product and, under certain conditions, dioxins and furans are formed. Halogenated compounds can be removed through adsorption on impregnated activated carbon. (Persson, 2003)

Dust and particles

Dust and particles are removed with filters with different mesh size. These filters also remove droplets of water and oil. (Persson & Wellinger, 2006)

3.2 Conventional upgrading technologies

There are a number of different technologies for the separation of CO₂ from CH₄ and the most common solutions are; absorption, adsorption and membrane separation. These three technologies will be briefly presented below.

3.2.1 Absorption

The absorption technology uses the differences in binding forces for different molecules to separate CO₂ and H₂S from CH₄. CO₂ and H₂S are more polar and therefore more soluble in a polar absorption fluid than the non-polar CH₄. Examples of absorption fluids are water and organic solvents. (Persson & Wellinger, 2006)

Water scrubbing

The most common upgrading technology in Sweden is water scrubbing, where water is the absorption fluid. The gas is compressed and fed into a column where it meets a counter flow of water. The column is filled with a package material to create a large surface between the gas and the liquid. Both CO₂ and H₂S are more soluble than CH₄, and can thus be separated from the gas stream. However, H₂S accumulates in the water and can cause corrosion and plugging of pipework. Also, it can cause problems with bad smell if it is vented to the atmosphere. Therefore, it is recommended to remove it before the absorption column. Upgraded gas leaving the column has a methane content of around 97 % and is saturated with water. (Persson & Wellinger, 2006)

Water leaving the column is enriched with CO₂, but also small amounts of dissolved CH₄. Leading the water through a flashing tank, where the pressure is reduced, makes this CH₄ to leave. The vent from the flashing is then sent back to the gas injection, reducing the CH₄ losses in the processes. (Persson & Wellinger, 2006)

The process can recirculate the water or it can use fresh water continuously. The latter is common at places with large water supply, for example sewage water treatment plants. (Persson & Wellinger, 2006) If water is recirculated, it is treated in a stripper column to remove CO₂ and other contaminants. An example over a process scheme of a water scrubber, with recirculating water, can be seen in Figure 4.

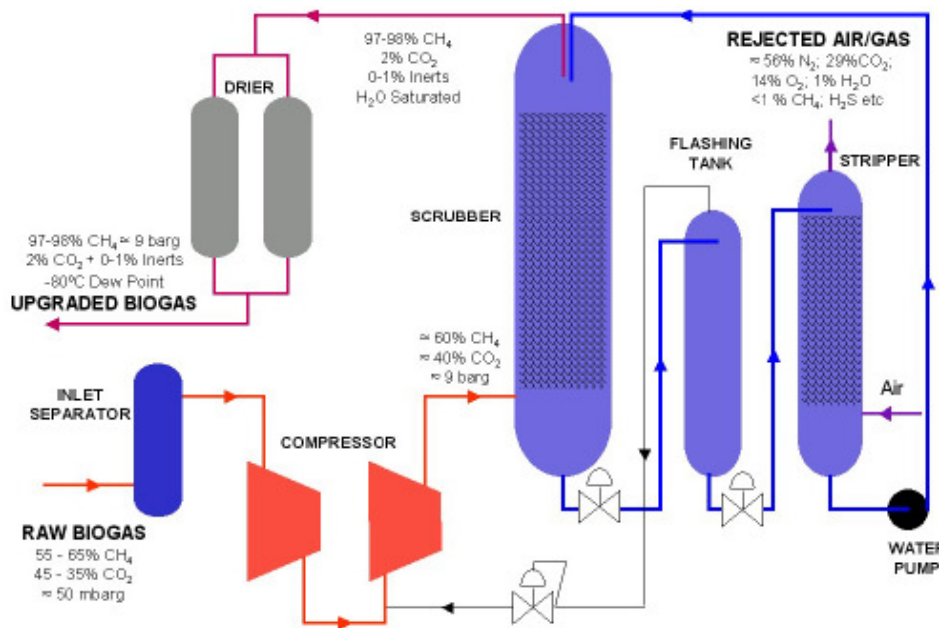


Figure 4 Process scheme over a water scrubber with recirculating water, developed by Flotech. (Flotech, 2006)

Organic solvent

Another type of absorption process is to use organic solvents as absorption fluids. These organic solvents can be polyethylene glycol or alkanol amines, where Selexol[®] and Genosorb[®] are examples of the first and LP Coaab[®] is an example of the latter. The reason to use an organic solvent is that CO₂ is more soluble in it, which results in smaller plants for the same gas flow. The solvent is regenerated in a regenerating process. (Persson & Wellinger, 2006)

In this report this technology is represented by the LP Coaab[®] process, developed by Purac/Läckeby Water Group, since it is represented in an upgrading plant in Gothenburg. LP Coaab is the name of the technology but also the name of the absorption liquid, a specifically composed amine. (Purac, 2008)

A sketch over a LP Coaab process can be seen in Figure 5. Saturated Coaab is regenerated in a CO₂ stripper, where heating of the absorption liquid makes CO₂ to leave. The process is a fully reversible chemical absorption process. (Purac, 2008)

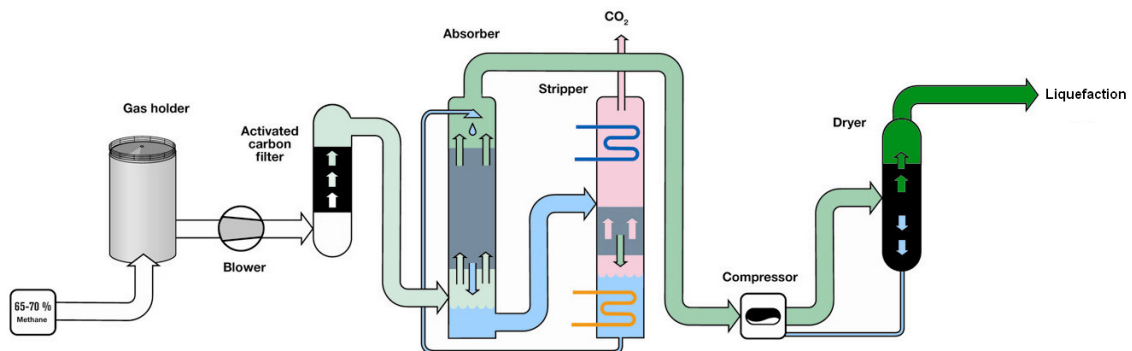


Figure 5 Process flow diagram over LP Coaab. (Karlsson, 2008)

3.2.2 Adsorption

In an adsorption process CO_2 is adsorbed on a material, like activated carbon or molecular sieves. The molecules are trapped in pore spaces in the adsorbent and the separation takes place due to different mesh size of different molecules. The most common adsorption processes, and the second most common upgrading process in Sweden, is Pressure Swing Adsorption, PSA, where the adsorption takes place at elevated pressure. Another type is Temperature Swing Adsorption, TSA and in this process the adsorption takes place at elevated temperature. (Persson, 2003)

Pressure Swing Adsorption - PSA

As mentioned above the adsorption takes place at an elevated pressure and separation is possible due to different mesh sizes of CH_4 and CO_2 . The system consists of a number of columns in series, for continuous operation. When the adsorption material is saturated in one column, the gas flow is led to the next, while reducing the pressure regenerates the saturated column. Reducing the pressure makes the adsorbed molecules to leave. The pressure is first reduced to atmospheric and then to a light vacuum. The vent from the first stage contains significant amounts of CH_4 and therefore it is sent back to the gas inlet, in order to keep the CH_4 losses low. In the second stage the vent mainly consists of CO_2 and is vented to the atmosphere. (Persson & Wellinger, 2006)

Before entering the adsorption column the gas needs to be dry and free from H_2S . H_2S is removed by adding an additional column with activated carbon. When the adsorption material is saturated it is exchanged. (Persson & Wellinger, 2006)

3.2.3 Membranes

There are two types of membranes; dry membranes and gas-liquid membranes. Only the first will be treated below. In the latter CO_2 diffuses through the membrane and is absorbed by an absorption liquid.

Dry membranes

Dry membranes are membranes with a gas phase on both sides and the driving force is the differences in partial pressure. Molecules with different sizes have different permeability and when a pressurized gas stream enters the membrane, CO_2 permeates to the low-pressure side, while CH_4 stays under pressure. (Persson & Wellinger, 2006) An example of a membrane can be seen in Figure 6.

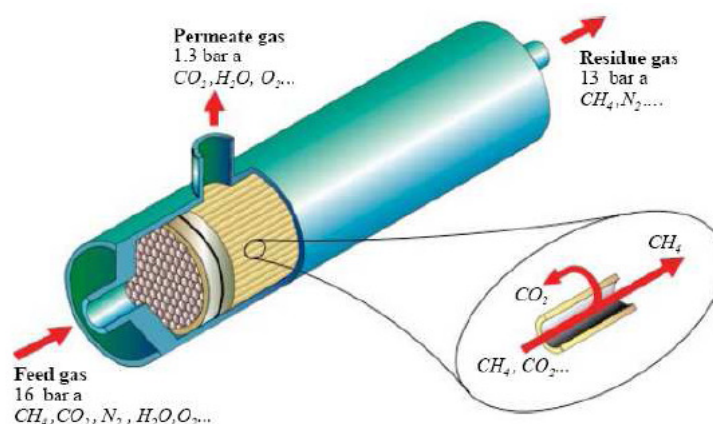


Figure 6 Principle sketch over a MEDAL membrane, a dry membrane from Air Liquide. (Rouaud, 2008)

Also, some CH₄ passes through the membranes. This leads to a conflict between high CH₄ levels in the gas and at the same time a high CH₄ recovery. A larger size of the membrane gives higher CH₄ levels in the outgoing gas stream but more CH₄ permeates through the membrane. Using membranes in series can reduce this problem. The vent from the first membrane is CO₂ rich and sent to a flare while the permeated gas from the second membrane consists of significant amounts of CH₄ and is sent back to the fed gas. (Brown, 2008)

The membranes separate some H₂S but since H₂S is corrosive it is recommended to remove it before the process. Also, the gas needs to be compressed and dried. (Persson & Wellinger, 2006)

One supplier of membranes is MEDAL, a division of Air Liquide. Air Liquide Advanced Technology has recently presented a total system for the production of LBG from landfill gas using these membranes for the separation of CO₂ and CH₄ (more information about this processes can be find in appendix 2). The MEDAL membranes are also used in Acrion's process (see chapter 4.1.2 and appendix 1).

4 Liquid biogas

As mentioned in the introduction to this report cryogenic technology refers to as the development and utilization of low temperatures (Barron, 2005). This technology can be used to purify and upgrade biogas. The theory is based on different condensation temperatures for included compounds in the biogas, which can be seen in Table 4. (Benjaminsson, 2006)

Table 4 Condensation temperatures, at atmospheric pressure, for different compounds in the biogas. (Benjaminsson, 2006)

Compound:	Condensation temperature (°C):
CO ₂	-78.5
CH ₄	-161
N ₂	-196

The technology can be used to upgrade biogas by chilling the gas to around -80°C (atmospheric pressure) but usually *cryogenic technology* is associated with the production of liquid biogas, LBG. Then the gas is further chilled to -162°C. Another product that often is mentioned when discussing cryogenic upgrading technology is liquid CO₂, LCO₂, which is used in many commercial applications (for more information, see chapter 8). (Pettersson et al, 2007)

Another alternative to produce LBG is to use conventional upgrading technology, like the ones described in chapter 3, connected with a small-scale liquefaction plant. Both solutions will be presented and discussed in this text.

Production of liquid biogas is a suitable upgrading technology for landfill gas. Landfill gas usually consists of significant amount of N₂, which is hard to separate from CH₄ with conventional technologies. However, when CH₄ is liquefied in the liquefaction step, N₂ can be separated due to its lower condensation temperature. (Benjaminsson, 2006)

Chilling biogas to very low temperatures is energy intensive but in some occasions the product is more valuable. If the biogas production plant is situated on the countryside, far from the end users, it is more space efficient to transport biogas in its liquid state. Today pressurised (200 bar) gas is delivered in gas vessels stored on a mobile CBG storage, leading to transportation of a huge share of steel, compared to gas. (Pettersson et al, 2006)

Producing LBG also leads to a renewable fuel available for heavier vehicles. The fuel can be stored as LBG on the vehicle, which increase the driving distance per tank. The requirement is that the vehicle is running frequently, otherwise LBG will vapourize and CH₄ will be vented to the atmosphere. LBG is only how the gas is stored on the vehicle. When it gets to the engine it is in its gas phase. (Storarr, 2008)

A variant of cryogenic technology is the production of LBG at pressure letdown stations in the natural gas grid. Here the pressure reduction is used to liquefy a part flow of the gas stream (around 20-30 %). (Pettersson et al, 2007) This alternative will be briefly discussed.

4.1 Cryogenic technology

Cryogenic technology means that impurities, but mainly CO₂, are separated from the gas flow through condensation. In this report three main suppliers of cryogenic technology have been identified. These three suppliers are:

- Scandinavian GtS
- Acrion Technologies/Terracastus Technologies
- Prometheus-Energy

Below follows a brief presentation of these companies, together with a description of their technology (for more technical information, see appendix 1). Two of the three technologies, the ones developed by Scandinavian GtS and Prometheus-Energy, are cryogenic technology. The third technology, developed by Acrion, is a combination of cryogenic technology and commercial technology connected with a liquefier, since it only separate a small part of the CO₂ content through condensation (writers' opinion).

4.1.1 Scandinavian GtS

Scandinavian GtS (SGtS) was formed in May 2007 through a partnership between the Dutch company Gastreatment Services (GtS), a company that works with gas treatment and gas processing and has developed their own technologies for this purpose, and the Swedish company Scandinavian Biogas, working with biogas production optimizing with patented technology and end-to-end expertise of the biogas process. Scandinavian GtS is now working with the development of a technology to produce LBG from landfill gas and digester gas. (Scandinavian GtS, 2008)

Scandinavian GtS uses a concept with four modules in series that can be seen in Figure 7.

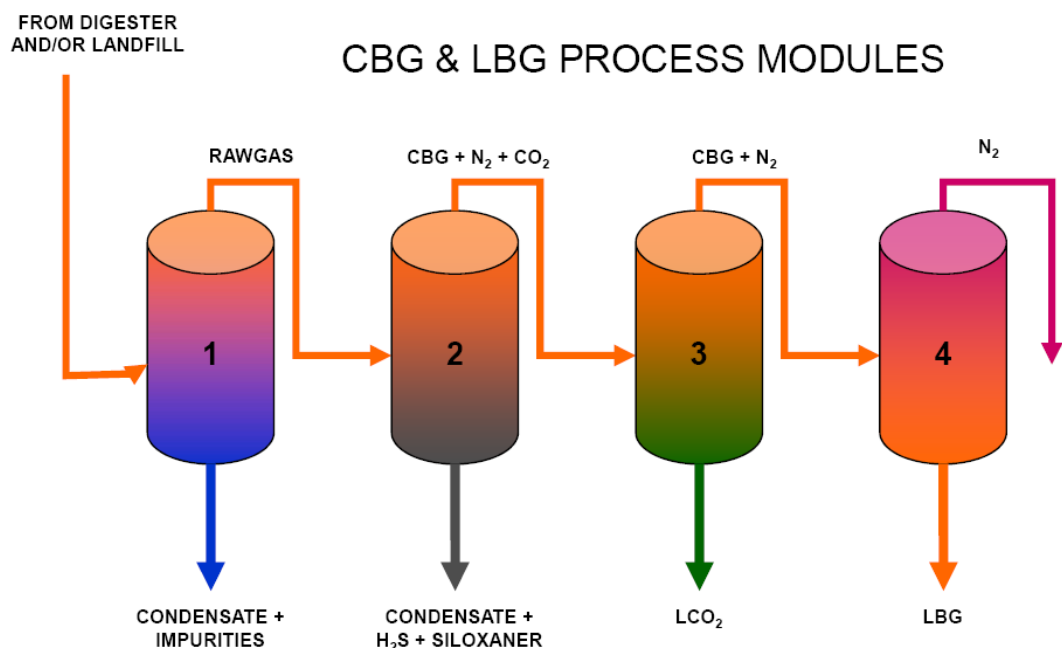


Figure 7 Scandinavian GtS uses a concept with four modules to produce LBG from raw biogas. (Scandinavian GtS, 2008)

In the 1st module the gas is cooled to +6°C. This makes the moisture condense, together with most contaminants. Many remaining contaminants dissolve in the condensed moisture and everything is drained. In module 2 H₂S and siloxanes are removed from the clean gas. The gas is further chilled to approximately -25°C to freeze out the remaining water and to condensate siloxanes. H₂S and the remaining traces of siloxanes are then removed with a SOXSIA[®]-filter. SOXSIA[®] is a catalyst that adsorb siloxanes at the same time as H₂S is converted to elementary sulphur, S, in a regenerating chemical reaction with iron oxide, Fe₂O₃. In the 3rd module, CO₂ is separated from the gas stream by chilling the gas to -78°C and then freeze the CO₂ out. The liquid CO₂, LCO₂, leaving module 3 is sent to a storage tank and will be used as a refrigerant in the process or as a valuable by-product. Gas leaving module 3 is dry and clean and could, after compression, be used as CBG, or further processed in module 4 to produce LBG. By chilling the gas to around -190°C, CH₄ will condense and be separated from N₂, which has a lower condensation temperature. The product is LBG with a CH₄ content over 99 %. (Kättström, 2008)

Combinations of module 1 and 2 have been in commercial use for three years, in around ten existing plants. A pilot of module 1-3 has been running on a landfill in Helsinki during the summer of 2008. The company that used the pilot does not want to reveal the results but according to Mr. Kättström, CEO, Scandinavian GtS, the tryouts “*achieved the purpose*” (2008). This pilot is now in the Netherlands and module 4 has been added to the process. The pilot is ready for show-off and the first commercial plant will take off during second quarter of 2009. (Kättström, 2008)

A commercial plant of module 1-3 will be built in Varberg for injection into the gas grid. Upgraded biogas will be produced from digester gas from an existing digester plant. This plant is at present producing 50 Nm³/h but is built for a higher capacity. The present work is focusing on finding more substrate and the project is still in the planning phase. (Kättström, 2008)

4.1.2 Acrion Technologies/Terracastus Technologies

Acrion Technologies is a small American company whose main area is separation and purification of CO₂ rich gases (> 10%) and the use of CO₂ as a working fluid to achieve separation and purification. In 1998 they received a small business innovation research grant from the U.S. Department of Energy. The purpose was to develop a technology for production of marketable LNG and liquid CO₂ from landfill gas, which resulted in *CO₂ Wash[®]*. (DOE, 2001) AB Volvo holds the license for the production of LNG with Acrion’s technology. Mack Trucks Inc. and Volvo Technology Transfer (VTT), both a part of AB Volvo organisation, have the responsibility for the commercialisation of the technology. Mack Trucks Inc. and VTT have recently started Terracastus Technologies that will work with the specific production of LBG from biogas sources. (Brown, 2008)

Acrion’s technology is a combination of cryogenic and conventional technology. They use a distillation column (*CO₂ Wash[®]*) to clean the raw gas followed by two membranes and a liquefaction step to produce LBG. Before entering the *CO₂ Wash[®]* the gas is compressed, desulphured and dried. H₂S is removed with a *Sulfa Treat* in which it reacts with iron oxide in a non-recoverable process. A sketch over the whole process can be seen in Figure 8. (Brown, 2008)

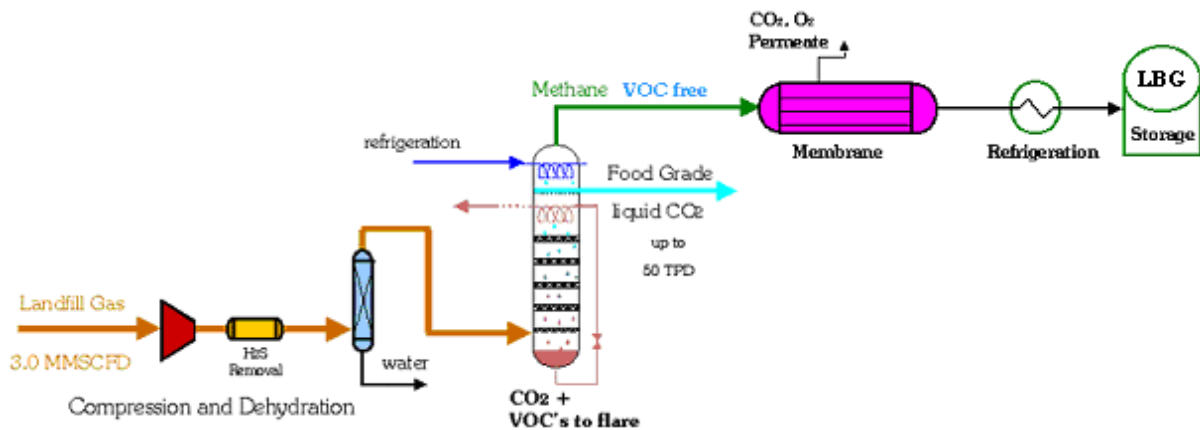


Figure 8 Process flow diagram over Acirion's process for the production of LBG from raw gas. The polishing step for CO₂ is missing in the figure. (Acirion, 2008b)

In the CO₂ Wash[®] a part of the CO₂ content in the gas stream is condensed and most of this is taken out in the top as LCO₂. The rest is used as an absorption liquid in the process, which efficiently removes contaminants such as siloxanes, halogenated compounds and non-methane organic compounds. The adsorbent and contaminants are taken out in the bottom and sent to a flare. At the top a clean gas stream containing CH₄, CO₂, O₂ and N₂, with a CO₂ content of approximately 25 %, exits the column. CO₂ and any existing O₂ are then separated from the gas stream with two MEDAL membranes. (Acirion, 2008a) Permeate from the first membrane is CO₂ rich and is used as fuel or sent to flare, while the second permeate has a high CH₄ concentration and is sent back to the feed gas. Gas leaving the membranes has a CO₂ content of around 1-2 % and before entering the refrigeration module the last CO₂ is removed with a mol sieve (not included in Figure 8). Any existing N₂ is separated and flashed in the refrigeration plant. (Brown, 2008)

In 2005 Acirion produced LBG from landfill gas, processed with CO₂ Wash[®], in a demo project at Burlington County Landfill, New Jersey, USA. The plant produced over 37,850 litres (equal to ~22,300 Nm³) of LBG and, according to VTT, the tryout was “a huge success”. (Elmqvist, 2008) They were able to produce a gas with a methane content over 99.2 % and the purity of the methane gas and liquid CO₂ were continuously analysed by a third part (the results can be found in Appendix 1). (Acirion, 2008b) This pilot project was only a small-scale project producing around 650 –1,100 Nm³/day and the most important experience with it was that they have been able to show that the whole chain works. VTT has just finished the work with designing large-scale plants able to produce 230, 465 and 930 Nm³/h (converted from *gallons/day*, see appendix 3). The technology has not yet been tested in full-scale commercial applications but discussions are held with a number of different companies, among others in Sweden and USA. (Elmqvist, 2008)

4.1.3 Prometheus-Energy

Prometheus-Energy is an American fuel company that produces, sell and distribute LNG. LNG is produced from small, overlooked sources of methane such as landfill sites, stranded gas wells, wastewater treatment facilities and coal mines. (Prometheus-Energy, 2008a) According to Mr. Barclay, Chief Technology Officer at Prometheus-Energy, it is important that it is not index sources, such as pipeline gas, because they do not want to focus on feed stock prices (2008). Their idea is to produce LNG from small-scale facilities close to the

consumers. The company was incorporated Prometheus-Energy Company in 2003. (Prometheus-Energy, 2008b)

Prometheus-Energy's idea is to use commercial technologies in integrated gas purification and liquefaction systems, to produce LBG with a CH₄ content over 97 %. By using basic equipment they minimize costs and manufacturing time and by integrating the different technologies it is possible to increase the overall efficiency. They do not want to reveal their technical characteristics but they build up their systems with a modular approach that can be seen in Figure 9. From this description the general technologies can be derived. (Barclay, 2008)

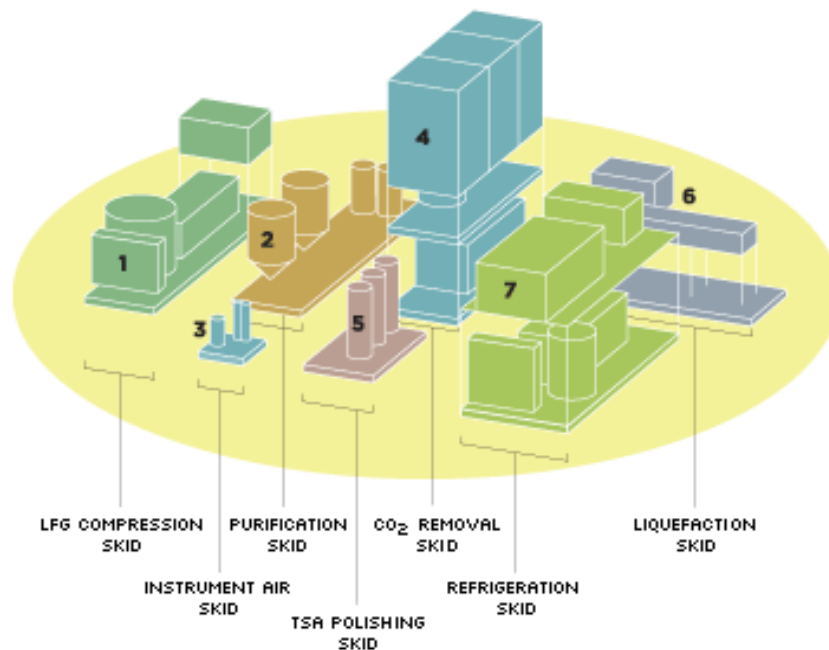


Figure 9 An overlook over Prometheus-Energy's modular approach. (Prometheus-Energy, 2008c)

In the pre-purification module (module 1 and 2) the gas is compressed and water, sulphur compounds and low concentrations of non-methane-organic compounds, including siloxanes, are removed. In the following bulk purification module (module 4) CO₂ is removed from the gas stream by using a proprietary cryogenic freezing technique that freeze out the CO₂ and at the same time pre-cools the methane and any N₂. The separated CO₂ is vented to the atmosphere. After the bulk purification module the dry and clean gas is led to the liquefaction and post-purification module (module 5 and 6). CH₄ is liquefied and after liquefaction the CH₄ concentration is enhanced through dynamic flash evaporation of N₂. The refrigerant module (module 7) provides the cooling to the process through a closed Brayton N₂ cycle. To increase the thermodynamic efficiency of the overall process the refrigerant cycle is designed to maximize the pre-cooling of the LFG gas stream. (Prometheus-Energy, 2008c)

A first pilot plant was built on Hartland Landfill, Victoria BC, in November 2000 producing methane to 96 % purity and 99 % pure CO₂. Since then they have increased the capabilities and performance of their biogas to LBG systems. The facility at Hartland landfill was designed to take care of the separated CO₂. The reason why they excluded this in the present design, according to Mr. Barley, is that the risk for legal actions in the USA is too high and the insurances are very expensive. They could however produce a gas with 99 % CO₂ "fairly

easy” and he adds that it could be a good and interesting alternative for projects in other countries, for example Sweden. (2008)



Figure 10 A picture over the LBG production plant at Bowerman landfill. (Prometheus-Energy, 2008)

In late 2006 a commercial facility for the production of LBG from LFG was installed at Frank R. Bowerman Landfill in California. There have been some technical issues, mainly to purify the landfill gas in an economical fashion. These problems are now solved and the facility has a commercial production of 6,700 Nm³/day (converted from gallons/day) slowly moving forward to the designed capacity of 11,150 Nm³/day. They sell all the produced LBG and a part is used for fuelling a fleet of over 200 buses in the Orange County, California. (Montague, 2008)

Prometheus-Energy is planning to build one more facility, twice this size, but the site is not yet decided. (Montague, 2008)

4.2 Small-scale liquefaction

The second main way to produce LBG is to upgrade the raw gas with traditional technologies, described in chapter 3, and then liquefy CH₄ using small-scale liquefaction technology. To prevent dry ice formation and corrosion in the downstream liquefaction step, the components in the upgraded biogas needs to live up to the concentrations in Table 5. (Pettersson et al, 2007)

Table 5 Maximum component concentrations to be able to liquefy biogas. (Pettersson et al, 2007)

Component:	Requirement:
CO ₂	< 25 ppmv
H ₂ S	< 4 ppmv
H ₂ O	< 1 ppmv

If the upgrading process does not reach these requirements an extra polishing step is needed before liquefaction.

There are several types of liquefaction techniques but here only the ones used for biogas liquefaction will be presented. These liquefier types are either closed-loop or opened-loop cycles. In closed-loop cycles an external refrigerant is used while in opened-loop cycles the refrigerant is a part of the gas stream. (Pettersson et al, 2007) The liquefaction techniques presented are well known and have been in use for several years in the technical gas industry, for example for the liquefaction of natural gas, but in a much larger scale.

4.2.1 Closed-loop cycle

In a closed-loop cycle the refrigerant and the gas stream are separated. The refrigeration can be done with one or more cooling cycles and a sketch over a closed-loop system can be seen in Figure 11.

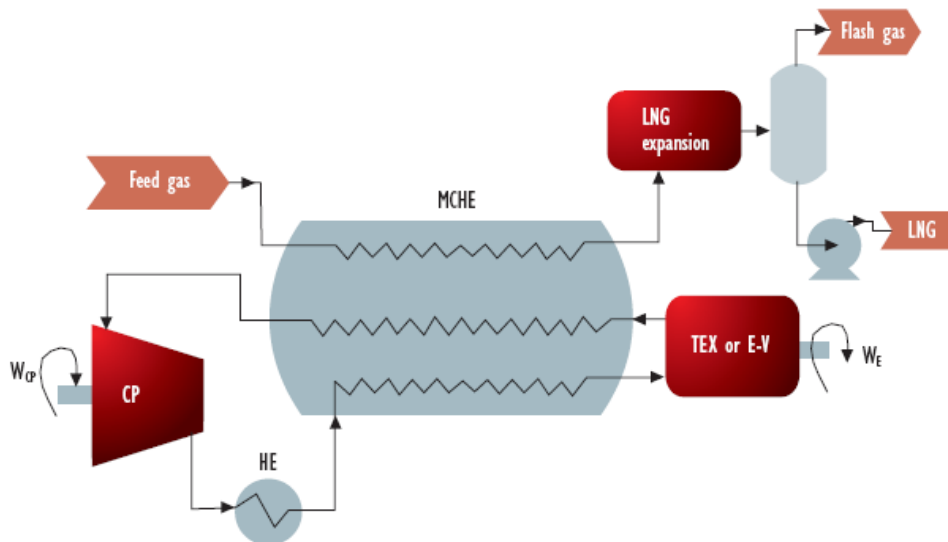


Figure 11 Sketch over a closed-loop cycle. (Pettersson et al, 2007)

The refrigerant is compressed in a compressor (CP) and cooled to room temperature in a heat exchanger (HE) before entering the cryogenic heat exchanger (MCHE) where it is further chilled. After the first pass through the cryogenic heat exchanger the refrigerant is cooled through expansion in an expansion valve (E-V) or in a turboexpander (TEX). In a turboexpander the refrigerant expands in a turbine producing work that can drive the compressor. Since energy is taken out it results in a drastic temperature drop. After expansion the refrigerant enters the cryogenic heat exchanger for the second time cooling the incoming biogas flow. The cycle is closed when the refrigerant returns to the compressor. (Pettersson et al, 2007)

The incoming biogas (feed gas) is cooled and partly condensed in the cryogenic heat exchanger. The remaining gas is condensed through expansion. Any gas that is dissolved in the liquid is separated in the following flash tank. (Pettersson et al, 2007)

Examples of closed-loop cycles are the Nitrogen/Brayton cycle and the Mixed-refrigerant cycle.

Nitrogen refrigeration cycle/closed Brayton cycle

In the Nitrogen cycle, also called closed Brayton cycle, N_2 is the working fluid. The system consists of a compressor, turboexpander and heat exchanger and the biogas is liquefied in the later. The system is simple and robust but has a low efficiency since the cooling curve for N_2 does not correspond to the one for CH_4 . (GTI, 2008a)

One supplier of this type of refrigerant process is Cryostar. They have developed a system they call *EcoRel* to re-liquefy boil off gas, BOG, on LNG ships. The *EcoRel* system is design to cope with big variations in gas flows, it is sturdy, easy-to-operate and allowing a quick start of the process. (Cryostar, 2007) This system is also applicable on small-scale projects on land.

It is built up with one compressor and one turboexpander in a closed Nitrogen cycle, with one electrical motor driving the compressor. The extracted energy in the turbo expander is used to drive the compression side of the machine. The technology is not in use in any projects on the mainland but they have 14 EcoRel-systems on order that will equip the largest ever built LNG carriers. (Rivollier, 2008)

The requirements of the gas to be liquefied are a content of $\text{H}_2\text{O} < 0.1 - 0.2$ ppmv and $\text{CO}_2 < 25-50$ ppmv. Cryostar does not have their own purification technology but they are reviewing different technologies that could be used to remove the last CO_2 . They want to be able to include it in a complete package. (Rivollier, 2008)

Another supplier of a closed N_2 cycle process is Air Liquide and they use this technology in their recently presented process for the production of LBG from landfill gas (for more information, see appendix 2).

Mixed-refrigerant cycle (MRC)

The Mixed-refrigerant cycle consists of multiple stages of expansion valves, phase separators and heat exchangers. (GTI, 2008a) The system has one working fluid that is a mix of N_2 , CH_4 and other hydrocarbons. The mix of refrigerants is designed to match the cooling curve for CH_4 resulting in a continually cooled gas stream and thereby a lower energy demand. However, this process is much more complex than using a single refrigerant, and therefore more difficult to scale down. (Pettersson et al, 2007)

A supplier of this technology is Hamworthy. They hold the licence rights to a patented MiniLNGTM technology for the production of 7,000 – 70,000 Nm^3 LBG/day (converted from *t LBG/day*), developed by SINTEF. They use an energy efficient closed loop mixed refrigerant cycle built up only using standard equipment, which reduce investment costs. (Hamworthy, 2008) The unique with their technology, according to Mr. Jacobsen at Hamworthy, is that they are able to use plate fin heat exchangers in their system, which gives a low energy demand for a liquefaction plant in this scale (2008). Before liquefaction the gas needs to be cleaned and pre-cooled to -10°C . This takes place in separate containers with conventional technology (PSA or TSA). (Jacobsen, 2008)

A fully instrumented pilot plant, five times smaller than a full-scale plant, have been operating since Oct 2003, producing 1,400 Nm^3 LNG/day. The technology is not in use in any commercial projects and they have no written contracts, but there are many interesting projects underway. (Jacobsen, 2008)

Another developer of small-scale mixed refrigerant cycle is GTI. The system is similar to the one developed by SINTEF. Linde BOC holds the license of the technology and a pre-commercial prototype has been producing 2,200 Nm^3 LBG/day (converted from *gallons/day*) over longer time periods. (GTI, 2008c)

4.2.2 Open-loop cycle

In the open-loop cycle the refrigerant is a part of the gas stream. A general sketch over a system can be seen in Figure 12. The biogas (feed gas) is compressed (CP) and then chilled to room temperature in heat exchangers (HE). LBG is then produced in a turbo expander at the same time as work is extracted. Finally any N_2 is separated from the liquid methane in the flash tank. (Pettersson et al, 2007)

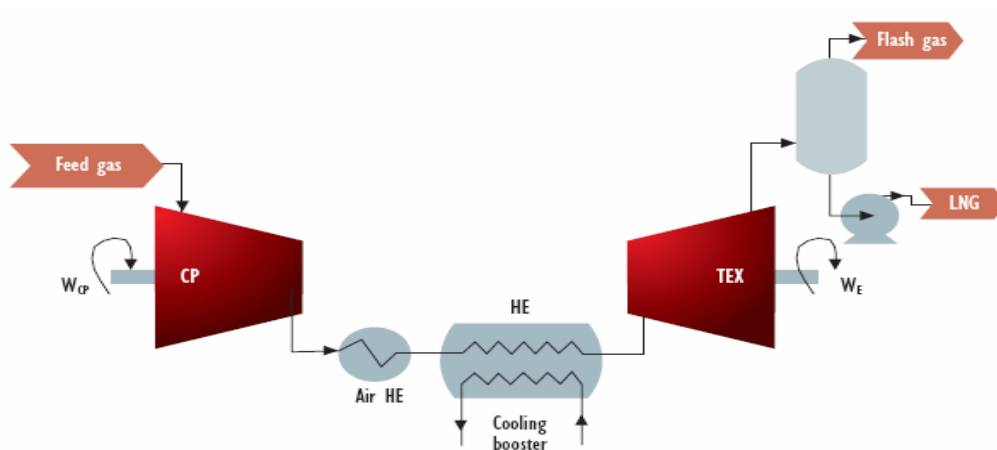


Figure 12 Sketch over an open-loop cycle. (Petterson et al, 2007)

Turboexpander at gas pressure drop

A special application of the open-looped cycle is at pressure letdown stations in the gas grid where high-pressure gas is received and low-pressure gas is sent out. Here the expansion of the gas can take place through a turboexpander. A fraction of the gas stream can then be liquefied with little or no power investment since the work taken out in the turboexpander drives the compressor. This process is very high efficient. (GTI, 2008a)

4.3 Pressure letdown station

If the biogas production and upgrading plant is situated close to the gas network one option could be to inject upgraded biogas into the gas grid and then produce LBG at a pressure letdown station. These stations are situated where the distribution network accesses the transmission pipeline and has the function to reduce the pressure to match the requested commercial distribution pressure. The pressure in the Swedish transmission pipeline has a capacity of 80 bar, but normally the pressure is between 55-65 bar. Most of the pressure letdown stations in Sweden are so called “80-4” stations, which in reality mean a pressure drop from around 60 to 4 bar. There are also a few “80-10” and “80-30” which means that the pressure is dropped from 60 bar to 10 and 30 bar, respectively. (Ahlstrom, 2008)

Biogas is liquefied at the pressure let down stations using the pressure drop to produce electricity driving the process, in whole or in part. The larger pressure drop, the more energy can be extracted. (Pettersson et al, 2007) One developer of this technology is Idaho National Laboratory, INL.

4.3.1 Idaho National Laboratory - INL

INL's technology for the small-scale liquefaction of natural gas at pressure letdown stations is able to liquefy 20-30 % of incoming gas flow. To prevent ice-formation in the downstream equipment, moisture is first removed from the gas stream through injection of methanol. Methanol bonds to the water and the methanol-mix can be separated from the gas stream through condensation. After this step the gas flow is split into two part flows, A and B. A enters a turbo expander that makes the gas expand as the same time as energy is extracted in a turbine, resulting in a powerful temperature drop. The electricity produced is then used to compress part flow B, which then enters a heat exchanger where it is cooled by the cold and expanded part flow A. Part flow A leaving the heat exchanger has a pressure of 4 bar and is injected into the distribution network. (Pettersson et al, 2006)

Part flow B will be further divided into two part flows and, by leading one of them through expansion valves and a heat exchanger tank, one flow will be liquefied while the other one is injected into the distribution network. During the final cooling liquid and solid CO₂ is formed and is separated from the methane stream by a separation tank, a hydro cyclone and a final filter. (Pettersson et al, 2006) The process tolerates a CO₂ content of 2.5 %. Both the methanol-mix condensate and the solid CO₂ are vaporized into the distribution network, not significantly changing the heating value. The hydro cyclone can also separate a part of the higher hydrocarbons such as ethane, propane and butane that exist in natural gas but according to the supplier it is easier to keep these in the LBG. (INL, 2008)

The technology is in use in a plant in Sacramento, California, USA. This facility produce 22,300 Nm³ LNG/day (converted from *gallons/day*), which correspond to 10-20 % of incoming gas flow. (INL, 2008)

5 Delivery and transportation of LBG

When LBG is delivered to remote fuel stations or storages it is transported in vacuum insulated pressure vessels. One manufacture of these semi-trailers is Cryo AB and the dimensions of a standard equipped semi-trailer, suitable for Nordic logistic conditions, can be seen in Figure 13.

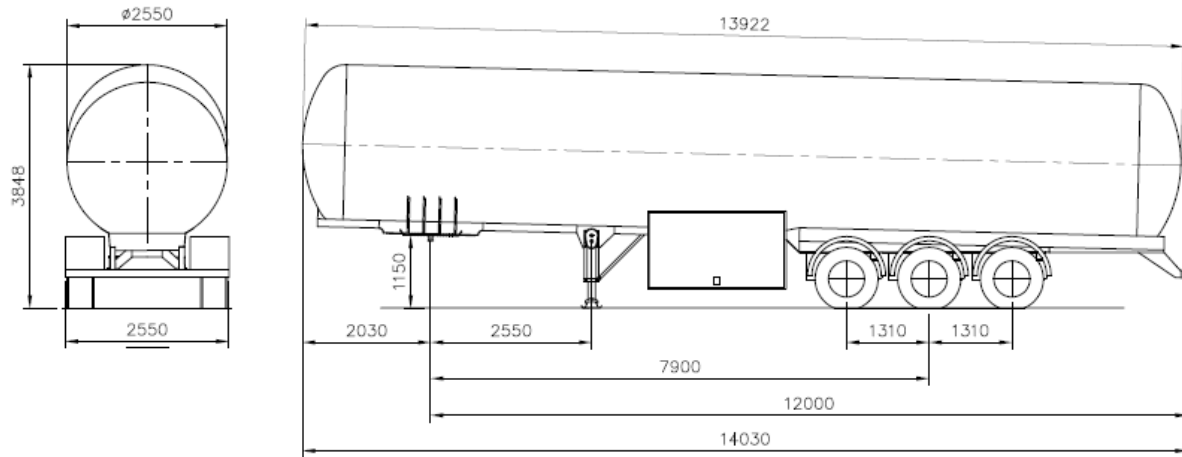


Figure 13 A standard equipped semitrailer/full trailer for the transportation of LNG/LBG. (Cryo AB, 2008)

This trailer is optimized for the transportation of LNG/LBG and has a tank capacity of 56,000 litres (~33,000 Nm³ LBG). It is vacuum insulated and the heat in-leakage is less than 0.9 % of maximum payload LBG per 24 hour. The maximum payload is 83.7 % filling rate at 0 bar (g) (=19,730 kg). The source of heat is the surrounding air and the heat in-leakage raises the pressure of the LBG. The maximum working pressure is 7.0 bar (g). If this pressure is exceeded gas is vented to the atmosphere through a safety valve. (Cryo AB, 2008)

6 Fuel station technology

There are three different types of fuel station available, using LBG as a feed stock:

- LBG refuelling station
- LCBG refuelling station
- Multi-purpose refuelling station

LBG stations fuel LBG to vehicles equipped with a cryogenic tank while LCBG stations refuel CBG. LCBG stands for *liquid to compressed biogas* and LBG is transformed to CBG at the refuelling station. Multi-purpose refuelling stations are able to fuel both LBG and CBG, and consist of one LBG part and one LCBG part. (Vanzetti Engineering, 2008a)

There are a number of companies in the LNG business working with the development of fuel stations using LBG as a feedstock. The presented data in this text is based on information from three different companies; Cryostar, Nexgen Fueling and Vanzetti Engineering. This report will focus on the multi-purpose station and since the three companies' designs are very similar, only a general description will be presented.

The reason why the multi-purpose station is chosen is because LBG could be a good alternative for heavier vehicles. Here it is assumed that these vehicles already are available and in use on a large extent. The refuelling station assumes to be situated in conjunction with one of the frequent roads in Sweden, not in vicinity with the gas network. The following requirements lie as a background for the design:

- Possibility to fuel both LBG and CBG
- One double dispenser for CBG; one nozzle for vehicles (NGV-1) and one nozzle for busses (NGV-2)
- One single nozzle for LBG
- Expected volume of sale: 3000 Nm³/day
- Pressure on CBG: up to 230 bar (200 bar at 15°C)

This proposal is the same as the one in SGC's report *SGC 177*, except for the expected volume of sale. Here it is assumed to be 3,000 Nm³/day instead of 2,000 Nm³/day. (Pettersson et Al, 2007)

The standard equipment on the multi-purpose station consists of a storage tank for LBG, cryogenic pumps, ambient vaporizer, odorant injection system and dispensers. (Cryostar, 2008a)

There are three types of cryogenic pumps:

- Reciprocating
- Centrifugal
- Submerged

Reciprocating pumps are able to function at very high pressures and are therefore used for the filling of buffer tanks and gas cylinders. Centrifugal pumps are able to produce high flow rates and are used for the transfer of cryogenic liquids between reservoir tanks or road tankers. (Cryostar, 2008b) A submerged pump is a centrifugal pump installed inside a vacuum

insulated cryogenic tank. This tank is totally submerged in the cryogenic liquid, which makes it stay in permanently cold conditions. (Vanzetti Engineering, 2008b)

A sketch over a multi-purpose station can be seen in Figure 14. LBG is stored in a vacuum insulated cryogenic vessel and LBG is delivered with semi-trailers. The volume of the storage tank is usually designed to match refilling on a weekly basis. The transfer from trailer is either done by gravity or by transfer pumps, the latter significantly reducing transfer time. (Vanzetti Engineering, 2008a) From the LBG storage tank the station is divided into two; the LBG part and the LCBG part.

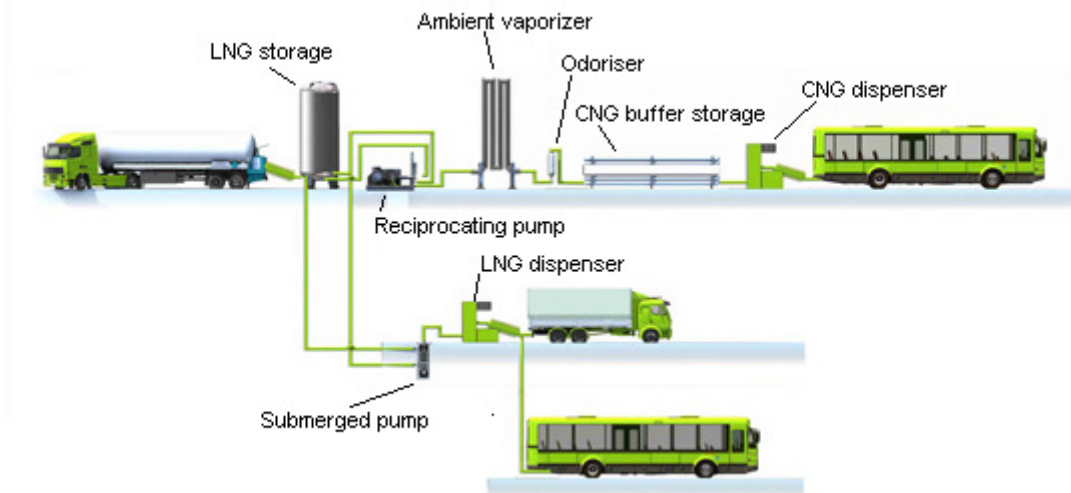


Figure 14 Sketch over a multi-purpose refuelling station (Vanzetti Engineering, 2008a).

The LCBG part consists of a reciprocating pump, an ambient vaporizer and a buffer storage. The reciprocating pump sucks LBG from the storage tank and raises the pressure to around 300 bar, before sending it to the ambient high pressure vaporizer. CBG is then odorized before going to the CBG storage and the dispenser. The buffer unit is a gas vessel storage, with a maximum working pressure of 300 bar, enabling fast filling of vehicles. (Nexgen Fueling, 2008)

The LBG part only consists of a centrifugal pump that transfers LBG from the storage tank, through vacuum insulated lines, to the LBG dispenser that dispense LBG at a pressure of 5-8 bar. (Nexgen Fueling, 2008) Some LBG dispensers are supplied with a system for the recovery of the vehicle boil of gas. (Cryostar, 2008a)

To reduce methane losses all venting lines are collected and sent back to the higher parts of the storage tank, to be reliquefied by the cold LBG. (Heisch, 2008)

7 Commercial LBG production plants

Linde BOC (The Linde Group) has developed a process, for the production of LBG, that is or will be used in two commercial projects where LBG is produced from landfill gas. However, Linde do not sell their technology and equipment. Their concept is to sell a product (LBG) taking all the responsibility for the production. They participate in projects by designing them and then they own and run the facility. They have a knowledge base at the company that they do not want to share and since they do not sell their technology they are not willing to reveal or promote it. However, a general description reveals that they use conventional technologies for the purification of the raw gas. In a first stage sulphur is removed and the gas is compressed. Then the gas is dried in a PSA or PSA-type system before a series of steps to remove VOC, CO₂ and N₂. The purification process is finished with a polishing system, a PSA-type system, for the removal of the residual CO₂. (Carson, 2008)

7.1 Biomethane project at Albury landfill, Surrey, U.K.

Gasrec is a British company producing compressed and liquid CH₄ fuels. They produce the CH₄ fuel from landfill sites or from digester plants. Together with BOC, a member of The Linde Group, and SITA UK, one of UK's leading recycling and waste management companies, they are producing LBG from landfill gas at Albury landfill. SITA UK owns and runs the landfill and collects the raw landfill gas while BOC provides plant operation and maintenance services, risk management and engineering expertise. The Linde Group provides the purification and liquefaction technology. (Gasrec, 2008)

On 18th of June 2008 they announced the first successful production of LBG at Albury Landfill. The plant will produce around 7 million Nm³ (converted from *tonnes*) of LBG per annum, with a 85 % methane recovery of the methane content in the raw landfill gas. This is enough to fuel up to 150 heavy goods vehicles or up to 500 light goods vehicle. Gasrec will sell the gas to the end users while Hardstaff Group, a U.K. company working in the road transport industry, and a developer of natural gas vehicle technology with their own patented dual fuel technology; OIGI[®] (Hardstaff group, 2008), will manage the transport logistics and fuel transportations. The LBG is already used in Albury's haulage trucks, which use Hardstaff group's dual fuel system. (Gasrec, 2008)

7.2 Altamont landfill, Livermore, California, USA

In April 2008 The Linde Group and Waste Management Inc. (North America's leading waste managing and recycling company) announced that they would build the world's largest plant for production of LBG from landfill gas at Altamont landfill, Livermore, California, USA. The plant will produce up to 29,000 Nm³ (converted from *gallons/day*) LBG a day and will begin operating in 2009. The Linde Group delivers the technology; and will stand for the engineering of the plant as well as for the purification and liquefaction; while Waste Management Inc. will supply the landfill gas. The liquid biogas will be used in Waste Management Inc.'s 300 trash and recycling collection vehicles in California. (The Linde Group, 2008)

8 Liquid CO₂

CO₂ is often referred to as a greenhouse gas, causing climate change on earth through the combustion of fossil fuels, but CO₂ in its liquid or solid phase can be used in many commercial applications. With some of the techniques for the upgrading of biogas, described in chapter 4.1, pure, liquid carbon dioxide, LCO₂, comes as a by-product.

At atmospheric pressure CO₂ is an invisible, odourless and tasteless gas that is heavier than air. The phase diagram in Figure 15 shows at which temperatures and pressures CO₂ is in its solid, liquid and gas phase. An interesting observation is that CO₂ cannot exist in its liquid state under a pressure of 5.1 atm (~5.2 bar). When solid CO₂ is heated, at atmospheric pressure, to -78.5 °C, it sublimates directly to its gas phase, instead of melting. Because of this property the solid phase is usually known as *dry ice*, it does not melt. (Change, 2003)

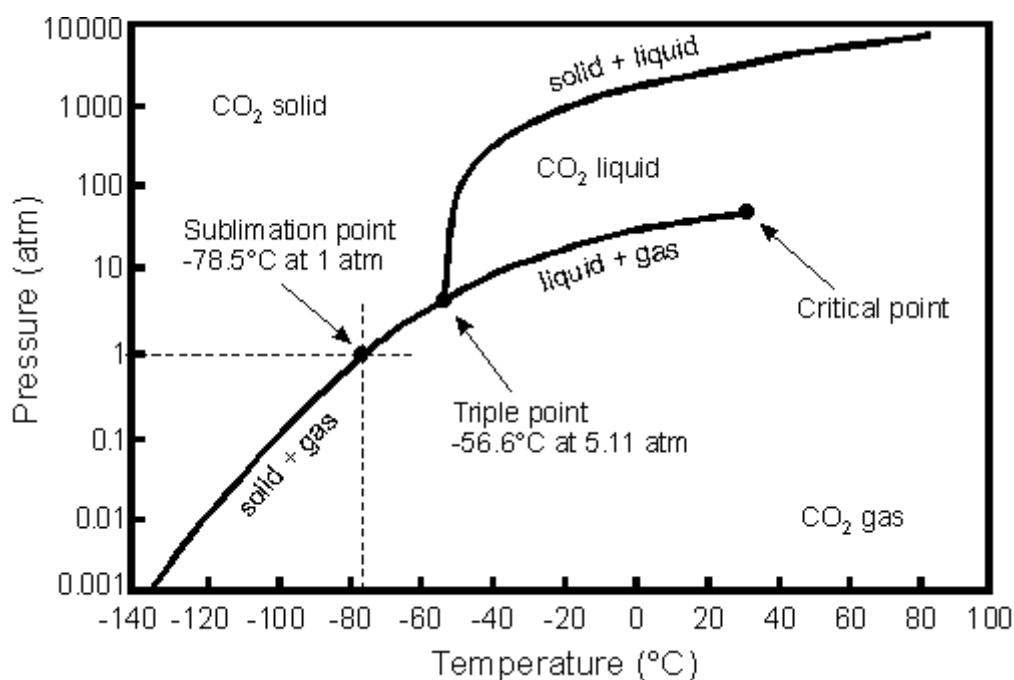


Figure 15 A pressure-temperature phase diagram for CO₂. (Benjaminsson, 2006)

Dry ice is used in commercial applications as a refrigerant. Other examples of possible use are carbonation of fizzing beverages, as a fertilizer in greenhouses, in wastewater treatment and in fire extinguishers were it dispirits the air. (Air Liquide, 2008).

In this report two big suppliers of CO₂ in Sweden have been identified and these are AGA and Air Liquide. Mr. Ljungkvist at AGA, says it would be interesting to extract CO₂ from cryogenic biogas upgrading plants if the amounts are big enough and the facility is not too far away. He point out that CO₂ is a by-product in many industrial processes and that there is a surplus of it. Many companies want to get rid of it to get away from the CO₂ taxes. AGA pays around 0.1-0.5 SEK/kg CO₂ for the raw gas, depending on the treatment needed. Today they get most of their CO₂ from a chemistry industry in Finland were they have their own treatment plant and clean the gas to food grade quality. (Ljungkvist, 2008) AGA has 9 public fuel stations for CO₂ (November 2008) and two more planned (AGA, 2008).

Air Liquide gets all their CO₂ from *The Absolut Company* in Kristianstad. They treat it to food grade quality and, as AGA, they do not pay much for the raw gas. They have received inquiries from biogas upgrading plants but these plants have been too far away to be of economical interest. (Pettersson, 2008)

In many CO₂ applications it is not necessary with food grade quality, as long as the gas does not smell or taste bad or contains any toxic compounds. A problem is however that AGA and Air Liquid cannot transport and store food grade and non-food grade quality gas in the same tanks, meaning that they have to invest in more equipment. If the CO₂ source is not big enough this is not economically sustainable. (Ljungkvist, 2008; Pettersson, 2008) Cryogenic upgrading plants would probably be a small source of CO₂ and therefore it can be assumed that the potential to sell the product to AGA or Air Liquide is small.

There is however a possibility to sell the gas directly to the end user. The benefit with LCO₂ from cryogenic upgrading is that it is very clean from the beginning and does not need much treatment. This means that it probably could be sold much cheaper than CO₂ from AGA and Air Liquide. This report will focus on applications where CO₂ is used as an alternative to fossil fuels. CO₂ is considered as a better alternative if it originates as a by-product and if the production of CO₂ does not influence on the production of the main product. In this way it will eventually be released to the atmosphere, but by using it in an extra step another fossil fuel source can be replaced. (Thermo King, 2003) In this report two such applications have been identified and these are cryogenic transport temperature control and cultivation of vegetables and plants in greenhouses.

8.1 Cryogenic transport temperature control

The conventional way to cool temperature-controlled goods during transportation is to use a diesel-driven cooling system with fluorinated refrigerants. Today many companies want to profile themselves as *environmentally friendly* and using cryogenic temperature-controlled systems is one alternative. The supplier of such systems in Sweden is Ingersoll-Rand (known as H-O Nilsson Service AB). They deliver patented systems from Thermo King. (Pettersson et al, 2007) A figure over a truck equipped with a CO₂ cooling system can be seen in Figure 16.

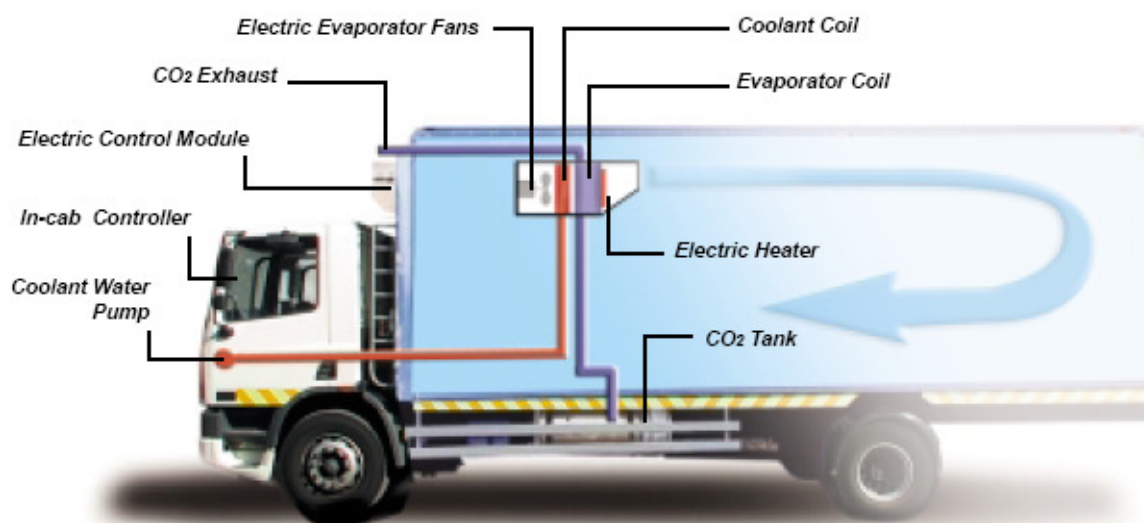


Figure 16 A full cryogenic system, installed in a truck, using 100% CO₂ for the transport temperature control. (Thermo King, 2003)

Liquid CO₂ from the storage tank is evaporated in the evaporator coil. The evaporator coil is sealed so that no CO₂ goes into the cargo space. The evaporated CO₂ absorbs heat from the temperature-controlled air that is circulated over the coil and through the cargo space. When all energy is extracted CO₂ is vented over the roof. Heat for defrosting is taken from the vehicle engine coolant. This source of heat is also used if the truck distributes other goods and the temperature outside is low. (Thermo King, 2003)

The system presented above is used by several suppliers of chilled and frozen goods and H-O Nilsson has delivered around 150 pieces. (Smedbro, 2008)

8.2 Cultivation of vegetables and plants in greenhouses

CO₂ fertilization in green houses is a well-known phenomenon, which has been in use for several years. The most common solution is to get CO₂ when burning natural gas or propane for heat generating for use in the greenhouse. It is a cheap source and gas is a clean fuel. However, these fuels are fossil and also, if the combustion is incomplete it results in emissions of CO, NO_x and ethylene (a substance which makes the plants age). Therefore it is getting more common to use pure CO₂ together with an external heat source; usually combustion of biomass but also district heating. Another renewable alternative could be the burning of biogas, but then there is still the problem with incomplete combustion. (Carlén, 2008)

9 Analysis and energy balances

In earlier chapters different technologies and solutions for the production of LBG have been presented, and in this chapter they will be analyzed. The identified cryogenic upgrading technologies will be compared to each other, and with the other technical solutions. The LBG system will then be compared to the present biogas infrastructure.

The main focus in the analysis lies on energy consumption. Important factors that are discussed are CH₄ losses, CH₄ recovery, waste heat recovery and use of LCO₂ in external processes. Different developers and suppliers of purification and liquefaction technology have different approaches, which results in differences, especially for the methane recovery. All original number from suppliers and literature can be found in Table A3 1, Appendix 3: Original data, data conversion and calculations. These have been converted to uniform units and for energy consumption this is to kWh/Nm³ clean biogas. *Clean biogas* is defined as 100 % CH₄. When using conventional upgrading technology the CH₄ concentration is however between 97-99 %, but to enable comparison all numbers are expressed per 100 % CH₄. When reading the analysis, keep in mind that all numbers are supplier's data and not the absolute truth.

Each analysis is completed with energy balances, were the different technologies are compared to each other. In these comparisons all original numbers have been converted to primary energy per Nm³ clean biogas. Primary energy is the energy needed to produce one unit of useful energy, for example heat or electricity. The primary energy factor is defined as the quota between primary energy and delivered useful energy. Included in the primary energy are extraction of fuel, transportations and conversion, transmission and distribution losses. By converting useful energy to primary energy it is possible to compare different energy carriers to each other. (Energimyndigheten, 2006)

In this report the electricity is converted to primary energy using a factor for Nordic electricity mix. This factor varies from year to year depending on water flows in the Nordic hydropower plants. In 2004 the Nordic mix consisted of 49 % hydropower, 25 % nuclear power, 18 % of other thermal power and 8 % other renewable sources, resulting in a primary energy factor of 1.64. In this report a mean value for 1991-2004 is used and this is 1.6. The primary energy factor for a Swedish mix would be slightly higher due to the higher part of nuclear power (around 45 %), which has lower electricity efficiency. (Energimyndigheten, 2006) The primary energy factor for nuclear power is 2.9, compared to 1.18 for hydropower (Persson et al, 2005).

The primary energy factor for conversion of heat to primary energy has been calculated from efficiency data on production of fuel and heat respectively (these data can be found in Table A3 16, Appendix 3). Heat is used in the Cooab process and it is assumed that it is produced in a boiler fuelled with forest fuel, which results in a primary energy factor of 1.16. (Persson et al, 2005) The heat could also be produced from the burning of the biogas, but because of the higher value of biogas, this alternative is not brought up.

The primary energy factor used for diesel fuel comes from life cycle analysis data. (Uppenberg et al, 2001)

The effects of different choices of primary energy factors are examined in a sensitivity analysis. There it is assumed that the electricity is produced on the margin, and that is done by a coal condensing plant. The primary energy factor for this fuel is 2.74, mostly due to the low electricity efficiency. The heat production in the sensitivity analysis assumes to be produced with natural gas. (Energimyndigheten, 2006) All primary energy factors can be found in Table 6.

Table 6 Primary energy factors for different energy carriers and fuels.

Energy carrier:	Technology:	Primary energy factor:	Source:
Electricity:	Nordic mix	1.60	(Energimyndigheten, 2006)
	Coal condensing plant	2.74	(Energimyndigheten, 2006)
Heat:	Forest fuel boiler	1.16	(Persson et al, 2005)
	Natural gas boiler	1.29	(Persson et al, 2005)
Fuel:	Diesel	1.06	(Uppenberg et al, 2001)

9.1 Production of LBG

9.1.1 Cryogenic technology

In this report three suppliers of cryogenic upgrading technology have been identified and their technical solutions have been presented. Both Scandinavian GtS (SGtS) and Prometheus-Energy work with well-integrated systems to increase the overall efficiency. SGtS use their own technologies to separate contaminants and CO₂ from the gas stream while Prometheus-Energy use commercial technologies for the purification of the gas, together with proprietary cryogenic technology to freeze out CO₂. Acrion use a different technology for the purification and upgrading. They use a distillation column (CO₂ Wash[®]) to purify the gas and CO₂ is then separated with membranes and a PSA.

In Table 7 information regarding different CH₄ and CO₂ parameters can be found. Because all CO₂ is removed before the liquefaction of CH₄ the rest in the LBG product is N₂. If the source is digester gas the CH₄ content should be almost 100 %. The CH₄ recovery reveals how much of the CH₄ in the raw gas that becomes LBG. This number is high for both SGtS and Acrion and for Prometheus-Energy there is no information available.

Table 7 Information regarding different CH₄ and CO₂ parameters for the three cryogenic upgrading technologies (for original data, see Appendix 3, Table A3 4).

Company:	CH ₄ content in LBG (%):	CH ₄ recovery (%):	CH ₄ loss (%):	CO ₂ recovery (%):	LCO ₂ purity (%):
Scandinavian GtS	99	>99	0.5	~100	“food grade”
Acrion	99.2*	>99	”None”	30 – 50**	99.99
Prometheus-Energy	>97	Not available	”None”	-	-

* CH₄ content in the gas produced during the pilot project at Burlington County Landfill

** Depends on economics; if the product can be sold or not.

Since CO₂ is freed out when using cryogenic technology it results in low CH₄ losses. Both Acrion and Prometheus-Energy have reported that it does not exist any CH₄ losses in their processes, since all vented CH₄ is collected and treated. *No* CH₄ losses is however not realistic. Usually vented CH₄ is burned in a flare, and to get zero CH₄ losses this burning process must be complete. (Lantz, 2008) A more convenient expression for the CH₄ losses could be *very low*.

A process that is connected to CH₄ losses is the flashing of N₂ and this is treated in chapter 9.1.5, regarding landfill gas. Another aspect that is important is the size of the membranes used by Acrion. A larger size of the membranes means a lower CO₂ concentration in the clean gas stream, but a larger size also means that more CH₄ is lost with the permeated CO₂. To keep CH₄ losses low it is important how the permeate is treated. (Persson & Wellinger, 2006)

Clean LCO₂ is produced in all the three processes but it is only SGtS and Acrion that takes care of it. In SGtS process the CO₂ recovery is almost 100 % while Acrion have a CO₂ recovery of 30-50 %, depending on the economics. If it is not sold SGtS use it as a refrigerant in the process. (Kättström, 2008)

Table 8 shows the electricity demand for the production of LBG from raw biogas and which scales the technology is suitable for. SGtS has the lowest energy consumption for the production of LBG, but they have not yet been able to test their technology, meaning that the numbers have not yet been verified. They only have experience from a pilot over module 1-3. A pilot over module 1-4 has just recently been finished for demonstration. The positive thing with Acrion and Prometheus-Energy is that they have tried their technologies in pilot plants and Prometheus-Energy even has a commercial plant running. The numbers on electricity consumption for Prometheus-Energy is calculated from information regarding the production of LBG at the Bowerman landfill. These numbers only show the magnitude and will not be included in the energy balances, since they are too vague.

Table 8 Electricity demand for production of LBG from raw gas. The specified scales for the technology from Acrion and Prometheus-Energy are the scales on actual available designs (for original and conversion data, see Appendix 3, Table A3 2 and Table A3 5).

Company:	Electricity: (kWh/Nm ³ clean gas)	Drive:	Scales: (Nm ³ raw gas /h)
Scandinavian GtS	0.8	Electricity	50 – 2400
Acrion	1.42*	Electricity	230, 465, 930
Prometheus-Energy**	1.54	LFG fired gas engine	90 – 930

* Mean value from two different sources.

** Based on actual numbers for the LBG production plant at Bowerman landfill (for calculations, see Appendix 3).

None of the technologies use any heat in the processes, but in the refrigeration processes low value waste heat is rejected. SGtS process is designed to reject waste heat with a temperature of 55°C. This heat could be used as process heat, for example in the digester plant (Kättström, 2008). Other alternatives are as space heating or it could be converted to electricity. (Barclay, 2008) How much of the waste heat that can be used is very site specific.

No chemicals are used in any of the three processes, however they handle a number of refrigerants. Prometheus-Energy probably only use one refrigerant, and that is nitrogen or a mixed refrigerant in the refrigerating module. Acrion use two, one in the distillation column and one in the liquefaction plant. The one used in the distillation column does not have to be cryogenic (Brown, 2008). In SGtS process refrigerants are used in module 2, 3 and 4. Some of these can be replaced with LCO₂ produced in the process. How much depends on if there is a buyer of the LCO₂ or not. Selling it would result in a better economy for the facility compared to lower electricity demand if it used as a refrigerant (Kättström, 2008).

Both SGtS and Acrion use iron oxide for the removal of H₂S. The difference is that SGtS use a regenerative technique, which results in the production of solid sulphur, S, while Acrion use

a non-recoverable process. Prometheus-Energy has not revealed which technology they use for H₂S removal.

The drain from module 1 and 2 in SGtS process is sent back to the digestate or the digestion chamber on a digester plant or to the leachate water treatment on a landfill (Kättström, 2008) while Acrion send the contaminants absorbed in the CO₂ to a flare or a thermal oxidizer (Acrion, 2008a) and Prometheus-Energy burn the drain in a flare or, on the landfill, treat it in the leachate water treatment (Barclay, 2008)

9.1.2 Conventional technologies connected with small-scale liquefaction

Conventional Technologies

To be able to compare cryogenic technologies with conventional technologies, connected with a small-scale liquefaction plant, numbers on energy consumption etc. have been collected from actual suppliers of the different technologies. The chosen companies are the biggest suppliers in Europe, in their area, and the companies are Malmberg Water and Flotech, suppliers of water scrubbers, Purac/Läckeby Water, supplier of LP Coaab (amine process), and CarboTech who is a supplier of PSA technology. (Ericsson, 2008) In the following text the companies will be represented with their technology, and not the company name. The contacted supplier of dry membranes has not been willing to hand over this kind of information, so this technology will not be represented in the analysis. The reason why they are presented in the report is because the membranes are used in Acrion's process and also, the supplier recently announced that they have a process for the production of LBG from landfill gas. More information about this process can be found in Appendix 2.

In Table 9 characteristics for each technology can be found. One parameter that is similar for all three technologies is that H₂S is removed before the separation of CO₂. By removing H₂S early in the process corrosion of the downstream equipment, due to H₂S, can be avoided. The PSA also include drying as a pretreatment, while in both absorption processes drying takes place after the CO₂ separation.

Table 9 Characteristics for the different conventional purification and upgrading technologies (for original data, see Appendix 3, Table A3 6)

Technology:	Regeneration:	Pre treatment:	After Treatment:	CH ₄ losses (%)
Water scrubbing	Air stripping	H ₂ S removal	Drying	0.1-1
Cooab (amine)	Heating	H ₂ S removal	Drying	< 0.1
PSA	Vacuum	H ₂ S removal, drying	-	1-2

When it comes to the regeneration all three technologies use different solutions. In this report the water scrubbing system use recirculated water and this water is regenerated through air stripping. The amine absorption liquid is regenerated by heating while the adsorption material in the PSA columns is regenerated through applying a light vacuum.

The CH₄ loss varies between the different technologies, depending on different affecting mechanisms. In water scrubbers some CH₄ is absorbed into the process water. Most of this is recovered in the flashing tank, and is sent back to the gas inlet, but some is lost in the water-regenerating step. One of the suppliers of water scrubbing design their plants for a CH₄ loss of 1 % while the other one claims that they have a process with a CH₄ loss of only 0.1 %. In the

amine process the CH₄ loss is <0.1 %. This is due to that the absorption liquid has a higher selectivity for CO₂, compared to water. The PSA process has the highest CH₄ losses, 1-2 %. Beyond CO₂, also some CH₄ is adsorbed in the adsorption column. During regeneration this CH₄ is released and, while the vent from the first regenerating step is sent back to the gas inlet, the vent from the second stage is vented to the atmosphere. (Persson, 2003)

The regeneration of absorption liquid in the amine process results in a large use of heat. This, and the use of electricity for each technology, can be found in Table 10.

Table 10 Electricity and heat consumption, in kWh/Nm³ clean biogas (100 % CH₄), for different upgrading technologies. (For original data and conversion factors, see Appendix 3 and Table A3 2)

Technology:	Electricity: (kWh/Nm ³ clean biogas)	Heat: (kWh/Nm ³ clean biogas)
Water scrubbing	0.40*	-
Cooab	0.18	0.20 (0.95**)
PSA	0.40	-

* The energy consumption is an average for the two suppliers.

** 0.95 is the gross heat consumption. 0.75 kWh can be reused as low value heat in external processes.

The electricity consumption for water scrubber and PSA is the same and half this size for the amine. However, as mentioned above, the amine process has a high heat consumption. According to the supplier of the amine technology as much as 0.75 of the total 0.95 kWh heat used can be reused as low value heat (60 °C) in for example the biogas production plant or other external facilities. (Karlsson, 2008a) This however requires that a user is situated close by, which is not always the reality.

To be able to liquefy purified and upgraded biogas the concentrations of CO₂, H₂S and moisture cannot exceed 25, 4 and 1 ppmv respectively. (Pettersson et al, 2007) Table 11 shows the actual concentration for each compound after upgrading with the different technologies.

Table 11 CO₂, H₂S and moisture concentrations after purification and upgrading (for original data, see appendix 3, Table A3 7).

Technology:	CO ₂ (%)	H ₂ S (ppmv)	Dew point (°C, 4 bar)
Water scrubbing	2	0.1-5	-80
Cooab	<25* ppmv	< 0.5	-60
PSA	1-3	<1	Not available

* If the upgrading unit is complemented with an extra absorption column.

All three technologies reach the H₂S and moisture requirements but not for CO₂. The amine process can reach a CO₂ content of 25 ppmv but then an extra column has to be added to the process. (Karlsson, 2008a) Another alternative to reduce the CO₂ concentration from around 2 % to 25 ppmv, which can be used by all three technologies, is to add a molecular sieve to the process.

Small-scale Liquefaction Technology

The small-scale liquefaction technologies presented in this report are closed Nitrogen Brayton cycle and a simplified closed Mixed-Refrigerant cycle (MRC). Both systems have one working fluid but, while the first only consists of N₂, the latter consist of a mix of refrigerants.

This mix consists of N₂, CH₄ and other hydrocarbons and is designed to match the cooling curve for biogas to increase the efficiency. The Nitrogen Brayton cycle has a low efficiency since N₂ does not match with the cooling curve for biogas, but it is not as complex as the MRC and only one refrigerant needs to be handled. Since the MRC is very complex it could be hard to scale down but Hamworthy has the license on a small-scale system based on mixed refrigerants. Hamworthy also include a polishing step, a PSA, in their system, for the last removal of CO₂.

The closed mixed refrigerant process has been developed on a research institute and the goal has been to reach a high efficiency and low investment costs by using a mixed refrigerant cycle and standard equipment. (Jacobsen, 2008) The closed nitrogen reversed Brayton cycle, developed by Cryostar, has been developed to reliquefy boil of gas on LNG carriers and is therefore designed to be stable and easy to operate. However, this system can be applied on the mainland and the presented numbers are from real simulations of such a plant. (Rivollier, 2008)

Table 12 shows the energy consumption and scales for the different refrigeration processes. It reveals that the energy consumption for a mixed refrigerant process is significantly lower than for the nitrogen cycle, as the theory stated.

Table 12 Scales and electricity demand to liquefy upgrade biogas with a N₂ or mixed refrigerant cycle (for original information, see Appendix 3, Table A3 1).

Company:	Refrigeration process:	Scales: (Nm ³ clean gas/h)	Electricity consumption: (kWh/Nm ³ clean gas)
Cryostar	Closed Nitrogen reversed Brayton cycle	From 300	0.63*
Hamworthy	Closed Mixed-Refrigerant cycle	300 – 2,900	0.43

* Calculated from simulations of a real case.

9.1.3 Linde BOC and Air Liquide

Both Linde BOC and Air Liquide have developed a process for the production of LBG from raw biogas, but Linde does not want to reveal their technology and Air Liquide could not leave any numbers on energy consumption. However, both companies are big and well established around the world and that is one reason why they are brought up in this report. Linde do also produce LBG in one commercial plant at Albury landfill.

Both Linde and Air Liquide have developed a process with a low CH₄ recovery; 85 % (Carsson, 2008) and 90 % respectively (Rouaud, 2008). Mr Carson at the Linde Group says that they are more concerned about the economic approach and they think it is too demanding to get the last CH₄ out. He adds that getting the extra CH₄ out also could cost more energy than you get and if it is cost efficient depends on the value of the extra CH₄ (2008).

9.1.4 Pressure let-down station

In Sweden today, biogas is only injected into the distribution network that has a working pressure of 4 bar. To be able to take advantage of the pressure drop at the pressure letdown stations the gas needs to be injected to the transmission pipeline with a pressure of around 60 bar. At the moment this is not possible due to transportation regulations for the transmission

network. The owner of the Swedish transmission network, Swedegas, is however reviewing these regulations. (Frisk, 2008)

To be able to inject biogas into the gas grid the heating value must be raised so it matches the one for natural gas. This is made through injection of propane, which is a non-renewable source of energy. Also, when the gas enters the pipeline it will mix with natural gas containing around 10 % of higher hydrocarbons. (Gasakademin, 2008) If not separated from the CH₄ during liquefaction, this will go with the LBG, resulting in a not 100 % renewable product. This last problem can be solved with the so-called “*green gas principle*”. When natural gas is fuelled at one place the quantity of the gas is reported so that the same amount of biogas can be inserted into the system at another place (Region Skåne, 2007).

9.1.5 Landfill gas

Landfill gas often contains significant amounts of N₂, which lower the heating value since it does not contain any energy. N₂ is hard to separate with the conventional technologies described in chapter 3, but this N₂ can be separated in the liquefaction process since it has a lower condensation temperature than CH₄. However, some N₂ is dissolved in the liquefied CH₄. This N₂ is separated through flashing, where the pressure reduction makes the dissolved N₂ to leave. A problem is that also some CH₄ will be released. Therefore, the CH₄ recovery is closely related to the N₂ concentration in the landfill gas. With a high N₂ concentration, more flashing is needed, which reduce the CH₄ recovery. (Barclay, 2008) To keep the CH₄ losses at a low level it is very important how the vent from the flashing system is treated. One way to take care of it is to use it as fuel in a gas engine, producing electricity for the plant.

The landfill gas at the LBG production plant at Bowerman Landfill (Prometheus-Energy) has a N₂ content of 10 %, while the product contains 3 %. The vent from the flashing system contains as much as 50 % CH₄ and it is used as fuel in a gas engine. (Montague, 2008). The N₂ and CH₄ gas is taken out late in the process, as a less valuable product compared to LBG. This result in higher energy consumption per unit LBG compared to if the N₂ concentration in the raw gas is low. Therefore it is important to collect the landfill gas in a way that minimizes the N₂ concentrations. However, then it is a risk that more CH₄ stays in the landfill and eventually results in emissions to the atmosphere.

9.1.6 Energy balances

Chilling gas to very low temperatures is energy intensive and therefore it is interesting to find out which way is most energy efficient. In these balances the energy consumption is expressed in kWh primary energy/Nm³ clean biogas (100 % CH₄) even though the CH₄ concentration in the product from conventional upgrading technology is between 97-99 %. The reason for this is to enable comparison between different technologies. Prometheus-Energy is not represented in the balances since the calculated numbers for their technology are too vague and also, they are based on landfill gas with a N₂ content of 10 %. One supplier that is represented in the balances is Linde BOC. They have left a general number over the energy consumption for the production of LBG from raw gas and, since they have build one commercial plant and have one under construction, it is interesting to compare it to the rest.

All original numbers on energy consumption is converted to primary energy and it is assumed that all processes are driven with electricity bought from the grid, which is converted using

the primary energy factor for a Nordic mix. Heat, used in the Coaab process, assumes to be produced in a boiler, fuelled with forest fuel, and the energy demand for this process is presented both with and without heat recovery. The reason for this is to elucidate the amount of heat that is used in the process. For more information regarding the relationship between electricity/heat consumption and primary energy consumption, see Appendix 3 and Table A3 2.

In Figure 17 the primary energy consumption for the different technologies can be seen. To the water scrubber, PSA and Coaab process, a mixed refrigerant or a N₂ cycle is connected. In the latter, the reduction of CO₂ from 3 % to 25 ppmv is not included.

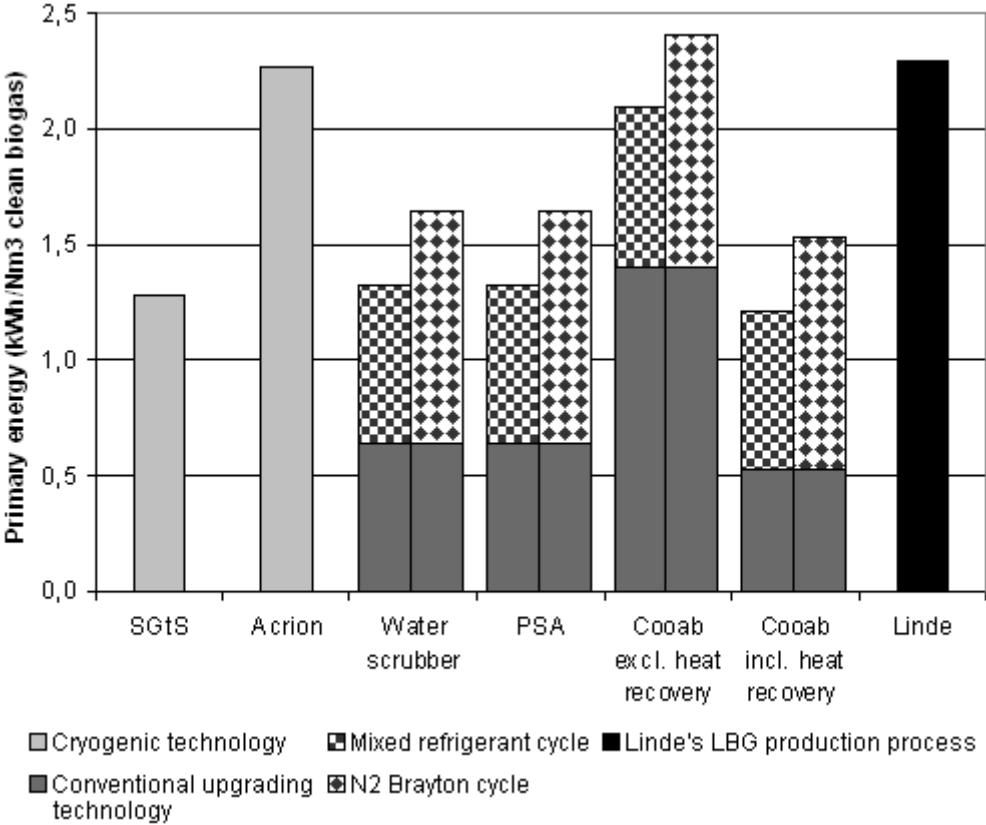


Figure 17 Primary energy consumption for different technical solutions for the production of LBG, where the water scrubber, PSA and Coaab process is connected with a mixed refrigerant or a N₂ cycle.

As mentioned earlier the energy consumption is higher for a N₂ cycle than for a mixed refrigerant cycle. When using a mixed refrigerant cycle the energy consumption for a water scrubber and a PSA system equals the one for SGTs process. If waste heat can be used in an external process, the Coaab process is comparable to these three. An interesting point is the relatively high energy consumption for Acrion’s and Linde’s technologies. Both these have been tested in pilot or commercial plants and maybe these numbers are more realistic.

CH₄ losses

The influence of CH₄ losses on energy consumption has been examined through applying a factor of one unit plus the methane losses to the primary energy consumption for each upgrading technology (for calculations, see Appendix 3). It is assumed that there are no CH₄ losses in the liquefaction step, which is a realistic assumption if the gas does not contain any

N₂. The results from the calculations show that this parameter has a very small influence on the energy consumption. However, loss of CH₄ is a loss of valuable income and also an emission of a greenhouse gas, 20 times stronger than CO₂, so it is important to keep them low.

Waste heat recovery

Another important factor is the use of waste heat in external applications. In the Coaab process, significant amounts of heat is used in the regenerating step and as much as 80 % of this can be used as low value heat in external applications. But also one of the water scrubber suppliers and SGtS has developed a system to reuse rejected heat from the process. These systems are able to recover around 80 % of the inserted energy as low value heat (around 60 °C) (see Appendix 3, Table A3 17).

When the rejected heat is converted to primary energy it is assumed that the heat is used for heating a digester chamber. The need for heat (inclusive hygienization at 70 °C) in such an application, with a mesophilic process, is around 10 % of the energy content in the upgraded biogas, (Berglund & Börjesson, 2003) which corresponds to 0.97 kWh/Nm³. This is twice as much as the rejected heat from the Coaab process. The rejected heat from the upgrading processes is assumed to replace forest fuel in a boiler with a primary energy factor of 1.16. The results can be seen in Figure 18. For calculation, see Appendix 3.

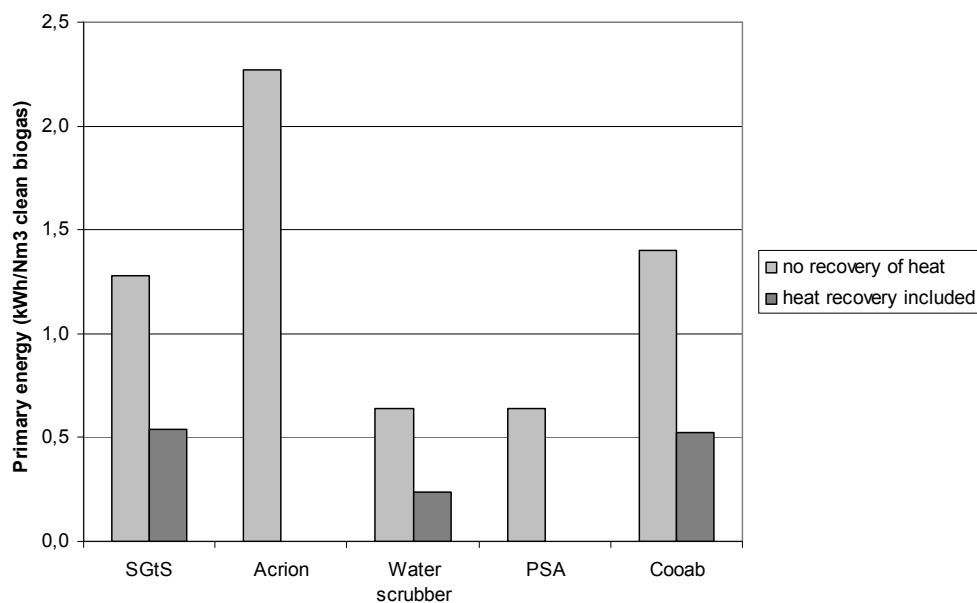


Figure 18 The primary energy consumption with and without heat recovery. The two bars to the left (SGtS and Acrion) show the primary energy consumption for production of LBG while the rest shows the primary energy consumption for upgrading of raw gas with conventional technology.

The use of waste heat in external applications has significant effects on the energy balance. The inserted primary energy is reduced with more than 50 % and this shows that use of waste heat in external applications is an energy efficient way to save energy.

Pressure letdown station

An alternative to produce LBG is to inject upgraded biogas into the gas grid and liquefy a part flow at a pressure letdown station. To be able to do so, the heating value of the biogas must be raised to match the one of natural gas, and the gas must be compressed to around 80 bar. Raising the heating value is achieved through addition of propane. It is assumed that the upgraded biogas has a CH₄ concentration of 97-100 %, depending on upgrading technology, and to reach a heating value of 11 kWh/Nm³ (the heating value for Danish natural gas) around 7-9 % of propane must be added to one Nm³ upgraded biogas.

In the energy balance calculation it is assumed that the liquefaction plant is designed so that no extra energy needs to be added to the process. The primary energy consumption for the production of LBG at pressure let down station is then calculated by adding the compression work of the biogas-propane mix from 4 to 80 bar to the primary energy consumption for the purification and upgrading (for calculations and assumptions, see Appendix 3). The addition of propane is also an energy input, but since this extra energy can be utilized when burned in the vehicle engine, it is not included in the energy balance. If the “Green gas principle” is practiced no considerations has to be taken to the fact that biogas will mix with natural gas in the pipelines.

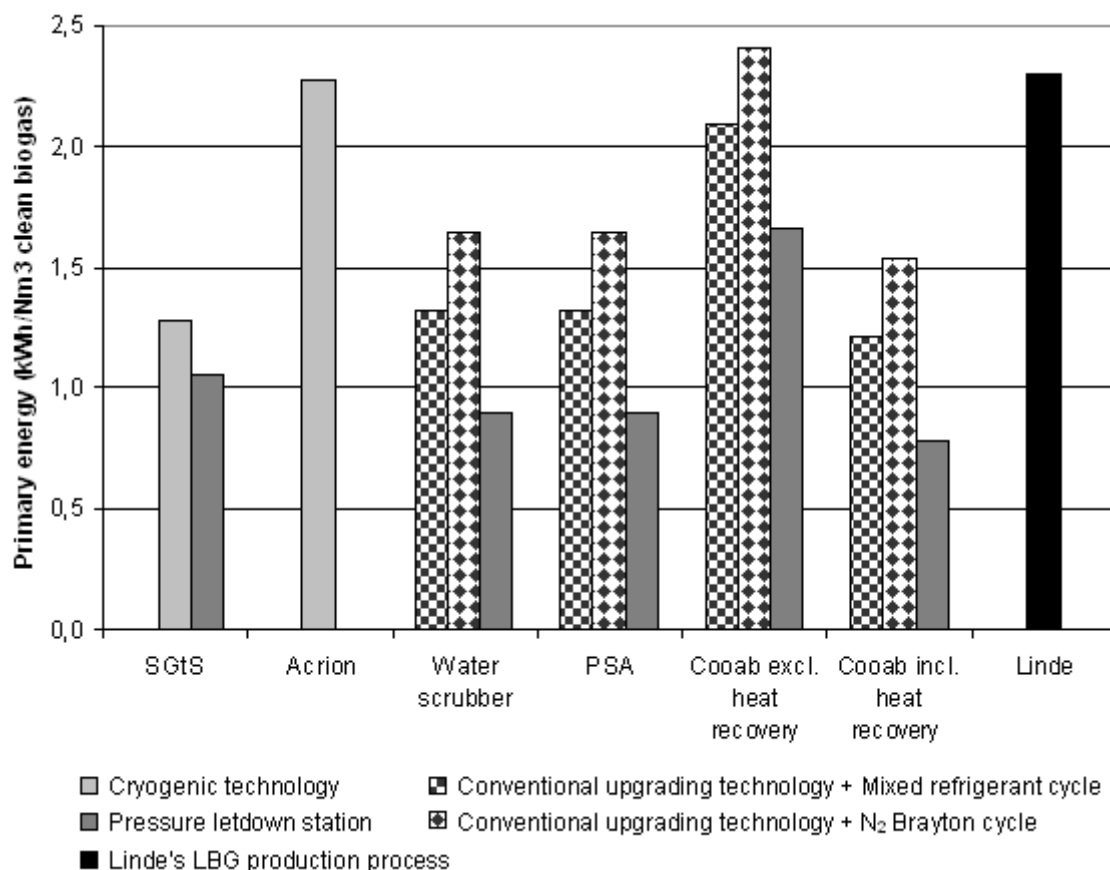


Figure 19 Primary energy consumption for LBG production at pressure letdown stations, in comparison to other LBG production alternatives. These other alternatives are cryogenic technology and conventional upgrading technologies connected with a mixed refrigerant or N₂ Brayton cycle. Included in the primary energy consumption for the LBG production at a pressure letdown station is the upgrading of raw biogas and the compression work to 80 bar.

Figure 19 shows the result of the calculations, in comparison to the other LBG production alternatives presented above. The only work added to liquefy upgraded biogas is the compression work for the biogas-propane mix. The light gray bar for SGtS process represents the energy consumption for purification and upgrading, using module 1-3, together with the energy to produce LBG at the pressure letdown station. The result shows that producing LBG at a pressure letdown station is energy efficient.

Sensitivity analysis

To determine the effects of the choice of primary energy factors a sensitivity analysis is made. In this it is assumed that the electricity is produced on the margin, and that is in a coal condensing plant, with a primary energy factor of 2.74. The heat is produced with natural gas instead of forest fuel (for calculations and assumptions, see Appendix 3). The result from this analysis can be seen in Figure 20. The light gray bars show the result when using primary energy factors for a Nordic electricity mix and forest fuel, while the dark gray bars represents the primary energy for electricity and heat produced on the margin. The figure shows that the differences are significant depending on choice of primary energy factors. A conclusion from this is that conversion to primary energy is a good tool to compare different energy carriers within a study, but not in comparison to other studies.

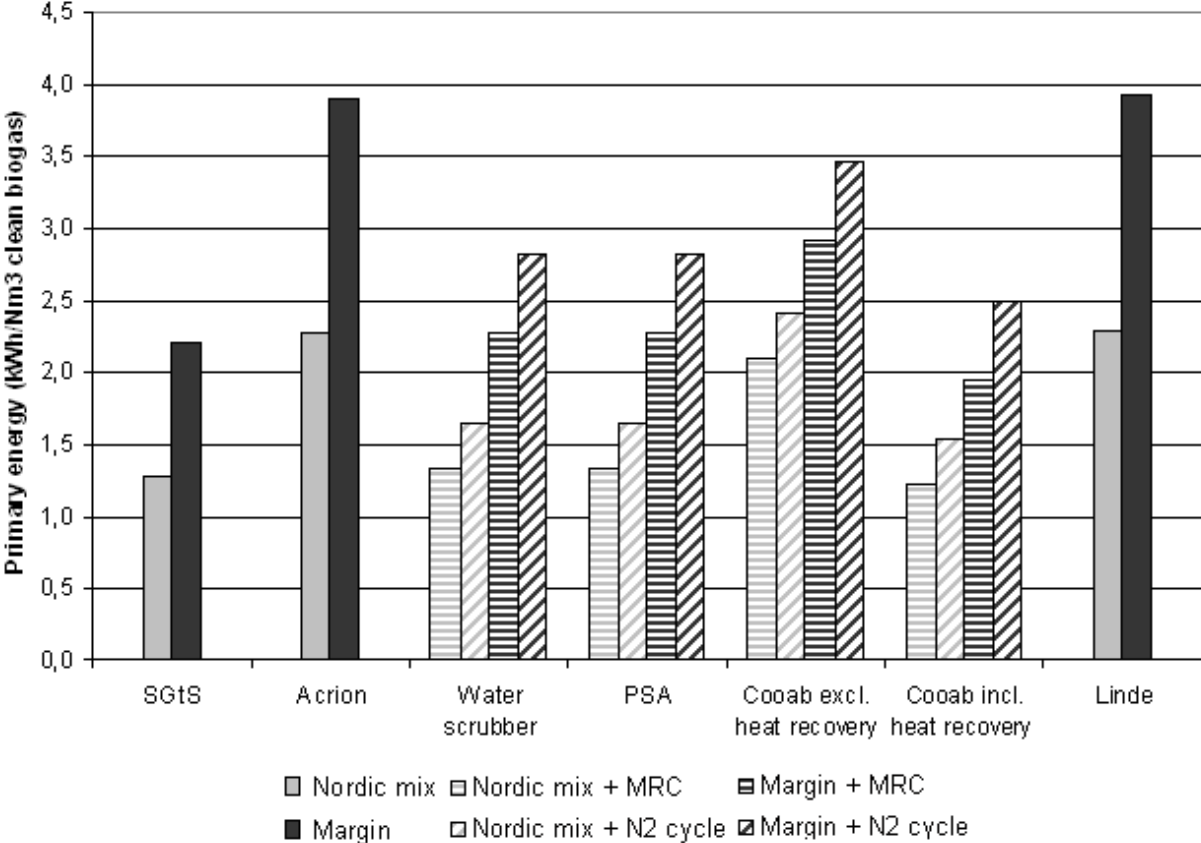


Figure 20 Differences in primary energy consumption, depending on choice of primary energy factor. The conventional upgrading technologies are connected with a N₂ cycle or a mixed refrigerant cycle.

9.2 Distribution and fuelling of biogas

9.2.1 Distribution

LBG is distributed in vacuum insulated pressure vessels and the semi-trailer presented in this report (delivered by Cryo AB) has a storage capacity of 27,500 Nm³. The vessel is filled to 83.7 % at atmospheric pressure to prevent venting to the atmosphere when the pressure is raised, due to heat in-leakage. If the pressure exceeds 7 bar the safety valves opens and CH₄ is vented to the atmosphere. (Cryo AB, 2008) A picture over a LBG distribution vehicle from Hardstaff Group can be seen in Figure 21.



Figure 21 Hardstaff Groups's truck for LBG distribution.

CBG is also transported on road. AGA uses a system where pressurized gas (200 bar) is stored on mobile storages. Each storage contains 147 gas bottles a 50 L made of steel. The storages are mounted on a steel frame and covered with a fireproofed cover. A picture over a mobile CBG storage from AGA can be seen in Figure 22. This distribution system results in transportation of a huge share of material compared to the share of biogas. Only 5 % of the total weight of the transport is biogas and the system is a very inefficient way to deliver gas. (Schröder, 2008)



Figure 22 To the left; a mobile CBG storage with a fireproofed cover and to the right; gas bottles arranged in the storage. (Schröder, 2008)

One truck transporting CBG can take 3 mobile CBG storages per trip resulting in a gas load of 5,160 Nm³. (Schröder) This is more than 5 times less than the load on a LBG semi-trailer (for original data, see Appendix 3, Table A3 9).

9.2.2 Fuelling

The most energy intensive equipments on a multi-purpose station are the pumps. A reciprocating pump is used to transfer LBG from the storage to the ambient vaporizer and at the same time raise the pressure to around 300 bar. The submerged centrifugal pump is used to transfer LBG from storage to the LBG dispenser. The rest of the electricity is used for lightning, control systems etc. In the calculations it is assumed that this electricity demand is the same for every fuel stations, both multi-purpose stations and CBG stations. The electricity consumption presented is therefore only the electricity needed for pumping and compression.

The example of a multipurpose station presented in this report is equipped with a buffer unit for fast filling. This is an important feature on a public station since, if it takes too much time for the customer to fuel the vehicle, he/she will probably chose another fuel. The stations are also equipped with an odorizing system, which is a precaution to enable detection of any leakages.

The multipurpose station is designed for a gas flow of 3,000 Nm³/day. This is a large volume of sale if only cars are refuelled at this station, but in this report it is assumed that the station will be used for refuelling both light and heavy vehicles. 3,000 Nm³/day could for example cover 9 trucks fuelled with LBG and 4 busses and 63 cars fuelled with CBG (for calculations, see Appendix 3). The busses are assumed to be tourist busses and alike. Busses for public transport, running on biogas, are usually fuelled at a specific site.

A CBG station can have many appearances. Biogas can be taken from the gas grid, it can be delivered through a pipe from the upgrading facility or it can be transported with mobile CBG storages. However, somewhere the gas must be compressed to approximately 250 bar. The gas is normally stored in gas vessels and CBG is also odorized before leaving the upgrading plant. (Karlsson, 2008b)

Table 13 shows the energy consumption for pump work on a multipurpose station and the compression work, from 4 to around 250 bar, on a CBG station. The presented numbers for the multipurpose station are averages from three different suppliers. It is approximately 5 times more energy efficient to raise the pressure through pumping than compression. Also, gas stored in its liquid state is space saving and easier to deliver to remote fuel stations.

Table 13 Average energy consumption for a multi-purpose and a CBG station (for original data and calculations, see Appendix 3, Table A3 11 and Table A3 21).

Station type:	Electricity consumption, (kWh/Nm ³ CH ₄):
Multi-purpose station*	
LBG part	0.0027
CBG part	0.055
CBG station	0.25

* Calculated from data from Cryostar and Vanzetti Engineering

9.2.3 Energy balances

Chilling to very low temperatures is energy intensive. Therefore it is interesting to find out when inserted energy for the LBG production is paid back, due to more efficient distribution. It is assumed that both the LBG and CBG distribution vehicle is running on diesel and numbers on fuel consumption comes from each distributor (see Appendix 3, Table A3 20). LBG is produced with SGtS's and Acrion's technologies while CBG is produced with water scrubber, PSA and Coaab, and added to these processes are the energy demand for pumping or compression to 250 bar. In the numbers for the Coaab process is heat recovery included. For calculations, see Appendix 3. Figure 23 reveals that LBG must be distributed 500 – 2,100 km, depending on technology, before the inserted energy is paid back. The consumed diesel fuel per kilometer is shared per loaded Nm^3 gas and added to the energy consumption for production of CBG (200 bar) and LBG respectively.

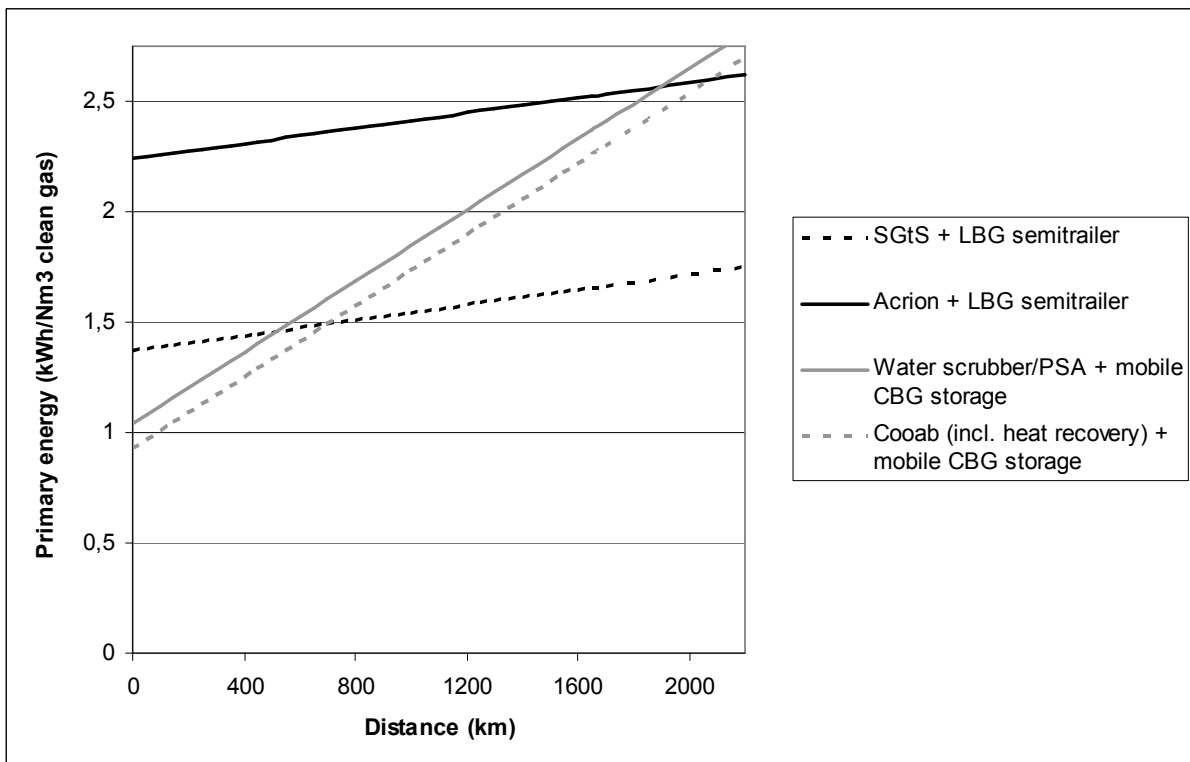


Figure 23 The Y-axis shows the inserted primary energy per Nm^3 clean biogas. The starting point of the lines represent the primary energy consumption for production of CBG+compression to 250 bar (water scrubber/PSA/Cooab) or for production of LBG+pump work on the fuel station (SGtS/Acrion). Added to this primary energy consumption is the inserted energy for delivery with a mobile CBG storage and semi-trailer respectively.

If the inserted energy should be paid back, due to more efficient distribution, the LBG must be distributed over long distances. However, to deliver the same amounts it takes more than five CBG distribution vehicles per LBG semi-trailer. It is therefore more economically efficient to distribute biogas as LBG.

9.3 Liquid CO₂

In this report two applications of LCO₂ have been presented and these are cryogenic transport temperature control and cultivation in greenhouses.

9.3.1 Cryogenic transport temperature control

There are many advantages using cryogenic transport temperature-control instead of temperature-controlling system that is diesel-generated. First, the use of fossil fuels and fluorinated refrigerants are avoided. Further it is very silent, which makes it suitable for urban areas with strong noise regulations and finally, the cooling effect is very good which enables rapid temperature pull-down and recovery (Thermo King, 2003). Because of the very good chilling effect it is suitable to use CO₂ as a refrigerant when transporting small packages on routes with frequent stops and also when transporting frozen goods. For chilled goods the effect needed is lower resulting in a minimum consumption of CO₂ that is bigger than necessary (Rydström, 2008).

Using CO₂ cooling systems is an option for companies that want to profile themselves as *environmentally friendly*. However, a problem could be the scarce access to public fuel stations. The driver of the vehicle fuel diesel (or gas) at one place and have to go to another place to fuel LCO₂, which takes extra time. The best would be if the driver could fuel both diesel and LCO₂ at the same place. (Larsson, 2008)

Another negative experience is the large consumption of CO₂, which makes it very expensive compared to diesel-driven cooling systems. The actual CO₂ consumption differs substantially from the supplier data. The consumption is also very season depended. In the summer the drivers have to refuel every day while the frequency is every third day in the autumn. The consumption is partly dependent on the driver. It is very important to turn of the cooling system when loading otherwise the cooling system is working on full effect. The consumption could also be lowered with the development of better steering and control system (Rydström, 2008).

Since the consumption of CO₂ is a big cost it could be attractive to buy it from another source, maybe to half the market price. Since CO₂ does not go into the cargo space, food grade quality is not required. However, the LBG production plant probably has to invest in an own LCO₂ distribution vehicle to be able to deliver it to the buyer.

9.3.2 Cultivation of plants and vegetables in greenhouses

The common way to get CO₂, and at the same time heat, for greenhouses has been to burn natural gas. However, natural gas is a fossil fuel and, to get a more *environmentally friendly* profile, many gardeners have switched to LCO₂ together with an external source of heat, often burning of forest fuel. This concept is at present much more expensive than burning natural gas. (Carlén, 2008) Therefore, it probably would be interesting to get CO₂ from another source, for maybe half the price.

An interesting solution could be to build a greenhouse in connection to a digester and upgrading plant. The greenhouse could get wasted heat, CO₂ and an organic fertilizer from the

biogas plant. However, the downside of it is that the plants will be dependent on each other. A back up system is probably needed, which would raise the investment costs.

9.3.3 Energy balances

When LCO₂ is used in cooling systems, 20-25 kg of CO₂/h can replace 1.5 litres of diesel/h. How much that can be replaced, per Nm³ clean biogas, depends on the CO₂ concentration in the raw gas and the CO₂ recovery applied by the suppliers. In this report two suppliers, SGtS and Acrion, produce LCO₂ with a CO₂ recovery from 30-100 % (see Appendix 3, Table A3 4). The primary energy that can be replaced for different CO₂ concentrations and recovery can be seen in Figure 24. For calculations and assumptions, see Appendix 3.

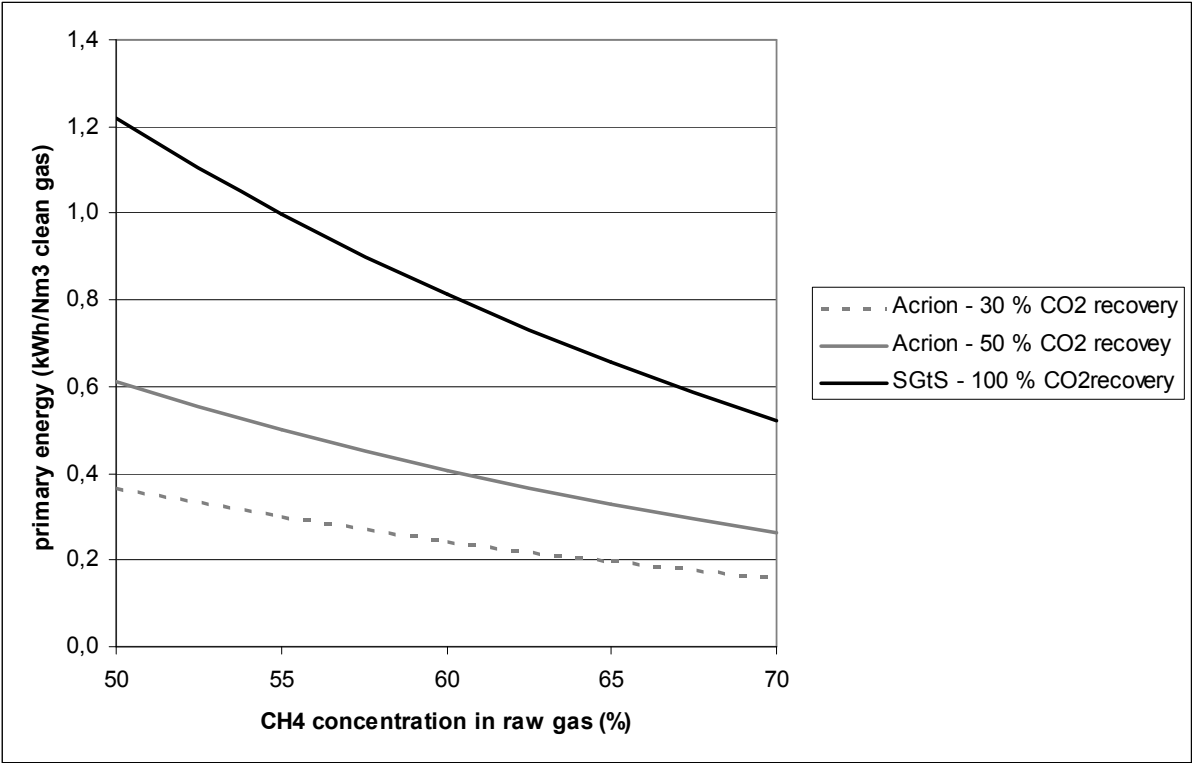


Figure 24 The amount of diesel, expressed in primary energy, that can be replaced per Nm³ clean biogas. The amount varies with CH₄ concentration in the raw gas and with the different CO₂ recovery applied by the suppliers of upgrading technology.

Logically, the amounts that can be replaced per Nm³ clean biogas decrease with decreasing CO₂ concentration and recovery.

In an arithmetic example it is assumed that a digester plant has a raw gas flow of 800 Nm³/h with 65 % CH₄. Based on this data, and the assumption that the CO₂ cooling system and the digester plant is running 10 and 24 h/day respectively, it results in the net primary energy consumptions presented in Figure 25. With a 100 % CO₂ recovery, as in SGtS process, the primary energy consumption is lowered with more than 50 %.

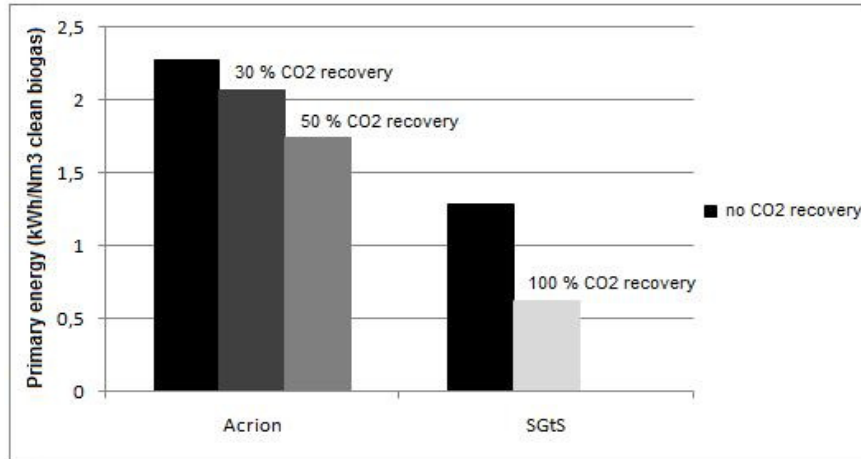


Figure 25 Net primary energy consumption for the production of LBG, for different processes with different CO₂ recovery. The numbers are valid for a raw gas flow of 800 Nm³/h and 35 % CO₂ (for calculations, see Appendix 3).

For each case in the arithmetic example above (30, 50 and 100 % CO₂ recovery respectively), 16, 27 and 53 cooling systems can be supplied. CO₂ cooling systems are most suitable for distribution of goods with frequent stops for loading and unloading, like in the city. In this environment it is also favourable because of its low noise and low emission of harmful compounds. According to Mr. Smedbro (2008) it exist around 100 CO₂ cooling systems on the Swedish market. This amount will probably grow over the next few years but the market is not huge.

The CO₂ consumption in a greenhouse is around 25 kg/(m² and year) and the size of a normal sized greenhouse is around 10,000 m² (Christensen, 2008). The traditional way to get CO₂ for a greenhouse is to burn natural gas. When instead using a pure CO₂ source, the heat from the natural gas burning must be replaced. This is usually done with the burning of forest fuel in a boiler, and since one source of heat will replace a new one, it probably will have very little effect on the energy balance. Therefore, it will not be brought up any further, except for an arithmetic example. However, a fossil source is replaced with a renewable source, which is an important aspect.

If it is assumed that a digester plant has a raw gas flow of 800 Nm³/h with 65 % CH₄, this plant could supply between 6-19 greenhouses, see Table 14 (For calculations and assumptions, see Appendix 3).

Table 14 CO₂ recovery and supplied numbers of greenhouses for the two companies that produce LCO₂ in their process. The numbers are valid for a raw gas flow of 800 Nm³/h and 35 % CO₂ (for calculations, see Appendix 3).

Company:	SGtS	Acrion	
CO ₂ recovery (%):	100	30	50
Numbers of supplied greenhouses (10,000 m ² /greenhouse):	19	6	10

According to the calculations above there is a surplus of LCO₂ from the biogas upgrading production plants. The market for the presented CO₂ applications is probably too small and to spread out to be a possible client. There are however many more fields of applications for CO₂, and if they are situated close by to the LBG production plant, the chances to sell the product increase.

Discussion and conclusions

There are different ways to produce liquid biogas, LBG, and the ways presented and analyzed in this report are cryogenic technology, conventional technology connected with a small-scale liquefaction plant and LBG production at pressure letdown stations. The analysis shows that the process developed by SGtS is twice as energy efficient compared to the other two cryogenic LBG processes. However, SGtS just recently finished a pilot over the whole process so the numbers from this supplier are not as reliable as the ones from Acrion and Prometheus-Energy, who already tested their technology in pilot scale. The numbers from Prometheus-Energy even origins from their commercial LBG production plant at Bowerman landfill. However, since the origin is landfill gas, with a N₂ content of 10 %, and the numbers are vague, Prometheus-Energy is not included in the analysis.

The analysis also shows that the most energy efficient ways to produce LBG are with one of the conventional technologies; water scrubber, PSA or Coaab (including heat recovery), connected with a mixed refrigerant process or with SGtS's process. One explanation to this might be that the conventional technologies have had more time to develop, which have lowered the energy consumption, while SGtS's process and the MRC have not yet been commercially tested, which makes these numbers on energy consumption more optimistically. Figure 26 show how many percent of the energy content in the product that is needed to produce LBG. 100 % represents the energy content in LBG, which is 9.97 kWh/Nm³ clean biogas, while the bars represent the inserted primary energy to produce the product. The conventional technologies are connected with a mixed refrigerant or a N₂ cycle.

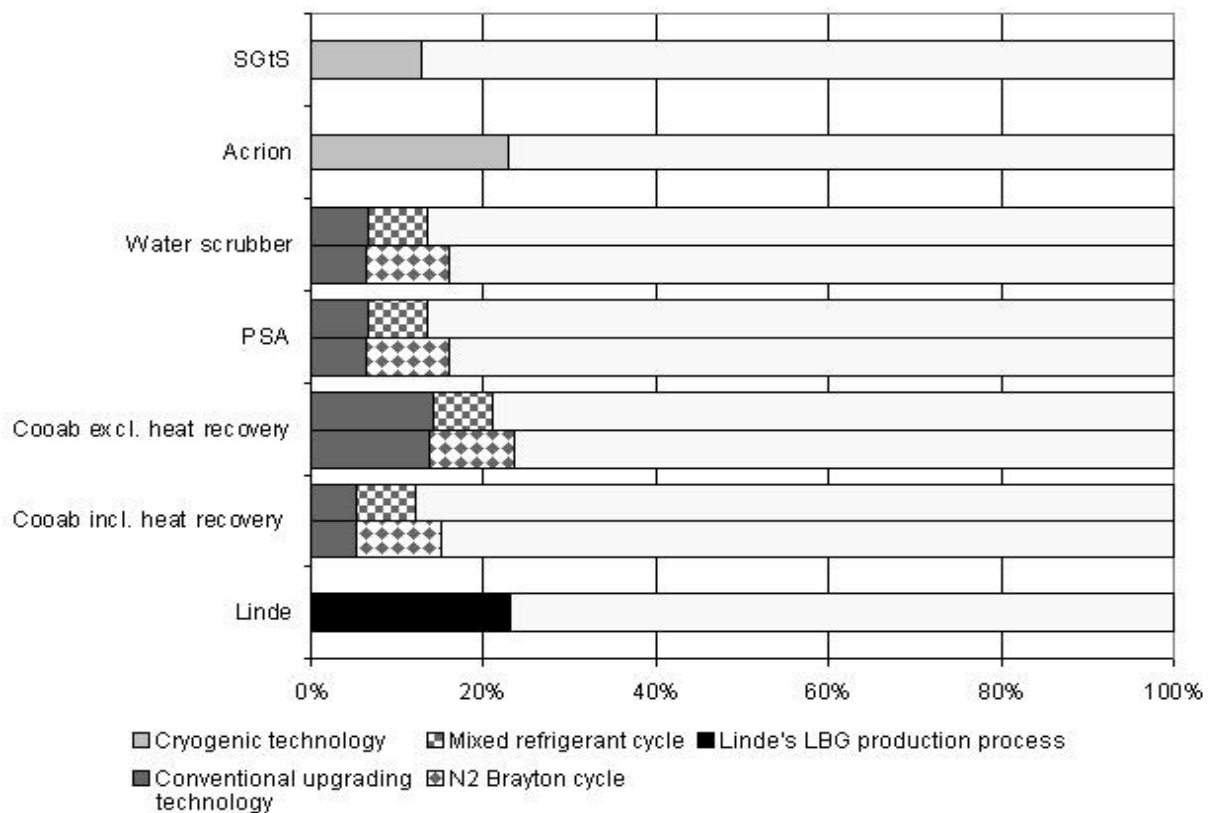


Figure 26 The bars represent how many percent of the energy content in the product that is used to produce LBG. The inserted energy is specified in primary energy and 100 % represents the energy content in the LBG product, which is 9.97kWh/Nm³ clean biogas.

Probably the numbers for Acirion's and Linde's processes are the most realistic. Numbers from these processes are similar to the numbers for Prometheus-Energy's plant at Bowerman landfill, and also, Linde have experience from one commercial facility, at Albury landfill. The numbers presented in Figure 26 is only for the upgrading and/or liquefaction. To these numbers will the energy consumption for the raw gas production be added.

Using cryogenic upgrading technology results in very low CH₄ losses. However, *none*, as Prometheus-Energy and Acirion express it, is not realistic. Small amounts will absorb in the liquid CO₂ and if the origin is landfill gas there is CH₄ losses connected to the flashing of N₂. Another source of CH₄ losses are the membranes used in Acirion process. The vent from these contains significant amounts of CH₄ so it is very important how it is treated. Loss of CH₄ is loss of income but, above all, it result in emissions of a greenhouse gas, 20 times stronger than CO₂. However, they have small influence on the energy balance.

An aspect that has big influence on the energy balance is the use of rejected low value heat in external processes. There are three identified suppliers that have developed technology to take care of this energy. The Coaab process uses significant amounts of heat and this technology would probably not be as interesting as if there were an available user of this. A possible user could be the biogas production plant but this require that the plant is situated close by, which is not always the reality. The dispose of low value waste heat in external processes is therefore very site specific.

An alternative to produce LBG is to inject upgraded biogas into the gas grid and then liquefy a small part at a pressure letdown station. Beyond the conventional upgrading technologies, both SGtS's module 1-3 and Acirion's CO₂ Wash[®] followed by membranes could be used to produce a biogas with 97 % CH₄. The energy balance shows that LBG production at pressure letdown stations is an energy efficient way to produce LBG. The only energy that needs to be added to produce LBG from upgraded biogas is the compression work from 4 to 80 bar.

Cryogenic technology is suitable for upgrading of landfill gas since N₂, which is difficult to separate with the conventional technologies, can be separated in the liquefaction step. However, significant amounts of N₂ will dissolve in the liquid CH₄. This N₂ will be removed through flashing but the flashing also makes some CH₄ to leave. Depending on the N₂ concentration in the raw gas, varying amounts of flashing is needed. A high N₂ concentration results in a lower CH₄ recovery and a higher CH₄ loss, but also in larger energy consumption since more compression work is needed. It is important how the vent from this flashing step is treated. On a landfill the access to cheap gas usually is good so the most likely use of the vent is as fuel in a gas engine.

When LBG is distributed to fuel station by a semi-trailer it is more than 5 times space efficient, compared to CBG distributed on a mobile CBG storage. Also, it is 5-10 times more energy efficient to raise the pressure by pumping (on a multi-purpose station) compared to compression. However, the LBG must be distributed 500-2,100 km, depending on upgrading process, before the higher energy consumption to produce LBG is paid back.

When using cryogenic technology to separate CO₂ it results in a very clean LCO₂ product that could be used in external processes. The disposal of this CO₂ is however very site specific. The possibility to sell it to AGA or Air Liquide (or other suppliers of industrial gases) is probably small. The scale of the biogas production plant is probably too small and too faraway to be of economic interest for them and they cannot use the same distribution vehicle for both

food grade and non-food grade gas quality. To be able to sell the separated CO₂ the LBG producers probably has to invest in an own distribution vehicle and a LCO₂ user has to be situated close by. Since the possible user will be dependent on the biogas production plant the use of this more insecure source must be connected with advantages, such as lower costs and easy access. The biogas producer on the other hand must be able to supply LCO₂ in situations of production stop and maintenance. They would probably need a back-up of LCO₂, which they have to buy from one of the market leading suppliers, to a significant cost.

In this report two possible applications for the use of LCO₂ from cryogenic upgrading have been identified and these are cryogenic temperature control and cultivation of plants and vegetables in greenhouses. The analysis shows that, if LCO₂ is included in the energy balance, the net primary energy consumption for cryogenic upgrading technology can be lowered with as much as 50 %, depending on the CO₂ concentration in the raw gas and the CO₂ recovery in the process. However, there is a surplus of LCO₂ from the LBG production plants for the presented applications. There are however many more fields for LCO₂ use that probably could constitute a possible buyer of LCO₂ from a biogas upgrading plant.

All analysis in this report is based on a number of assumptions, which affects the results in different ways. Most of the data also comes from the supplier of each process, which makes the analysis even more insecure. However, the analysis shows the magnitude of each interesting aspect and this is enough when comparing different technologies with each other. To be able to do the comparisons, all data used in the energy balances have been converted to primary energy. Behind these conversions lies an assumption on how the energy carrier is produced or what the energy carrier would replace in an external application. This assumption affects the result in different ways, which is examined with a sensitivity analysis. Using primary energy factors is useful when comparing different technologies within a study, but it is not that practicable when comparing different studies to each other, since different assumption on energy factors can lead to big differences in the results. For the interested, all original numbers and assumptions can be found in Appendix 3: Original data, data conversion and calculations.

In this report no consideration of costs for different technologies has been taken. This becomes extra clear when analysing the distribution of LBG. LBG must be transported at least 500 km before the inserted energy is paid back. However, it is more economically efficient to distribute biogas as LBG, since it takes more than 5 CBG distribution vehicles per each LBG. In a country like Sweden, where the gas network is limited and the interest of using biogas as a vehicle fuel is growing, producing LBG is a good alternative to CBG. The production is more energy intensive, but it probably results in a more valuable product since it becomes available for a bigger market. The use of cryogenic technology also opens for more smart solutions were LCO₂ and rejected heat could replace fossil fuel sources in external applications, significantly affecting the energy balance in a positive way. LBG also makes biogas available for heavier vehicle, since it can be stored in its liquid state and thereby significantly increasing the driving range.

In Table 15, positive and negative aspects with each process identified in the work with this report are summarized.

Table 15 Identified positive and negative aspects with each process.

Company/process:	Positive:	Negative:
SGtS	Low CH ₄ losses	Energy intensive compared to upgrading of biogas with conventional technology
	Extraction of LCO ₂	Pilot plant only recently available
	Appropriate for landfill gas	No commercial experience
	Take care of waste heat for use in external process	
Acrion	Low CH ₄ losses	Energy intensive process
	Extraction of LCO ₂	No commercial experience
	Experience from pilot plant	
	Appropriate for landfill gas	
Prometheus-Energy	Low CH ₄ losses	Energy intensive
	Experience from commercial and pilot plants	No extraction of LCO ₂
	Appropriate for landfill gas	
Water scrubbing	Experience from commercial applications	High water consumption
		Relatively high CH ₄ losses
Amine	Low CH ₄ losses	Use of chemicals
	Experience from commercial applications	Large use of heat
PSA	Experience from commercial applications	Relatively high CH ₄ losses
Membrane	Experience from commercial applications	High CH ₄ losses in the membranes
N ₂ Brayton cycle	Simple and sturdy	Low efficiency
		No commercial experience
Mixed refrigerant cycle	High efficiency	Complex for small-scale applications
	CO ₂ polishing step included	No commercial experience
	Experience from pilot plant	

In the following points the main conclusions will be summarized:

- It takes around 0.8-1.8 kWh electricity (and heat)/Nm³ clean biogas to produce LBG.
- Inserted primary energy to produce LBG is between 12-23 % of the energy content in the product. If the energy consumption instead is expressed in electricity (and heat), it correspond to 8-18 % of the energy content in the product.
- The most energy efficient ways to produce LBG are to use one of the conventional upgrading technologies; water scrubber, PSA or Coaab (including heat recovery), connected with a mixed refrigerant process or to use SGtS's process.
- Loss of CH₄ has little influence on the energy balance.
- Disposal of low value waste heat has a big impact on the energy balance but the possibility is very site specific.
- Producing LBG at a pressure letdown station is energy efficient in comparison to cryogenic technology and conventional technology connected with a small-scale liquefaction plant. However, as much as 7-9 % of propane must be added to the biogas, which is an extra cost and an input of a non-renewable energy source.
- The results on primary energy consumption are very dependent on choice of primary energy factors. However, it is very useful to use primary energy factors when different energy carriers are compared to each other, as in this report.
- Cryogenic technology is suitable for upgrading of landfill gas since N₂ can be separated in the CH₄ liquefaction step. To keep CH₄ losses low, it is very important how the vent from the flashing system is treated.
- It is more than 5 times space efficient to deliver LBG in a vacuum insulated vessel compared to CBG (200 bar) delivery on a mobile CBG storage.
- LBG has to be transported 500-2,100 km before the extra inserted energy to produce LBG is paid back, due to more efficient delivery, compared to a CBG system.
- The possibility to sell LCO₂ to one of the industrial gas producers is probably small. An alternative is to sell directly to a buyer, but the markets for cryogenic transport temperature control and cultivation of plants and vegetables in greenhouses are probably too small.

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Appendix 1: Cryogenic upgrading technology

In the text below follows more detailed descriptions over cryogenic upgrading technologies identified in this report.

Scandinavian GtS

A detailed sketch over SGtS process, divided into module 1-3, can be seen in Figure A1 1. The first design for module 4 is finished but since SGtS patent application is pending they are prohibited to reveal the technical details of the design. (Kättström, 2008)

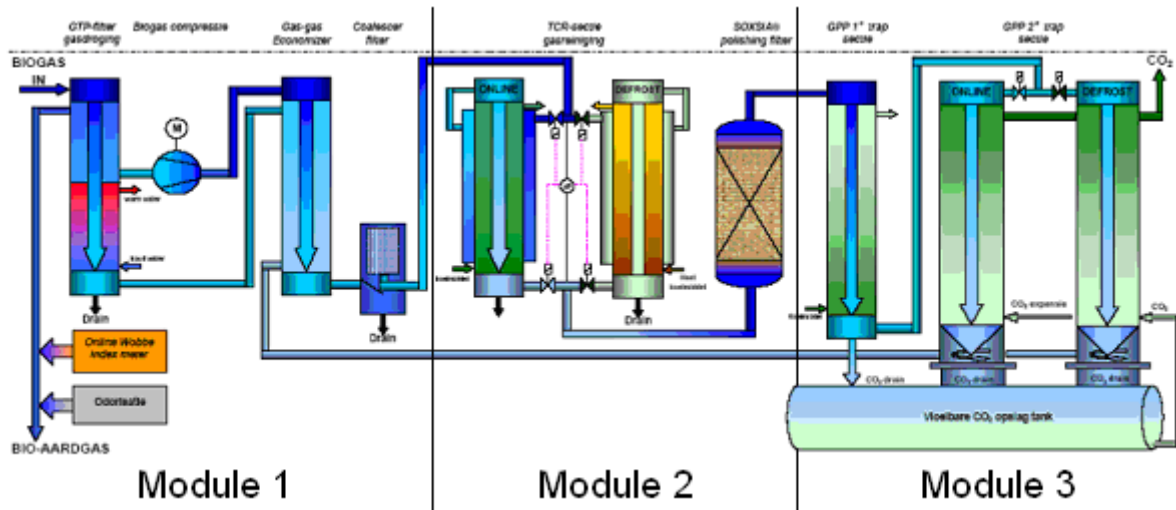


Figure A1 1 A detailed sketch over module 1-3 (Scandinavian GtS, 2008b)

Module 1

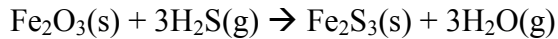
Module 1 consists of a GTP (Gas Treatment Package)-filter followed by biogas compression, a gas-gas economizer and a coalescer filter. In the GTP-filter the gas is cooled to +6 °C by the outgoing gas flow and cold water. This makes the moisture condense, together with most contaminants. Many remaining contaminants dissolve in the condensed moisture and everything is drained. The gas is then compressed to around 25 – 35 bar, depending on the optimum pressure for the refrigerating machines to reach highest efficiency. After compression the gas is further cooled in a heat exchanger, the *gas-gas economizer*, using the clean and cold (-60 - -80 °C) outgoing biogas flow as a refrigerant. (When module 4 eventually will be joined to the process the outgoing cold gas flow no longer will be used as a refrigerant in the gas-gas economizer.) Finally the gas flow is led through a coalescer filter to deposit more water. (Kättström, 2008)

Module 2

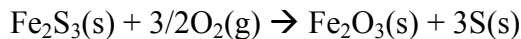
Module 2 consists of a TCR (Total Contaminant Removal)-system connected with a SOXSIA[®]-filter. The main task for the TCR-system is to clean the gas from hydrogen sulphide, H₂S, and siloxanes. The gas is chilled to approximately -25 °C, depending on working pressure, to freeze out the remaining water and to condensate siloxanes. (Kättström, 2008) In this process deposits are formed on the walls so the TCR-unit switches between two identical heat exchangers. While one is *online*, the other one is defrosted by a warm refrigerant and, before the unit is getting online again, the remaining water and siloxanes are drained. (Scandinavian GtS, 2008)

After the TCR-system the dry gas enters a SOXSIA[®]-filter. This filter is used to remove remaining traces of H₂S and siloxanes. SOXSIA[®] stands for *Sulphur Oxidation and Siloxane Adsorption* and is a catalyst that adsorbs siloxanes and at the same time converts H₂S to elementary sulphur, S. It is designed to hold 40 % more siloxanes per unit of volume and it can operate at low temperatures, atmospheric pressure and relatively high feed gas flows.

H₂S is removed from the gas flow through a chemical reaction with iron oxide, Fe₂O₃:



To regenerate Fe₂O₃ the absorber is purged with inert gas at 20 °C to 50 °C. Then a small amount of oxygen is added, up to 0.2 %, and the following chemical reaction takes place:



SOXSIA[®] can hold up to 26 % of its own weight on elementary sulphur and after around six month the filter need to be switched. The used filter is desulphured by heating it to approximately 450 °C. This makes the elementary sulphur melt and after desulphurisation the SOXSIA[®]-filter is ready to be reused. (Scandinavian GtS, 2008)

Module 3

Module 3 consists of a GPP[®] (Gastreatment Power Package)-system that removes CO₂ from the clean and dry gas. The gas is first chilled to -78 °C, which makes the CO₂ condense. After that the gas is led into a unit consisting of two identical heat exchangers, like the TCR-system, but with a different refrigerant. The *online* part freezes out the CO₂, with a very good separation as a result, while the *defrost* part is defrosted producing clean liquid CO₂, LCO₂. This LCO₂ is stored in a storage tank. A part of it is used as a refrigerant in the process while the rest could be used as a valuable by-product. (Kättström, 2008)

Gas leaving module 3 is dry and clean and could, after compression, be used as CBG, or further processed to produce LBG.

Module 4

As mentioned earlier the design of module 4 is finished and since the patent application is pending SGtS is not able to reveal the technical characteristics of the design. The purpose with module 4 is to liquefy CH₄ by chilling the gas flow to around -190 °C. CH₄ will be separated from any nitrogen, N₂, which has a lower condensation temperature, and the product is LBG with a methane content over 99 %. (Kättström, 2008)

Acrion/Terracastus Technologies

CO₂ Wash[®]

CO₂ Wash[®] is a distillation column which efficiently removes contaminants such as siloxanes, halogenated compounds and non-methane organic compounds from the raw gas. A sketch over the column can be seen in Figure A1 2. The compressed, desulphured and dried gas is fed into the bottom of the column. As the gas drift upward CO₂ condense due to refrigeration at the top. Most of this liquid CO₂ exits at the top with a purity of 99.99 %. (Acrion, 2008a) A small portion is however washed back down the column. This reversed stream of LCO₂ absorbs the contaminants in the upward gas stream and is then taken out in the bottom. It is vaporized and finally incinerated in the landfill flare or oxidized in a regenerative thermal oxidizer. At the top a clean gas stream containing CH₄, CO₂, O₂ and N₂, with a CO₂ content of approximately 25 %, exits the column. (Elmqvist, 2008)

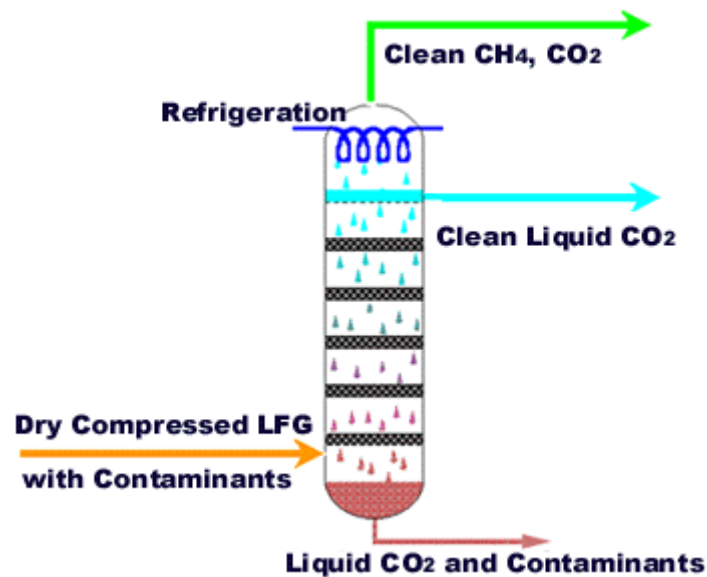


Figure A1 2 Sketch over CO₂ Wash[®]. (Acrion, 2008a)

LBG process

Before entering the CO₂ Wash[®] the gas is compressed, desulphured and dried. Since H₂S is a very volatile molecule it is hard to reduce it to desired levels with CO₂ Wash[®]. It is instead removed with a *Sulfa Treat* upstream the process, where H₂S reacts with iron oxide in a non-recoverable process. (Brown, 2008) A sketch over the total process can be seen in Figure A1 3.

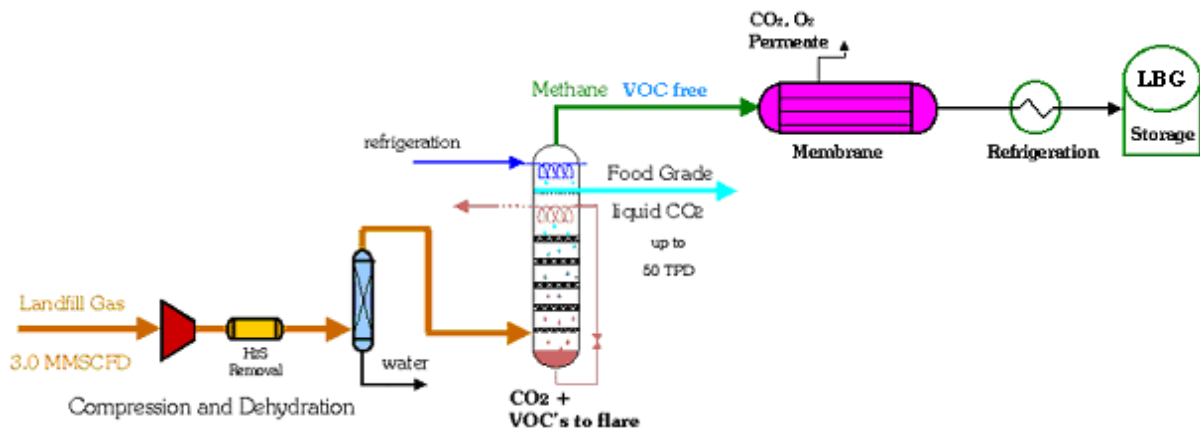


Figure A1 3 Process flow diagram over Acirion’s process for the production of LBG from raw gas. The CO₂ polishing step is not included in the figure. (Acirion, 2008a & b)

To prevent dry ice formation in the liquefaction step the clean gas stream from CO₂ Wash[®] needs to be further processed. CO₂ and any existing O₂ are separated from the gas stream with two MEDAL membranes. The process is not designed to recover CO₂ from these membranes. However, depending on the economical value of LCO₂, it is possible to process the outgoing CO₂ to get clean LCO₂ from them as well. Gas leaving the membranes has a CO₂ content of around 1-2 %. (Brown, 2008)

Before entering the refrigerating process the last CO₂ is removed with a *mol sieve* and the CO₂ concentration is reduced to 100 ppmv. Any existing N₂ is separated and flashed in the refrigeration plant. The methane is then liquefied in a liquefaction plant using a Brayton N₂ cycle or a mixed refrigerant cycle. (Brown, 2008)

Analysis results – Burlington County landfill

Purification results from the pilot project at Burlington County landfill can be found in Table A1 1 and Table A1 2. The results were continuously observed and analysed by an independent third party; Atlantic Analytical Laboratory. (Acirion, 2008b)

Table A1 1 Contaminant concentrations in the clean gas stream leaving the CO₂ Wash[®] column. (Acirion, 2008b)

Compound:	Concentration:
Siloxane Compounds	below detection limit of 5 ppb
Chlorinated Hydrocarbons	below detection limit of 10 ppb
Total Sulphur Compounds	100 ppb

Table A1 2 Contaminant concentrations in the liquid carbon dioxide product, LCO₂, from CO₂ Wash[®]. (Acirion, 2008b)

Compound:	Concentration:
Propane	67 ppm
COS	0.25 ppm
Benzene	below detection limit of 5 ppb
H ₂ S and Other Sulphur Compounds	below detection limit of 50 ppb
Light hydrocarbons (C ₁ -C ₃)	below detection limit of 1 ppm
Total chlorinated hydrocarbons	below detection limit of 500 ppb
Hydrocarbons (BTX, Alcohols, C ₅ +)	below detection limit of 200 – 500 ppb
Inert components (N ₂ , CO, O ₂ , H ₂)	below detection limit of 4 ppm

Prometheus-Energy

Prometheus-Energy's process is built up in modules. In the pre-purification module the pressure is raised and water, sulphur compounds and low concentrations of non-methane-organic compounds including siloxanes are removed. The following bulk purification module removes CO₂ from the gas stream by using a proprietary cryogenic freezing technique that freeze out the CO₂ and at the same time pre-cools the methane and any nitrogen. In the liquefaction step the dry and clean gas is liquefied. In the next step, the post-purification step, the CH₄-concentration is enhanced through dynamic flash evaporation of the N₂ and the N₂ concentration is reduced to less than 3 %. The refrigerant module provides the cooling to the process through a closed Brayton N₂ cycle. By using a separate refrigerant instead of LCO₂ or/and N₂ from the process stream, the refrigerating module does not respond to flow variations. To increase the thermodynamic efficiency of the overall process the refrigerant cycle is designed to maximize the pre-cooling of the gas stream. (Prometheus-Energy, 2008c)

Prometheus-Energy does not want to reveal any technical details over their present technology. However, there is an available process flow diagram over the pilot project at Hartland Landfill. Prometheus-Energy was then known as CryoFuel Systems. The process can be seen in Figure A1 4. LFG entering the process is compressed and chilled in the *chiller* for removal of water and heavy organic compounds. These are vaporized and burned in the flare. The remaining contaminants are then removed in the TSA columns before the gas enters the *cold box*. In the cold box CO₂ is separated from the gas stream through condensation in the heat exchangers and sent to LCO₂ storage. Then the CH₄ is condensed and the cold for the condensation of CO₂ and CH₄ is produced in a closed Brayton cycle. Any N₂ is then separated from the LBG in the blue/green columns. The N₂ concentration in the raw gas at Hartland Landfill varied between 14-38 %, which is a very high N₂ concentration, resulting in a high CH₄ concentration in the flash vent. This vent is sent to the gas engine driving the process. (Pettersson et al, 2006)

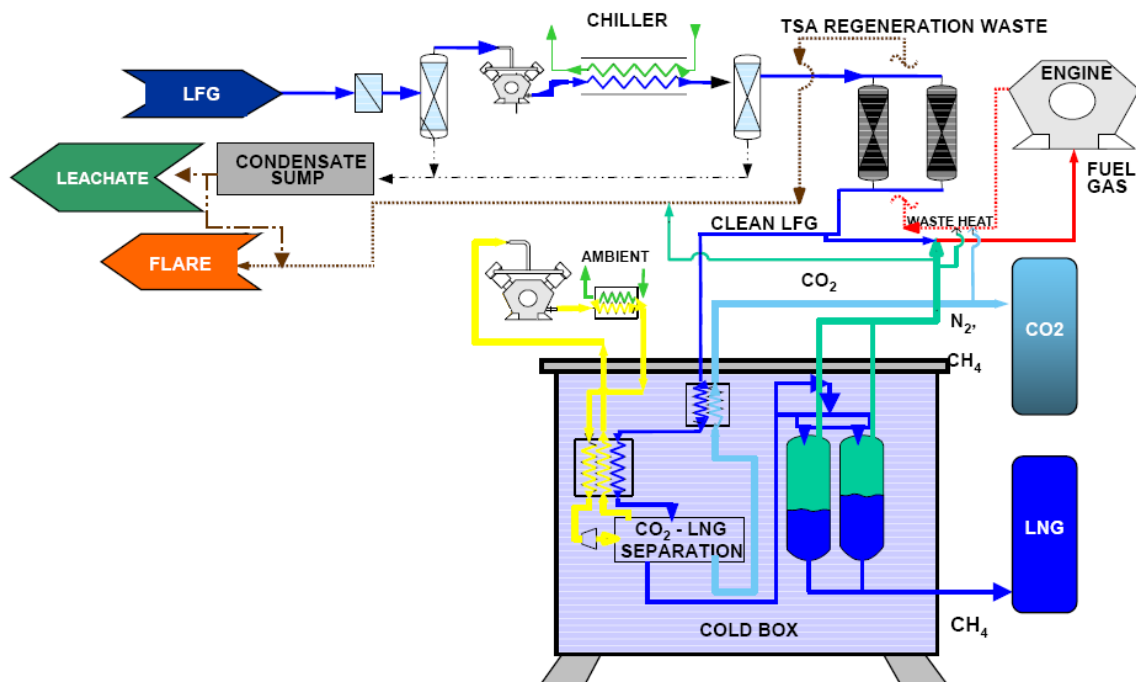


Figure A1 4 Process flow diagram over the pilot at Hartland Landfill. (Pettersson et al, 2006)

Appendix 2: Air Liquide Advanced Technology

Air Liquide Advanced Technology recently presented a process for the production of LBG from landfill gas, using MEDAL membranes for the separation of CO₂.

Figure A2 1 shows an example of a system process flow diagram for the purification and upgrading of landfill gas. Raw gas from the landfill is first compressed and dried before H₂S is removed. The gas is further dehydrated in the coalescer filter before the last water, siloxanes and halogen compounds are removed in a PSA column. The skid is equipped with two PSA columns; one is in operation while the other one is regenerated. For the removal of hydrocarbons two activated carbon beds, in series, are used. The significant amount is removed in the first bed while the second is used for a finishing polish. Now the gas is free from impurities and CH₄ and CO₂ is separated in a two-stage membrane. (Rouaud, 2008)

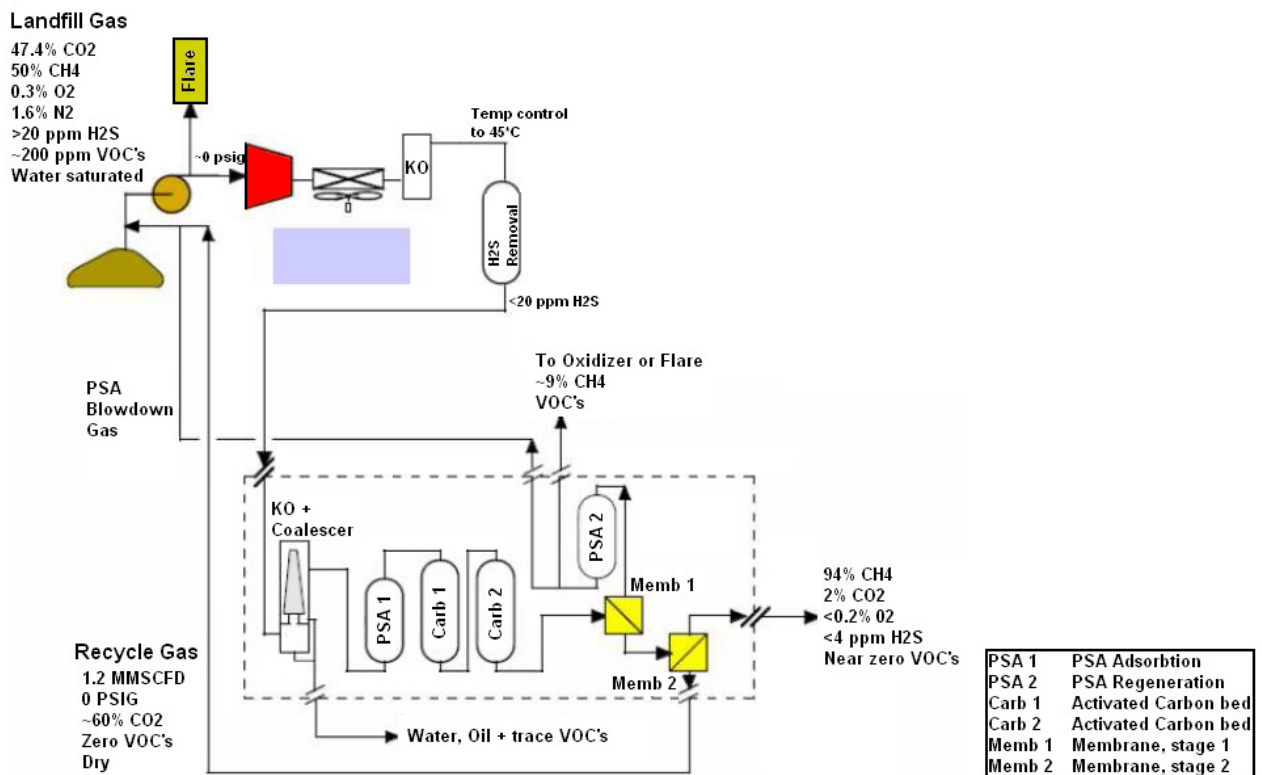


Figure A2 1 A system process flow diagram over the purification and upgrading of landfill gas (Rouaud, 2008).

These membranes are called MEDAL and Figure A2 2 shows a principle sketch over a carbon dioxide membrane. (Acirion also uses these membranes in their process, after the CO₂ Wash[®], see chapter 4.1.2 and Appendix 1) It is made up of a polymeric hollow fibre and the driving force is the differences in partial pressure for CO₂ and CH₄ across the membrane. Pressurised feed gas enters the membrane to the left in the figure. CO₂ is the *fast* gas and permeate through the membrane to the low-pressure side where it is collected while CH₄ stays under pressure. With a two-stage system, as in the system process flow diagram in Figure A2 1, the MEDAL membranes reduce the CO₂ content to below 2 %, with a 90 %+ CH₄ recovery. (Rouaud, 2008)

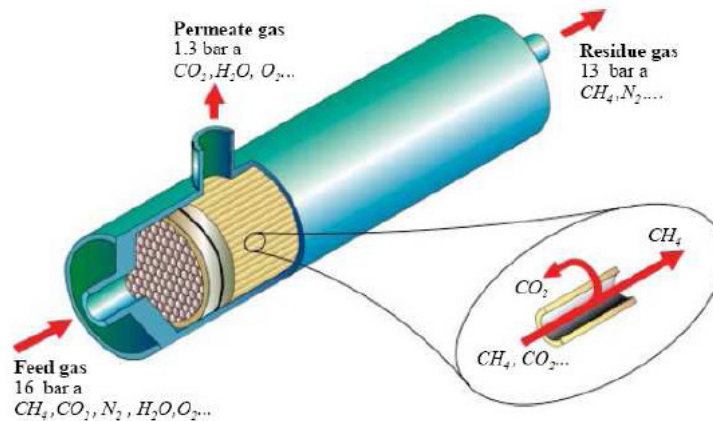


Figure A2 2 Principle sketch over a MEDAL Membrane from Air Liquide (Rouaud, 2008).

Before the liquefaction step the last CO_2 is removed with a proprietary PSA at ambient temperature and medium pressure. If the gas contains any N_2 this will be removed with a cryogenic heat exchanger placed after the membranes. (Rouaud, 2008)

For the liquefaction Air Liquide use a plate-fin heat exchanger where the cold power is provided through a closed nitrogen Brayton cycle, see Figure A2 3. N_2 is compressed and cooled (cooling water) to room temperature. Before entering the turbo expander, where the cold power is produced, it is pre-cooled in the plate-fin heat exchanger by the outgoing N_2 . Incoming CH_4 is then liquefied in the heat exchanger. (Rouaud, 2008)

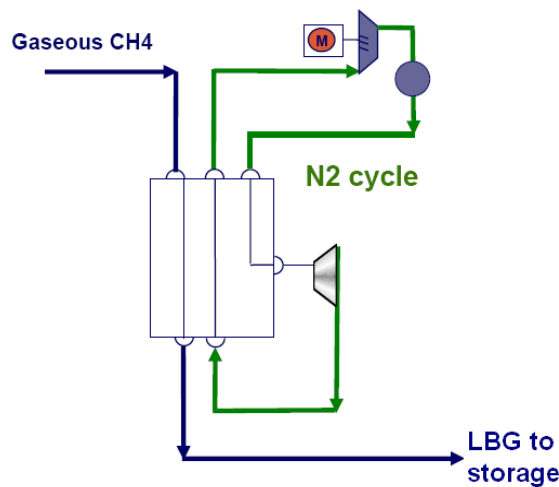


Figure A2 3 Liquefaction unit based on a N_2 Brayton cycle (Rouaud, 2008).

Air Liquide Advanced Technology is also developing an alternative liquefier based on a N_2 Brayton cycle for the combined separation of N_2 and CH_4 and production of LBG. (Rouaud, 2008)

Appendix 3: Original data, data conversion and calculations

Table A3 1 Reported original data regarding energy consumption and scales from suppliers of different technologies.

Company:	Specification:	CH ₄ concentration (%)	Electricity:	Heat:	Unit:	Scale:	Unit:	Source:	
		Raw gas:	Product:						
Cryogenic Technology:									
Scandinavian GtS	Raw gas to clean biogas (4-6 bar)	50	100	0.25	-	kWh/Nm ³ raw gas	50-2,400	Nm ³ /h	(Kättström, 2008)
	Clean biogas (CH ₄ + N ₂) to LBG	-	100	0.15	-	kWh/Nm ³ clean gas	-	-	(Kättström, 2008)
	Raw gas to LBG	50	100	0.4	-	kWh/Nm ³ raw gas	-	-	(Kättström, 2008)
Acron	Raw gas to LBG	-	100	0.15	-	kWh/kWh LNG	2,500, 5,000, 10,000	gallons LBG/day	(Elmqvist, 2008)
Prometheus-Energy	Raw gas to LBG	-	100	3	-	kWh/gallon LNG	-	-	(Brown, 2008)
	-	-	-	-	-	-	1,000-10,000	gallons LBG/day	(Montague, 2008)
Conventional Technology:									
Malmberg Water	Raw gas to upgraded biogas (6-7 bar)	60	97	0.24	-	kWh/Nm ³ raw gas	no data	no data	(Malmberg, 2008)
Flotech	Raw gas to upgraded biogas (9 barg)	60	97	0.215-0.27**	-	kWh/Nm ³ raw gas	no data	no data	(Flotech, 2008)
Läckeby Water	Raw gas to upgraded biogas (4 barg)	65	99	0.12	-	0.13 kWh/Nm ³ raw gas (0.62***)	no data	no data	(Karlsson, 2008)
CarboTech	Raw gas to upgraded biogas	60	97	0.24	-	kWh/Nm ³ raw gas	no data	no data	(Schulte, 2008)
Liquefaction Technology:									
Cryostar	Clean biogas to LBG	-	100	0.625****	-	kWh/Nm ³ clean gas	>5	ton LNG/day	(Rivollier, 2008)
Hamworthy	Clean biogas to LBG	-	100	0.6	-	kWh/kg LNG	5-50	ton LNG/day	(Jacobsen, 2008)
Conventional Technology + Liquefaction:									
The Linde Group	Raw gas to LBG	-	100	2	no data	kWh/kg LNG	no data	no data	(Carson, 2008)

* Assumed CH₄ concentration in the product, supposed that the biogas origin is digester gas

** Energy consumption varies between 0.215-0.27 depending on the scale; the larger scale the smaller energy demand.

*** 0.62 is the gross heat consumption, but of these 0.49 kWh can be reused for heating in external processes. This results in a net consumption of 0.13 kWh.

**** The energy consumption is calculated from simulations on a real case.

Table A3 2 Original data converted to kWh/Nm³ clean biogas and to primary energy in kWh/Nm³ clean biogas.

Company:	Specification:	CH ₄ concentration (%)		Original data		Energy consumption ^{**} (kWh/Nm ³ clean biogas)		Primary energy consumption ^{***} (kWh/Nm ³ clean biogas)		
		Raw gas:	Product: [*]	Electricity:	Heat:	Unit:	Electricity:	Heat:	Electricity:	Heat:
Cryogenic Technology:										
Scandinavian GtS	Raw gas to clean biogas (4-6 bar)	50	100	0.25	-	kWh/Nm ³ raw gas	0.50	-	0.80	-
	Clean biogas (CH ₄ + N ₂) to LBG	-	100	0.15	-	kWh/Nm ³ clean gas	0.15	-	0.24	-
Accion	Raw gas to LBG	50	100	0.4	-	kWh/Nm ³ raw gas	0.80	-	1.28	-
	Raw gas to LBG	-	100	0.15	-	kWh/kWh LNG	1.50	-	2.39	-
	Raw gas to LBG	-	100	3	-	kWh/gallon LNG	1.34	-	2.15	-
Conventional Technology:										
Malmberg Water	Raw gas to upgraded biogas (6-7 bar)	60	97	0.24	-	kWh/Nm ³ raw gas	0.40	-	0.64	-
	Raw gas to upgraded biogas (9 barg)	60	97	0.215-0.27*	-	kWh/Nm ³ raw gas	0.36-0.45	-	0.57-0.72	-
Läckeby Water	Raw gas to upgraded biogas (4 barg)	65	99	0.12	0.13	kWh/Nm ³ raw gas	0.18	0.20	0.30	0.23
					(0.62**)			(0.95)		(1.11)
CarboTech	Raw gas to upgraded biogas	60	99	0.24	-	kWh/Nm ³ raw gas	0.40	-	0.64	-
Liquefaction Technology:										
Cryostar	Clean biogas to LBG	-	100	0.625***	-	kWh/Nm ³ clean gas	0.63	-	1.01	-
	Clean biogas to LBG	-	100	0.6	-	kWh/kg LNG	0.43	-	0.69	-
Conventional Technology + Liquefaction:										
The Linde Group	Raw gas to LBG	-	100	2	-	kWh/kg LNG	1.43	-	2.29	-

* Assumed CH₄ concentration in the product, supposed that the biogas origin is digester gas

** Original data converted to one unit using conversion data and assumptions described in Appendix 3: Original data, data conversion and calculations

*** Energy consumption converted to primary energy consumption using primary energy factors for Nordic electricity mix (1.6) and for forest fuel (1.16)

Assumptions and conversion data

All numbers on energy consumption are presented in one unit: kWh/Nm³ clean gas. If the original numbers are specified in another unit it is converted to this unit. If the original numbers are presented per raw gas, and the CH₄ content in the gas varies in a wide range or is not known, it is assumed that the raw gas consists of 60 % CH₄. The lower CH₄ content in the raw gas, the higher energy demand per unit clean gas, since CO₂ and N₂ does not contain any energy. A CH₄ concentration of 50 % is relevant for landfill gas but it is very low for digester gas.

LNG contains significant amounts of higher hydrocarbons, which results in a higher heating value, compared to LBG. However, in this report, if a value is specified in LNG, it is assumed that it consists of 100 % CH₄ (if nothing else is said). This is a relevant assumption for the presented upgrading technologies since they work in the biogas field and since biogas from landfills and digester plants do not contain any higher hydrocarbons. However, LBG can contain up to 3 % N₂, especially if the source is landfill gas, and this results in a slightly lower heating value. Table A3 3 shows data for CH₄ for different characteristics, which are used in the conversion of original numbers.

Table A3 3 CH₄ data for different characteristics.

CH ₄ characteristics:	Specifications:	Value:	Unit:	Source:
Boiling point	1.013 bar	-161.6	°C	Air Liquide
Density	0°C, 1.013 bar	0.717	kg/Nm ³	SGC
	LBG, 1.013 bar	422.62	kg/m ³	Air Liquide
Lower heating value	-	9.97	kWh/Nm ³	SGC

$$1 \text{ kg LBG} = 1/0.717 \text{ Nm}^3 \approx 1.395 \text{ Nm}^3 = 1.395 * 9.97 \text{ kWh} \approx 13.9 \text{ kWh}$$

$$1 \text{ kWh} = 1/9.97 \text{ Nm}^3 \approx 0.1 \text{ Nm}^3$$

$$1 \text{ L LNG (1.013 bar)} = 1 * 10^{-3} \text{ m}^3 = 1 * 10^{-3} * 422.62 \text{ kg} \approx 0.42262 \text{ kg} = 0.42262 * 1.395 \text{ Nm}^3 \approx 0.5896 \text{ Nm}^3 \text{ CH}_4$$

Units:

$$\begin{aligned} 1 \text{ U.S. gallon (liquid)} &= 3.7854 * 10^{-3} \text{ m}^3 \\ 1 \text{ ft}^3 \text{ (cubic feet)} &= 0.0283 \text{ m}^3 \\ 1 \text{ HP (horse power)} &= 0.7457 \text{ kW} \\ 1 \text{ meter/min} &= 0.43 \text{ gallon/min} \end{aligned}$$

$$\begin{aligned} 1 \text{ meter/min} &= 0.43 \text{ gallon/min} = 0.43 * 3.7854 * 10^{-3} \text{ m}^3/\text{min} \approx 0.001628 \text{ m}^3/\text{min} = \\ &0.001628 * 422.62 \text{ kg/min} \approx 0.6879 \text{ kg/min} = 0.6879 * 1.395 \text{ Nm}^3/\text{min} \approx 0.9596 \text{ Nm}^3/\text{min} \end{aligned}$$

$$1 \text{ U.S. gallon} = 3.7854 * 10^{-3} \text{ m}^3 = 3.7854 * 10^{-3} * 422.62 \text{ kg} \approx 1.5998 \text{ kg} = 1.5998 * 1.395 \text{ Nm}^3 \approx 2.2317 \text{ Nm}^3$$

Conversion data:

$$\begin{aligned} 1 \text{ kg LBG} &= 1.39 \text{ Nm}^3 \\ 1 \text{ kg LBG} &= 13.9 \text{ kWh} \\ 1 \text{ kWh} &= 0.10 \text{ Nm}^3 \\ 1 \text{ U.S. gallon} &= 2.23 \text{ Nm}^3 \end{aligned}$$

Cryogenic technology

Table A3 4 Information regarding different CH₄ and CO₂ parameters for the three identified cryogenic upgrading technologies.

Company:	CH ₄ content in LBG (%)*:	CH ₄ recovery (%):	CH ₄ loss (%):	CO ₂ recovery (%):	LCO ₂ purity (%):	Source:
Scandinavian GtS	99	>99	0.5	~100	“food grade“	(Kättström, 2008)
Acirion	99.2**	>99	”None”	30 – 50***	99.99	(Brown, 2008)
Prometheus-Energy	>97	no data	”None”	-	-	Barclay, 2008)

* In this table the CH₄ concentration in the product is less than 100 %. However if the biogas source is digester gas it would be very close to 100 %.

** CH₄ content in the gas produced during the pilot project at Burlington County Landfill

*** Depends on economics; if the product can be sold or not.

Table A3 5 Scales and electricity consumption for production of LBG from raw gas. The specified scales for the technology from Acirion and Prometheus-Energy are the scales on actual available designs. The data have been converted from original data in Table A3 1.

Company:	Electricity: (kWh/Nm ³ clean gas)	Drive:	Scales: (Nm ³ raw gas /h)	Source:
Scandinavian GtS	0.8	Electricity	50 – 2400	(Kättström, 2008)
Acirion	1.42*	Electricity	230, 465, 930	(Brown, 2008)
Prometheus-Energy**	1.54	LFG fired gas engine	90 – 930	Barclay, 2008)

* Mean value from two different sources.

** Based on actual numbers for the production of LBG at the Bowerman landfill plant.

Prometheus-Energy – Bowerman Landfill

The numbers on energy consumption from Prometheus-Energy is actual numbers for their facility at Bowerman Landfill. The plant is driven with a LFG fired engine, which requires approximately 800 – 1,000 kW per hour to operate at full capacity. The efficiency off the engine is 30 – 35 %. Each day 1.15 MMscf (million standard cubic feet) LFG is taken out from the landfill to power the facility and to produce 5,000 gallons of LBG. The composition of the landfill gas is approximately 50 % CH₄, 40 % CO₂ and 10 % N₂. (Montague, 2008)

Scf stands for “standard cubic foot” which is the volume at 60 F and 1 atm.

$$60\text{F} = 15.56\text{ }^{\circ}\text{C} = 288.71\text{ K}$$

$$1\text{ ft}^3 = 0.02832\text{ m}^3$$

What does 1 scf correspond to in Nm³?

Assumption: ideal gas

$$\rightarrow 1\text{ scf} = 26.79 \cdot 10^{-3}\text{ Nm}^3$$

$$\rightarrow 1.15\text{ MMscf} = 1.15 \cdot 10^6 \cdot 26.79 \cdot 10^{-3}\text{ Nm}^3 = \underline{30,812.75\text{ Nm}^3\text{ landfill gas}}$$

$$1\text{ U.S. gallon (liquid)} = 2.2317\text{ Nm}^3$$

This conversion factor is for 100 % CH₄ in the product. The product from Bowerman landfill contains 3 % N₂ but, since this calculation only will determine the magnitude of the energy consumption, it is assumed that the product contains 100 % CH₄.

$$\rightarrow 5,000\text{ gallons} = 5,000 \cdot 2.2317\text{ Nm}^3 = 11,158.5\text{ Nm}^3\text{ CH}_4$$

Which LFG volume does this correspond to?

The composition of the gases is specified in volume fractions, which correspond to mole fractions.

$$LFG : \frac{5}{10} CH_4, \frac{4}{10} CO_2, \frac{1}{10} N_2$$

$$LFG : \frac{97}{100} CH_4, \frac{3}{100} N_2$$

How many moles CO₂ and N₂ should be added to the LBG to correspond to the LFG composition?

$$LFG : 97 \text{ mole } CH_4, 3 + X \text{ mole } N_2, Y \text{ mole } N_2$$

$$\begin{cases} \frac{97}{97 + 3 + X + Y} = \frac{5}{10} \\ \frac{3 + X}{97 + 3 + X + Y} = \frac{1}{10} \end{cases}$$

$$\rightarrow X = 16.4$$

$$\rightarrow Y = 77.6$$

16.4 + 77.6 = 94 moles need to be added to the 100 moles of LBG to correspond to the composition of LFG. One volume fraction of LBG therefore corresponds to the LFG volume of 194/100 = 1.94.

$$\rightarrow 11,158.5 \text{ Nm}^3 \text{ LBG (3\% N}_2) = 1.94 * 11,158.5 \text{ Nm}^3 = \underline{21,647.51 \text{ Nm}^3 \text{ raw LFG}}$$

LFG outtake: 30,812.75 Nm³ LFG/day

LBG production: 21,647.51 Nm³ LFG/day

LFG to engine:

$$30,812.75 - 21,647.51 = 9,165.24 \text{ Nm}^3$$

$$9,165.24/24 = 381.89 \text{ Nm}^3/\text{h}$$

The efficiency of the gas engine varies between 30-35 % and the effect of the engine is between 800-1,000 kW:

$$\text{Min: } 800/381.89 * 0.3 = 0.623 \text{ kWh/Nm}^3 \text{ raw gas}$$

$$\text{Max: } 1,000/381.89 * 0.35 = 0.917 \text{ kWh/Nm}^3 \text{ raw gas}$$

$$\rightarrow \text{Average: } 0.77 \text{ kWh/Nm}^3 \text{ LFG (50 \% CH}_4)$$

$$\rightarrow \underline{1.54 \text{ Nm}^3/\text{Nm}^3 \text{ clean gas (100 \% CH}_4)}$$

Conventional technologies

Table A3 6 Characteristics for the different purification and upgrading technologies, including CH₄ losses.

Company:	Technology:	Regeneration:	Pre treatment:	After Treatment:	CH ₄ losses (%)	Source:
Malmberg Water	Water scrubbing	Air stripping	H ₂ S removal	Drying	1	(Simonsson, 2008)
Flotech	Water scrubbing	Air stripping	H ₂ S removal	Drying	0.1	(Flotech, 2008)
Läckeby Water	Amine (Cooab)	Heating	H ₂ S removal	Drying	< 0.1	(Karlsson, 2008)
CarboTech	PSA	Vacuum	H ₂ S removal, drying	-	1-2	(Schulte, 2008)

Table A3 7 CO₂, H₂S and moisture concentrations after purification and upgrading.

Company:	Technology:	CO ₂ (%)	H ₂ S (ppmv)	Dew point (°C, 4bar)	Source:
Malmberg Water	Water scrubbing	2	5	-80	(Simonsson, 2008)
Flotech	Water scrubbing	2	0.1	-80	(Flotech, 2008)
Läckeby Water	Amine (Cooab)	<25*	< 0.5	-60	(Karlsson, 2008)
CarboTech	PSA	1-3	<1	Not available	(Schulte, 2008)

* If the upgrading unit is complemented with an extra absorption column.

Small-scale liquefaction technology

Table A3 8 Scales and electricity (converted from original data in Table A3 1) demand for the N₂ Brayton cycle from Cryostar and the MRC from Hamworthy.

Company:	Refrigeration process:	Scales: (Nm ³ clean gas/h)	Power investment (kWh/Nm ³ clean gas)
Cryostar	Closed Nitrogen reversed Brayton cycle	From 300	0.63*
Hamworthy	Closed Mixed-Refrigerant cycle	300 – 2,900	0.43

* Calculated from simulations of a real simulation

Cryostar

Original information:

To liquefy a gas flow of 400 Nm³ purified gas (8 bar, 20°C)/h a power investment of 250kW is needed. This power investment is calculated through simulations of a real case. (Rivollier, 2008)

→ 250 kW/400 Nm³/h = 0.625 kWh/Nm³ clean gas

Distribution

Table A3 9 Original numbers on storage capacity for biogas distribution.

Fuel type:	Specification:	Storage capacity:	Source:
LNG	83.7 % filling rate, 1.013 bar	19,730 kg	(Cryo AB, 2008)
CNG	1 mobile CBG storage, 200 bar	1,720 Nm ³	(Schröder, 2008)

1 truck can take 3 mobile CBG storages each trip

→ 3*1,720 = 5,160 Nm³/trip

Fuel station technology

Cryostar and Nexgen Fuels left a proposal over a fuel station design, including pump specifications like numbers on energy consumption. Vanzetti Engineering on the other hand left the names of the pump series to use. In each series there is a number of pumps to choose between and from these, pumps have been chosen to match the request. In Table A3 10 data over the different pumps can be found.

Table A3 10 Data on motor power and flow capacity for reciprocating and submerged centrifugal pumps from different suppliers.

Company:		Reciprocating Pump:	Unit:	Submerged centrifugal pump:	Unit:	Source:
Cryostar	Motor power	22	kW	15	kW	(Cryostar, 2008)
	Max flow capacity	600	Nm ³ /h	360	L/min	
Vanzetti Engineering	Motor power	22	kW	13	kW	Vanzetti Engineering, 2008b)
	Max flow capacity	16.8	L/min	no data	no data	
Nexgen Fueling	Motor power	70	HP	15 – 25	HP	(Emmer, 2008)
	Max flow capacity	10	meters/min	10	m ³ LNG/min	

Each fuel station is supplied with a buffer unit. When calculating the energy demand to power the pumps it is assumed that the pumps work on max capacity to refill the buffer unit and then stop when it is full. The energy demand for each supplier, together with an average, can be found in Table A3 11.

Table A3 11 Energy demand for different pumps and suppliers, together with an average.

Company:	Reciprocating pump (kWh/Nm ³)	Submerged centrifugal pump (kWh/Nm ³)
Cryostar	0.037	0.0018
Vanzetti Engineering	0.037	-
Nexgen Fuels	0.09	0.00316
Average:	0.055	0.0027

Fuel storage capacity

Table A3 12 Storage capacity and pressure for different vehicle types.

Storage and vehicle:	Parameter:	Value:	Unit:	Source:
CBG car	Tank volume (average)	20	Nm ³	(miljöfordon.se, 2008)
	Storage pressure	200	bar	(miljöfordon.se, 2008)
LBG truck*	Tank volume	72	U.S. gallon	(Storrrar, 2008)
CBG truck*	Tank volume	310	dm ³	(Storrrar, 2008)
	Storage pressure	250	Bar	(Storrrar, 2008)

* Vehicles with dual-fuel

CNG trucks:

How many Nm³ gas fits in a 310 litres tank at 200 bar (storage pressure in Sweden)?

Since the pressure is high the compressibility factor, Z, is added to the ideal gas law:

$$pV = nZRT$$

$$nR = \text{constant}$$

$$\rightarrow \frac{p_1 V_1}{Z_1 T_1} = \frac{p_2 V_2}{Z_2 T_2}$$

$$\rightarrow V_2 = \frac{Z_2 T_2 p_1 V_1}{Z_1 T_1 p_2}$$

The storage temperature is assumed to be 15 °C and the gas is assumed to consist of 100 % CH₄. Z is calculated from a calculation program available at SGC's homepage (SGC, 2008).

$$p_1 = 200 \text{ bar}, p_2 = 1,013 \text{ bar}$$

$$T_1 = 15 \text{ °C} = 288.15 \text{ K}, T_2 = 0 \text{ °C} = 273.15 \text{ K}$$

$$Z_1 = 0.7625, Z_2 \approx 1$$

$$V_1 = 310 \text{ dm}^3 = 0.31 \text{ m}^3$$

$$\rightarrow V_2 = 76.09 \text{ Nm}^3$$

Table A3 13 Storage capacities, in Nm³, for different vehicle types (calculated from data in Table A3 12).

Vehicle type* :	Storage capacity (Nm ³):
Car, CBG storage	20
Truck, CBG storage	76
Tuck, LBG storage	160

* The trucks are of the dual-fuel technology

Arithmetic example

The expected volume of sale on the fuel station is assumed to be 3000 Nm³/day. It is interesting to know how many vehicles this volume can supply. Below follows an arithmetic example based on the calculated tank volumes in Table A3 13 and on a number of assumptions:

The first assumption is that 1 truck/h fuel LBG between 8 am and 5 pm.

$$\rightarrow 9 * 160.68 = 1446.12 \text{ Nm}^3$$

The second assumption is that 4 busses fuel CBG/day. These busses are assumed to be tourist busses. Bus fleets for public transportation usually have their own fuel supply system.

$$\rightarrow 4 * 76.09 = 304.36 \text{ Nm}^3$$

The rest is fuelled as CBG for CBG cars.

$$\rightarrow 3000 - 1446.12 - 304.69 = 1249.52 \text{ Nm}^3$$

$$\rightarrow 1249.52 / 20 \approx 62.5 \text{ cars/day}$$

If most of these cars is fuelled between 7 am and 1 pm this means:

$$62.5 / 13 \approx 4.9 \text{ cars/hour}$$

Energy balances

Table A3 14 Background information used in the energy balances.

Company/Technology:	Energy consumption:		Heat recovery: (kWh/Nm ³ clean biogas)	CH ₄ loss: (%)	LCO ₂ recovery: (%)
	Electricity (kWh/Nm ³ clean biogas):	Heat (kWh/Nm ³ clean biogas):			
Cryogenic Technology					
SGtS	0.8	-	Yes	0.5	~100
Acrion	1.42	-	-	“none”	30-50
Prometheus-Energy	1.54	-	-	“none”	-
Conventional technology					
Water scrubbing	0.40	-	Yes	0.1-1	-
Cooab	0.18	0.95	0.75	< 0.1	-
PSA	0.40	-	-	1-2	-
Small-scale liquefaction technology					
Cryostar/N ₂ cycle	0.63	-	-	-	-
Hamworthy/MRC	0.43	-	-	-	-

Abbreviations:

PE = primary energy

PEF = primary energy factor

The numbers on primary energy is given in kWh/Nm³ clean gas. If the original numbers are given in another unit these are first converted to kWh energy/Nm³ clean gas. Then the energy consumption is converted to primary energy by multiplying the primary energy factor:

$$PE_{\text{energy carrier}} = \text{Energy consumption} * PEF_{\text{energy carrier}} \text{ (kWh/Nm}^3 \text{ clean biogas)}$$

All used primary energy factors can be found in Table A3 15.

Table A3 15 Primary energy factors for different technologies and energy carriers.

Energy carrier:	Technology:	Primary energy factor:	Source:
Electricity:	Nordic mix	1.6	(Energimyndigheten, 2006)
	Coal condensing plant	2.74	(Energimyndigheten, 2006)
Heat:	Forest fuel boiler	1.16	(Persson et al, 2005)
	Natural gas boiler	1.29	(Persson et al, 2005)
Fuel:	Diesel	1.06	(Uppenberg et al., 2001)

The factors for conversion of heat are calculated from efficiency data on production of fuel and heat respectively, see Table A3 16. It is assumed that the heat is produced in a boiler without exhaust gas condensation.

Table A3 16 Efficiencies for the production of fuel and heat respectively (Persson et al, 2005).

Fuel:	Efficiency (%):	
	Production of fuel, including transportations	Heat production
Forest fuel	0.96	0.9
Natural gas	0.86	0.9

Methane losses

The influence of CH₄ losses on energy consumption has been examined through applying a factor of one unit plus the methane losses to the primary energy consumption for each upgrading technology:

$$\text{Net } PE_{\text{consumption}} = PE_{\text{upgrading of biogas/production of LBG}} * (1 + CH_{4, \text{ loss}}/100)$$

Waste heat recovery

Some of the companies presented in this report have developed systems to take care of the wasted heat, which is rejected in their systems, for use in other processes. These companies can be found in Table A3 17, together with the amounts of waste heat and the temperature of it.

Table A3 17 Companies taking care of waste heat and the amount and temperature of this heat.

Company:	Use of waste heat:	Temperature:	Source:
Läckeby Water	0.49 kWh/Nm ³ raw gas	60 °C	(Karlsson, 2008a)
SGtS	80 % of used electricity	55 °C	(Kättström, 2008)
Malmberg Water	80 % of used electricity	55 °C	(Malmberg, 2008)

When including the use of waste heat in the energy balances, this heat replaces another energy carrier. It is the primary energy factor for the replaced energy carrier that is used for the conversion to primary energy. In this case it is assumed that the biogas source is digester gas and that wasted heat will replace forest fuel in a boiler:

$$\text{Net PE}_{\text{consumption}} = \text{PE}_{\text{upgrading of biogas/production of LBG}} - (\text{Waste heat recovery} * \text{PEF}_{\text{forest fuel}})$$

The assumptions regarding CH₄ content in raw gas and product are the same as before, but when adding the heat recovery system to the water scrubber, the energy demand is raised from 0.24 to 0.26 kWh/Nm³ raw gas (original numbers). (Malmberg, 2008)

Pressure letdown station

When biogas is injected into the gas grid propane, C₃H₈, is added to raise the heating value to match the one of natural gas. The heating values for the different gases can be seen in Table A3 18.

Table A3 18 Heating value for different gases. (SGC, 2008b)

Gas:	Heating value (kWh/Nm ³):
CH ₄	9.97
Natural gas (Danish)	~11
Propane	25.89

The C₃H₈ part, X, in one Nm³ gas injected into the gas grid is calculated with:

$$(\text{CH}_4 \text{ part}) * 9.97(1-X) + 25.89X = 11 \text{ kWh/Nm}^3$$

This means that to one Nm³ biogas, Y Nm³ C₃H₈ must be added:

$$Y/(1+Y) = X$$

In Table A3 19 X and Y for different CH₄ concentration can be found together with the volume of biogas/propane mix to be compressed per volume clean biogas.

Table A3 19 Calculated amount of added C₃H₈ and volume to be compressed for different CH₄ concentrations per Nm³ clean biogas.

Character:	Unit:	97 % CH ₄	99 % CH ₄	100 % CH ₄
X	%	8.2	7.1	6.5
Y	%	8.9	7.6	6.9
V _{comp}	Nm ³ /Nm ³ clean biogas	1.089	1.076	1.069

To be able to inject biogas into the gas grid biogas must be compressed to 80 bar. To calculate the compression work the formula for isentropic work, no loss of energy, is used:

Isentropic compression work:

$$w_s = \frac{\kappa R(T_1 - T_2)}{\kappa - 1} = \frac{\kappa R T_1}{\kappa - 1} \left[1 - \left(\frac{p_2}{p_1} \right)^{(\kappa-1)/\kappa} \right] \quad (\text{kJ/kg})$$

$$\kappa = c_p / c_v$$

$$R = R_u / M, R_u = 8.314 \text{ kJ}/(\text{kmol} * \text{K})$$

$$\eta = \frac{\text{isentropic compression work}}{\text{actual compression work}} = \frac{w_s}{w_a}$$

The specific heat at constant pressure (C_p) and volume (C_v) vary with temperature and in the calculations a mean value for T_1 and T_2 is used. T_1 is set to 298.15 K and the right T_2 is found by trial and error:

- guessed T_2
- c_p and c_v at guessed T_2
- calculate w_s by using the right part in the equation above
- calculate T_2 by using the left part in the equation above
- if T_2 guessed \neq T_2 calculated → return to first point
- if T_2 guessed = T_2 calculated → right T_2 !

The efficiency is assumed to be 10 % and the needed compression work is approximately 0.15 kWh/Nm³ clean biogas.

The energy balance is calculated by adding the primary energy for the compression work to the primary energy for the purification and upgrading process. Since the energy balance is given in kWh/Nm³ clean biogas the volume to be compressed per Nm³ clean biogas is 1+Y. It is assumed that the compressors are driven with electricity bought from the grid:

$$\text{Net PE}_{\text{consumption}} = \text{PE}_{\text{upgrading of biogas}} + (w_{\text{comp}} * \text{PEF}_{\text{Nordic mix}} * V_{\text{comp}})$$

Gas distribution and fuelling

Table A3 20 shows the fuel consumption for different biogas distribution vehicles. It is an average, meaning that the vehicle is loaded one way and empty on the return. The fuel consumption for a truck with three mobile CBG storages is 0.45 l/km and, if the gas were transported in an efficient way, this would be the average fuel consumption since the truck would take three empty CBG storages in return. However, according to the supplier, the truck often delivers three mobile CBG storages and return without load, resulting in a fuel consumption of 0.35 l/km on the way back.

Table A3 20 Average fuel consumption for biogas distribution vehicles.

Vehicle:	Fuel consumption: (l/km)	Source:
Semi-trailer (LBG)	0.46	(Reinemo, 2008)
Mobile CBG storage	0.4	(Eriksson, 2008)

1 litre of diesel contains 9.8 kWh (SGC, 2007). This energy content has been converted to primary energy using a factor of 1.05. Further, the fuel consumption per km has been divided per loaded Nm³ and added to the primary energy consumption for upgrading of biogas or production of LBG and pressure raise. It is assumed that the biogas will be fuelled as CBG and the pressure raise is achieved through compression and pumping (see Table A3 21) for upgraded biogas and LBG respectively:

$$\text{Net PE}_{\text{consumption}} = \text{PE}_{\text{upgrading of biogas/production of LBG}} + \text{PE}_{\text{compression/pump work}} + (\text{fuel consumption}/(\text{km and Nm}^3) * \text{PEF}_{\text{diesel}} * \text{distance})$$

Table A3 21 Energy consumption for pressure raise through compression and pumping, respectively.

Pressure raise:	Electricity consumption, (kWh/Nm ³ clean biogas):	Source:
Pumping	0.055	Table A3 11
CBG station	0.25	(Karlsson, 2008b)

* Calculated from data from Cryostar and Vanzetti Engineering

LCO₂

Cryogenic transport temperature control:

If the vehicle is well insulated, the driver handles the cooling system in a correct way and the chilled or frozen goods have the right temperature when loaded the CO₂ consumption is 20-25 kg/h and cooling system. This CO₂ can replace around 1.5 l diesel/h. (Smedbro, 2008)

The CO₂ density is 1.978 kg/Nm³ (Air Liquide, 2008)

→ 25 kg CO₂/h = 12.64 Nm³ CO₂/h

Replaced diesel per Nm³ CO₂:

1.5/12.64 = 0.12 l/Nm³ CO₂

Energy content in diesel:

1 L diesel = 9.8 kWh

Primary energy factor: 1.05

→ 1 L diesel = 10.3 kWh primary energy

Replaced primary energy:

0.12*10.3 = 1.22 kWh/Nm³ CO₂

The replaced primary energy per Nm³ CH₄ depends on the CO₂ concentration in the raw gas.

Per each Nm³ LBG (100 % CH₄) produced X Nm³ CO₂ is separated:

$$\frac{CO_2}{CH_4} = X$$

$$\text{Net PE}_{\text{consumption}} = \text{PE}_{\text{production of LBG}} - (1.22 * X * (\text{CO}_{2, \text{recover}}/100)) \text{ (kWh/Nm}^3 \text{ clean biogas)}$$

Assume a digester plant with a gas flow of 800 Nm³/h and a CH₄ concentration of 65 %. Table A3 22 shows how many kWh of primary energy that can be replaced per Nm³ clean biogas and how many cooling systems than can be supplied with this gas stream. In the calculation it is assumed that a CO₂ cooling system and the LBG production plant are running 10 and 24 hours per day, respectively.

Table A3 22 CO₂ recovery, replaced diesel, in primary energy, and supplied numbers of cooling systems for the two companies that produce LCO₂ in their process. The numbers are valid for a raw gas flow of 800 Nm³/h and 35 % CO₂.

Company:	SGtS	Acrion	
CO ₂ recovery (%):	100	30	50
PE consumption for LBG production (kWh/Nm ³ CH ₄):	1.28	2.27	2.27
Replaced PE in CO ₂ cooling system (kWh/Nm ³ CH ₄):	0.66	0.20	0.33
Net primary energy consumption for production of LBG (kWh/Nm ³ CH ₄):	0.62	2.07	1.74
Numbers of supplied cooling systems	53	16	27

Cultivation of plants and vegetables:

The consumption of CO₂ in greenhouses is approximately 25 kg/m² and year (vegetables) and the size of a normal sized greenhouse is around 10,000 m². (Christensen, 2008)

Assume a digester plant with a gas flow of 800 Nm³/h and a CH₄ concentration of 65 %. Table A3 23 shows how many greenhouses that can be supplied, if the biogas plant is producing biogas 24 h per day, 365 days a year.

Table A3 23 CO₂ recovery and supplied numbers of greenhouses for the two companies that produce LCO₂ in their process. The numbers are valid for a raw gas flow of 800 Nm³/h and 35 % CO₂.

Company:	SGtS	Acrion	
CO ₂ recovery (%):	100	30	50
Numbers of supplied greenhouses (10,000 m ² /greenhouse):	19	6	10