BIOMETHANE LIQUEFACTION – TECHNOLOGY AND FUTURE POTENTIAL

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Liquid biomethane (LBM) may represent, under certain conditions, a competitive renewable energy resource and a promising biofuel for transport. LBM, with its carbon neutral footprint, is produced through the upgrading and liquefaction of biogas obtained by anaerobic digestion of organic material. Use of LBM is advantageous in reducing greenhouse gas emissions and ensuring a more sustainable environment. Given its importance, the LBM is constantly increasing, hence more biogas liquefaction plants will be installed in the future. This paper describes two small-scale biogas liquefaction plants: single mixed refrigerant process and nitrogen expander process. In addition, it shows the present and future production potential of biomethane and liquefied biomethane in Europe.

Keywords: liquid biomethane, single mixed refrigerant, nitrogen expander, potential

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1. Introduction

Biomethane is methane sourced from renewable biomass. The pre-stage of biomethane is known as biogas, which is produced by anaerobic digestion of organic material, such as manure, sewage sludge, the organic fractions of household and industry waste, and energy crops. Biogas is also produced during anaerobic fermentation in landfills and is, then, referred to as landfill gas [1].

Biogas produced from anaerobic digestion processes and landfill consists mainly of methane (CH_4) and carbon dioxide (CO_2). Apart from methane and carbon dioxide, biogas can also contain other gases and contaminants, such as water (H_2O), hydrogen sulphide (H_2S), nitrogen (N_2), oxygen (O_2), ammonia (NH_3), siloxanes and solid matter. The concentrations of these impurities mainly depend on the composition of the substrate from which the gas is produced. Typical properties of biogas generated from anaerobic digestion plants and landfill sites are summarized and compared with those of natural gas in Table 1.

Tab. 1 Characteristics of natural gas, biogas and landfill gas

Parameter	Landfill gas	Biogas (digestion)	Natural gas
CH_4 (vol %)	35–65	50–70	85–98
CO_2 (vol %)	15–40	50–30	0–1
C_xH_y (vol %)	0	0	<12
N_2 (vol %)	2–5	<1	0.3–5
O_2 (vol %)	0–5	0	<0.1
H_2S (vol %)	<1	<2	0–5
NH_3 (vol %)	<0.1	<1	0
H_2O (vol %)	<10	<10	0

Biogas can be utilized as a fuel for on-site heat, steam and electricity generation in industry, as a substrate in fuel cells, as a substitute of natural gas for domestic and industrial use prior to injection into natural gas grids and as a vehicle fuel [2]. Depending on the end use, different biogas treatment steps are necessary. When it is important to have a high energy gas product, e.g. as vehicle fuel or for grid injection, the gas needs to be upgraded, i.e. CO_2 must be removed [3].

Upgrading of biogas has gained increased attention due to increasing demand for renewable fuel as an alternative to fossil fuel in many countries in order to reduce greenhouse gas emissions. In Europe by 2030, the total biomethane production is estimated to be around 190 TWh corresponding to about 3 % of the European natural gas consumption [4].

Several different biogas upgrading techniques are on the market today including water scrubbing, chemical scrubbing, physical scrubbing, pressure swing adsorption, membrane separation, and cryogenic separation [5]. Further information about these technologies can be found in Table 2.

Biogas must be liquefied for certain applications especially when customer sites located remotely from a gas grid and for storing gas in case of production and developing new generation for biofuel. Liquefied biomethane occupies 600 times less space than in gaseous state under normal conditions and around 3 times less space compared to compressed biogas (CBG) at 200 bar [6], which makes it possible to store and transport it in large quantities. LBM can be transported over large distances and can be dispensed to either LNG vehicles or CNG vehicles.

There are two main ways to produce LBM and these are cryogenic upgrading technology and conventional upgrading technology connected with a small-scale liquefaction plant.

Tab. 2 Biogas upgrading technologies.

Operation	Technology	Description
Absorption	Water scrubbing	Water absorbs CO_2 under high pressure conditions. Regenerated by depressurizing.
	Chemical scrubbing	Amine solution absorbs CO_2. The amine solution is regenerated by heating.
	Physical scrubbing	Polyethylene glycol absorbs CO_2. It is regenerated by heating or depressurizing.
Adsorption	Pressure swing adsorption	Highly pressurized gas is passed through a medium such as activated carbon. Once the pressure is reduced the CO_2 is released from the carbon, regenerating it.
Membrane	Membrane separation	Pressurized biogas is passed through a membrane which is selective for CO_2.
Cryogenic	Cryogenic separation	Biogas is cooled until the CO_2 changes to a liquid or solid phase while the methane remains a gas. This allows for easy separation.

This work firstly describes and compares two small-scale liquefaction biomethane processes: single mixed refrigerant process and nitrogen expander process. Secondly, it shows the present and future potential of biomethane and liquefied biomethane production in Europe.

2. Small-scale liquefaction plant

The main way to produce liquefied biomethane is to upgrade the raw biogas then liquefy methane 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 3.

Tab. 3 Maximum component concentrations acceptable to liquefy biogas. [7]

Component	Required (ml.m^{-3})
Carbon dioxide (CO_2)	< 25
Hydrogen sulphide (H_2S)	< 4
Water (H_2O)	< 1

If the upgrading method does not reach these requirements an additional treatment step is needed before liquefaction.

Due to the similarities between the biogas and natural gas (NG), NG liquefaction processes (single mixed refrigerant, nitrogen expander) can be adopted for biomethane liquefaction. These liquefaction processes are classified into two groups: open-loop cycle and closed-loop cycle which are described below.

2.1. Open-loop cycle

In the open-loop cycle, the refrigerant which is used as a cooling agent in liquefaction is a part of the gas stream. The biogas upgrading (feed gas) is compressed (CP) and then cooled down to ambient temperature via heat exchangers (HE). LBM is then produced in a turbo-expander at the same time as work is extracted. This work produced can be used to supply the compressor with a part of its work input. Finally, the liquefied biomethane (LBM) is sent to a flash tank to remove gas content. See Figure 1.

2.2. Closed-loop cycle

In contrast to open-loop cycle, the refrigerant is not a part of the gas stream in closed-loop cycle. The refrigerant is supplied externally. In this process, the biogas is not compressed before being heat exchanged. This results in a lower temperature difference between two streams biogas upgrading and cryogenic refrigerant.

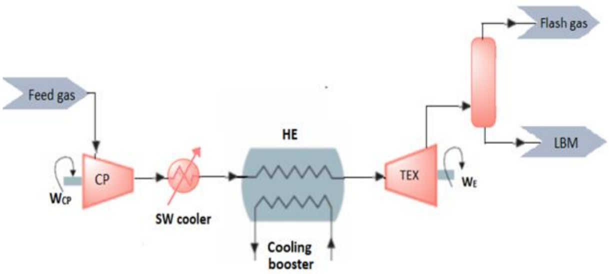

Fig. 1 Open-loop cycle proces diagram. [8]

The cooling process can be driven by a single cryogenic refrigerant as methane or nitrogen (Nitrogen/Brayton cycle) or a mixture of these with other hydrocarbons (Mixed-refrigerant cycle).

The biogas upgrading (feed gas) is first cooled and partly condensed to a low temperature in the cryogenic heat exchanger (HE) and thereafter expanded through a throttling valve (VLV). This decreases both the pressure and the temperature which results in condensation of the methane. Finally, any gas that is dissolved in the liquid biomethane (LBM) is separated in the flash tank [8]. See Figure 2.

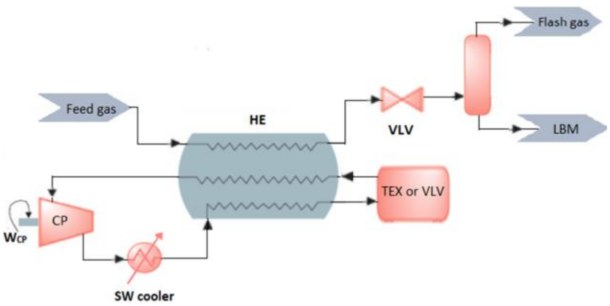

Fig. 2 Open-loop cycle proces diagram. [8]

3. Description of biomethane liquefaction processes

3.1. Single mixed refrigerant process (SMR)

Single mixed refrigerant process, also called PRICO (Poly Refrigerated Cycle Operation) process, uses a single cycle of mixed refrigerant made up of nitrogen, methane, ethane, propane and butane [9, 10]. This process includes two blocks: the mixed refrigerant and the upgraded biogas blocks [11].

In the first block, upgraded biogas is first cooled by indirect heat exchange with the cooled mixed refrigerant stream into the heat exchanger (LNG-HE). Then, the cooled gas is expanded through a throttling valve (VLV). Finally, the liquefied biomethane (LMB) obtained at −162°C and atmospheric pressure is separated to flash gas in the flash separator (SEP).

In the second block, the mixed refrigerant is pressurized by the compressor (K-1) and then pre-cooled in the water cooler (E-1). After that, it is cooled in the cryogenic heat exchanger (LNG-HE). Refrigerant leaving the cryogenic heat exchanger is expanded across a throttling valve (VLV) to obtain low pressure refrigerant at a low temperature. After expansion, the refrigerant enters the cryogenic heat exchanger (LNG-HE) for the second time cooling the incoming upgraded biogas flow. Finally, the vaporized refrigerant is returned to the compressor (K-1) and the cycle is repeated. This process is shown in Figure 3. [12]

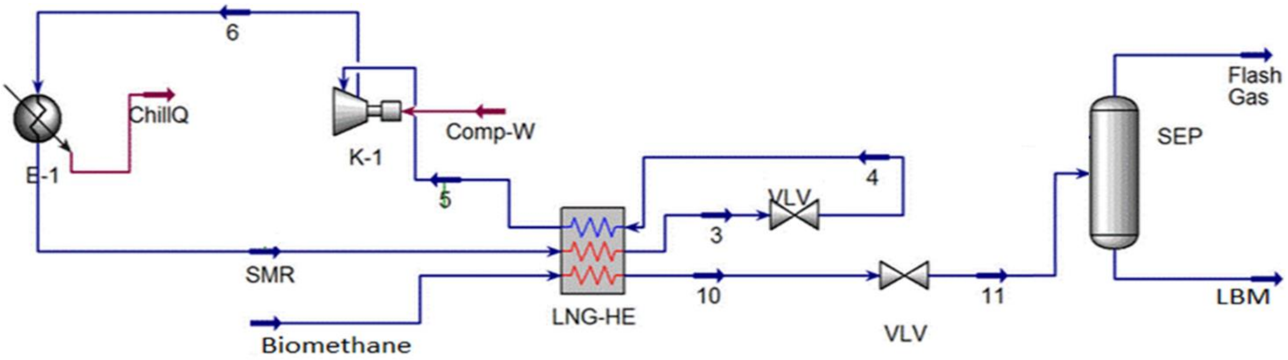

Fig. 3 Process Flow Diagram of single mixed refrigerant (SMR) process

Its low production capacity can be considered a disadvantage for this process because more trains will be required to produce in high capacities [9]. In contrast, this process has many advantages. Khan [13] summarizes SMR:

-it is an effective process.
-it has the lowest capital cost of all competing.
-it is a simplified refrigeration system require minimum equipment and simplified control.
-it has a compact and modular design using an aluminium plate fin exchanger.
-it is flexible in feed gas composition because it receives upgraded biogas from multiple sources.

3.2. Nitrogen expander process

In this process, also called closed Brayton cycle [14], nitrogen in gaseous state is used as a refrigerant.

This process is very similar in design to the single mixed refrigerant. Both include two blocks: one for liquefaction of upgraded biogas and the other for the refrigerant. See Figure 4. The only difference between them is that the refrigerant leaving the cryogenic heat exchanger is expanded in a turbo-expander (K-2) in this process instead of a throttling valve in the single mixed refrigerant [12]. The throttling valve provides only cooling while a turbo-expander produces work (that can drive the compressor) in addition to cooling [15].

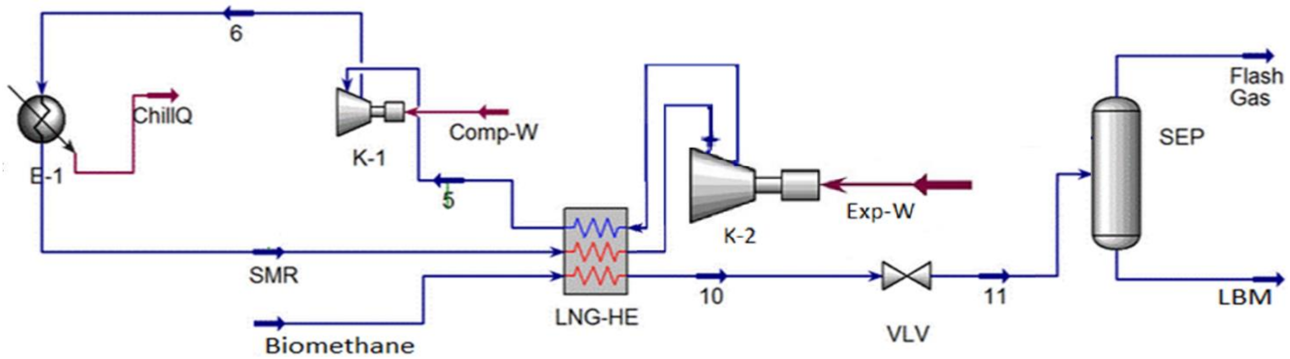

Fig.4 Process Flow Diagram of nitrogen expander process

The nitrogen expander process has many advantages: cost-effective, safety (no-hydrocarbons in the refrigerant cycle) and simplicity of operation. But in contrast, this process has a low efficiency which is due to the fact that a single refrigerant is used in a single phase (gas). Besides that, it is used over a wide range of temperatures. Thus, the temperature difference in some temperature domains is large which leads to thermodynamic inefficiencies and hence higher power requirements [16].

4. Potential of liquefied biomethane in Europe

4.1. Present status

Biomethane production has seen a significant growth in the recent years in Europe.

Nowadays, in Europe, biomethane is produced in 15 countries in 497 upgrading plants with a total capacity of approximately 1 million $m^3.h^{-1}$ [17]. The produced biomethane is used in the following applications; as vehicle fuel (in 12 countries), fed into gas grids (in 11 countries) or for heating purposes.

Most biomethane production plants are in Germany (194 plants), UK (85 plants) and Sweden (63 plants). In other countries, the biomethane production volumes are still marginal. [17]

Table 4 shows the production biomethane in each country of Europe for the year 2018. [17]

Only a very few numbers of plants, in Sweden, Norway, UK and a smaller plant in the Netherlands, produce liquefied biomethane in Europe today to be used as vehicle fuel. The liquefaction plants that exists uses biogas from anaerobic digestion, in UK from landfills and the others from biogas plants.

Lidköping (SE)

The plant in Lidköping is the first LBM production plant in Sweden. It is operating since April 2012. It uses industrial waste and agricultural waste for biogas production through digestion. The biogas is then upgraded in a water scrubber to fulfil the Swedish standard for biogas as a vehicle fuel. After that, the gas enters the liquefaction unit consisting of a pressure swing adsorption unit polishing step to remove further CO_2 (<25 $ml.m^{-3}$) followed by the liquefaction process. The technology for methane liquefaction is based on a closed nitrogen Brayton cycle. The capacity of the plant is 1.2 m³ LBM/h corresponding approximately to a production capacity of 4000 tonnes of LBM per year. [18]

Albury (UK)

At Albury landfill site in Surrey, the UK's first plant for liquefied biomethane opened in June 2008. From the collected landfill gas contaminants are removed and the gas is upgraded by pressure swing adsorption. Biogas is compressed to a pressure between 4-10 bar and is fed to a vessel (column) where is put in contact with an adsorbent (carbon molecular sieves). The purified CH_4 which is recovered at the top of the column is then liquefied by a closed-loop mixed refrigerant cycle. The production capacity of this plant is 5000 tonnes of LBM per year. [19]

Nes (NO)

A plant to produced biomethane was built and inaugurated February 2014 in Nes close to Oslo, Norway. The gas from household waste is upgraded to vehicle quality by a water scrubber unit. After compression to 20- 30 bar in an oil free piston compressor the gas is polished in order to further remove carbon dioxide and hydrogen sulphide. This polishing step is done by pressure swing adsorption in a molecular sieve. After that, the produced biomethane is liquefied by a closed-loop mixed refrigerant cycle. The production capacity of this plant is 4000 tonnes of LBM per year. [20]

Tab. 4 Biomethane production in Europe for the year 2018. [17]

Country	No. of plants	Production ($m^3.h^{-1}$)
Austria	15	2 685
Denmark	22	7 109
Finland	12	1 507
France	30	4 387
Germany	194	933 494
Hungary	2	800
Iceland	2	550
Italy	7	N/A
Luxembourg	3	680
The Netherlands	26	14 212
Norway	4	1 275
Spain	1	6 500
Sweden	63	14 620
Switzerland	31	3 333
United Kingdom	85	61 170
Total	497	1 052 322

4.2. Future potential

Several studies have been performed to determine the potential of biogas and biomethane. The potential for biogas and biomethane production is in some studies calculated as a theoretical potential and sometimes as the realistic potential at a specific year taking into account economic, technical as well as political factors.

The predicted biogas production estimated from all countries in Europe is around 326 TWh for year 2020 and 500 TWh in 2030 if the development is continuing at the same pace. [21]

The share of biogas to be upgraded to biomethane is estimated by the Green Gas Grid project to be 25 % of the biogas production in 2020 and 38 % in 2030, corresponding to 80 TWh in 2020 and approximately 190 TWh in 2030. For 2030, this corresponds to about 3 % of the European natural gas consumption. [5]

As shown, the production of liquefied biomethane is still limited in Europe, and only a few liquefaction biogas plants have been established.

This lower development of the LBM market seems to be related to political incentives, technology and economy rather than to supply of feedstock. The development of the market for liquefied biomethane is dependent on several factors; among others:

- A sufficient biomethane potential for a region or a country.
- EU and national targets aiming at increasing the market share of LBM including long-term incentives.
- Availability of LBM driven vehicles.
- Development of distribution networks like fuelling stations.
- Maturity and cost competitiveness of the upgrading/liquefaction techniques.
- Market mechanisms - demand for LBM as a vehicle fuel.

Based on the criteria's stipulated above, several studies have been carried out. The results of these studies show that the production potential for LBM in Europe could reach 7 TWh in 2020 and 20 TWh in 2030 [5]. Some countries, most likely the ones where the LBM production has already started, will be the forerunners showing the strongest growth in the near future. When the technology is tried in many commercial-scale plants and the infrastructure associated with the distribution is in place more countries will follow.

5. Conclusion

LBM is produced after cleaning (the trace components harmful to the end use are removed), upgrading (CO_2 is removed to obtain a high energy gas product) and liquefaction of biogas. In a biogas liquefaction plant, purified and upgraded biogas is converted to liquefied biomethane under -162 °C at atmospheric pressure. Feasible small-scale liquefaction plant is either a closed nitrogen/Brayton cycle or a closed single mixed refrigerant cycle. These two processes differ by their configurations, components and working fluids, and have therefore various operating conditions and equipment inventory.

Nitrogen/Brayton cycle uses turbo-expander to decrease the pressure and temperature of the refrigerant stream instead of throttling valve used by mixed refrigerant cycle. In addition, nitrogen/Brayton has low efficiency but it is not as complex as the mixed refrigerant cycle since it only uses one refrigerant (nitrogen).

Rapid growth in the population and increasing urbanization and modernization across the globe is increasing the demand for energy. Nowadays, major energy demand is supplied by renewable fuel sources of clean energy to reduce the consumption of fossil fuels and to bring down the effect of global warming.

For this reason, the production potential of renewable gas is estimated to increase in the future. In Europe, the total biogas production is estimated to be around 326 TWh in 2020 and 500 TWh in 2030. Of the total biomethane production of 80 and 190 TWh for the year 2020 and 2030 respectively, it is estimated that some 10 % of the potential would be converted to Liquefied biomethane (LBM), which is particularly beneficial either in locations where gas infrastructure for injection is missing or where can be used as automotive fuel.

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