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REVIEW

Biogas upgrading by cryogenic techniques

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Abstract

The scarcity of fossil fuels and the worldwide pollution have led the scientific community to seek renewable energy alternatives. In particular, biogas has become a potential alternative fuel to be employed instead of traditional energies. Biogas is mainly composed by methane (CH_4) and carbon dioxide (CO_2). To obtain pure biomethane, a proper biogas upgrading to remove CO_2 and other minority compounds is needed. For this purpose, upgrading processes have been developed, such as water or chemical scrubbing, membrane separation, pressure swing adsorption, and cryogenic techniques. Cryogenic techniques represent a good option to be optimized because these techniques yield high-purity products, ranging between 95 and 99%. Therefore, we present here a review on cryogenic techniques. In spite of many advantages, the high-energy penalty makes cryogenic techniques commercially inapplicable actually. Several authors have proposed novel configurations to reduce the energy consumption. Cryogenic packed-bed technology was recently tested in a coal-fired plant with an energy consumption of 1.8 MJ/kg CO_2. Economic analyses were carried out for anti-sublimation CO_2 capture, giving a cost of 34.5 €/ton CO_2. Among the different alternatives of cryogenic hybrid systems, cryogenic membrane processes stand out due to a 54.4% of capital cost savings.

Keywords Biogas upgrading · Cryogenic techniques · Hybrid cryogenic systems · CO_2 utilization

Introduction

Biogas from anaerobic digestion of biomass is a promising renewable energy source. Biogas composition is based mainly in methane (CH_4) and carbon dioxide (CO_2), although there are minor presence of other chemicals such as hydrogen sulfide (H_2S), nitrogen (N_2), oxygen (O_2), and siloxanes (Aguirre-Villegas et al. 2015; Montingelli et al. 2015; Chatterjee et al. 2016; Sahota et al. 2018). CH_4 content in biogas ranges between 50 and 70%, which makes this renewable energy suitable for replacing traditional natural gas (Álvarez-Gutiérrez et al. 2015; Kadam and Panwar 2017; Angelidaki et al. 2018; Kougias and Angelidaki 2018). Nevertheless, both CO_2 and the minority contaminants should be removed to meet the technical specifications of different countries for its injection in natural gas grid (Petrakopoulou et al. 2015; Miltner et al. 2017; Xiao et al. 2019), and many efforts have been done by scientific community for this end (Zhang et al. 2014a, b, 2018; Yan et al. 2014; Ravina and Genon 2015; Baena-Moreno et al. 2018a, 2019a; Chaterjee and Krupadam 2018; Liu et al. 2018; Pan et al. 2018). For this purpose, in the last years, the number of biomethane production operating plants in Europe has been doubled as shown in Fig. 1 (Baena-Moreno et al. 2019b).

Biogas upgrading technologies have been studied in deep by several authors (Kadam and Panwar 2017; Ullah Khan et al. 2017; Baena-Moreno et al. 2018a; Hajilary et al. 2018), and some of the latest studies are summarized in Table 1. The most known biogas upgrading techniques are water scrubbing, organic physical scrubbing, chemical scrubbing, pressure swing adsorption systems, membrane technology, and cryogenic separation (Sun et al. 2015). According to the literature (Persson et al. 2007; Deremince and Königsberger

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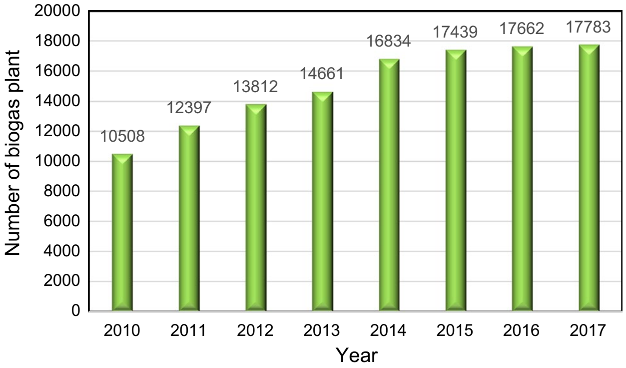


Fig. 1 Number of biogas plants in operation in Europe in 2010–2017. Note the fast increase from 2010 to 2014 and the slow down from 2014. Adapted from EBA (2018)

2017), the commercial technology most extended for biogas purification is water scrubbing, followed by membrane technology and chemical scrubbing.

Cryogenic separation is the less employed as commercial technology due to the high investment and operation costs (Tuinier et al. 2011a; Pan et al. 2013; Song et al. 2018). Nevertheless, in the last years, the research projects by this technology have increased to develop new approaches to minimize the extra cost, since cryogenic technologies have advantages such as avoiding the use of chemical solvents with no secondary pollution (Pellegrini et al. 2018). Table 2 includes some of the more recent works carried out with cryogenic technologies. In this sense, the use of cryogenic technologies for future works needs a review of the advantages and challenges for different existing configurations (Song et al. 2019). The present paper proposes a comprehensive guide for those exploring this technology as solution for biogas upgrading, focusing on CO_2 capture as majority contaminant but also giving insights for removal of minority compounds. Different configurations for low-temperature biogas upgrading are reviewed and presented, analyzing the advantages and handicaps which affect the overall process performance. Furthermore, hybrid systems which involve cryogenic technologies are presented as a potential solution for the low-temperature employment to upgrade biogas.

Cryogenic biogas upgrading

The bases of this technology are the different liquefaction temperatures for biogas compounds (Yousef et al. 2018). A gradual decrease in the gas temperature allows the selective separation of CH_4 from the rest of the components (Tan et al. 2017b). Thus, a high-purity biomethane is obtained in agreement with the standards marked by each country. This gas product is usually known as liquefied natural gas. The easiest path to remove the impurities contained in biogas by means of cryogenic methods employs a constant pressure of 10 bar (Song et al. 2019). The liquefaction is achieved by decreasing the temperature sequentially in order to remove each contaminant (or some of them) in different steps. The first point is usually set up at − 25 °C, where mainly H_2O, H_2S, and siloxanes are obtained. A second set point is appointed at − 55 °C to liquefied partially CO_2, followed by a new decrease until

Table 1 Recent biogas upgrading studies

Scope	References
Comparison of water scrubbing, cryogenic separation, amine scrubbing and caustic scrubbing as biogas upgrading processes in terms of efficiency	Hosseinipour and Mehrpooya (2019)
CO_2 and H_2 fermentation under mesophilic conditions to produce volatile fatty acids as well as biogas upgrading to biomethane in one stage	Omar et al. (2018)
Microbial electrochemical application to separate CO_2 from biogas and posterior regeneration to be employed	Kokkoli et al. (2018)
Hybrid biogas upgrading configuration proposal composed of two-stage thermophilic reactors	Corbellini et al. (2018)
Pressure swing adsorption through carbon molecular sieve adsorbent to upgrade biogas	Canevesi et al. (2018)

Table 2 Recent cryogenic technologies works

Scope	References
A novel cryogenic CO_2 separation process and cold recovery, as well as optimization of the innovative process	Spitoni et al. (2019)
Flue gas from oxy-fuel combustion is purified by cryogenic technologies	Knapik et al. (2018)
New combined cycle for CO_2 cryogenic capture and hydrogen production	Mehrpooya et al. (2017)
Simulation of a novel proposal for cryogenic CO_2 capture by Stirling cooler	Song et al. (2017a)
Evaluation of viscosity and thermal conductivity for CO_2 cryogenic mixtures	Tan et al. (2017a)

– 85 °C to completely removed the remaining CO_2 by a solidification stage (Riva et al. 2014). The liquefied CO_2 obtained in the second temperature point can be sold as high-purity by-products to enhance the overall economic process performance. Another more typically employed option consists of a preliminary dry of the gas followed by a multistage compression up to 80 bar. This allows keeping a higher operational temperature of between – 45 and – 55 °C, having as main disadvantage a necessary intermediate cooling in the multistage compression (Awe et al. 2017). In addition to those explained before, in the recent years, others configurations have been proposed and tested by several authors (Johansson 2008; Tuinier et al. 2010, 2011b; Kumar et al. 2011; Ryckebosch et al. 2011; Langè et al. 2015; Maqsood et al. 2017). These processes can be grouped, and they are explained in deep in the next sections.

Cryogenic distillation

Overall, this technique is characterized for having a high energy demand which makes the operational cost less competitive compared to other biogas upgrading technologies (Langè et al. 2015). To mitigate this high energy consumption, several ideas have been proposed during the last years (Zanganeh et al. 2009; Li et al. 2013; Xu et al. 2014; Ebrahimzadeh et al. 2016). These solutions are commonly based on integration and intensification techniques joined with hybrid systems, obtaining promising reductions in energy consumption. For instance, Maqsood et al. 2017 obtained an enhancement of almost 70% by mixing process intensification and hybrid cryogenic distillation.

Despite that this technology has not been tested for biogas upgrading under real conditions, its validity for CO_2 separation in natural gas purification has been reported by several authors (Maqsood et al. 2014a, b, c). Hence, this is a novel idea for those interested in biogas upgrading via cryogenic technologies. Figure 2 shows a typical scheme for natural gas cryogenic distillation. In this system, the raw gas is cooled in two single steps before coming into the distillation column, where the two final products are separated. The top product contains the majority of the CH_4 and is extracted from the column by a partial condenser. In natural gas processing, this stream is high-purity CH_4. Nevertheless, in biogas upgrading, necessary analysis should be done to corroborate whether any other compounds are present in the stream. On the other hand, the bottom product reveals a majority composition of high-purity CO_2 which could be sold to improve the overall economic of the process. CO_2 bottom stream is partially recycled to the column previous vaporization to keep a proper vaporization heat, whereas the other part is extracted as product.

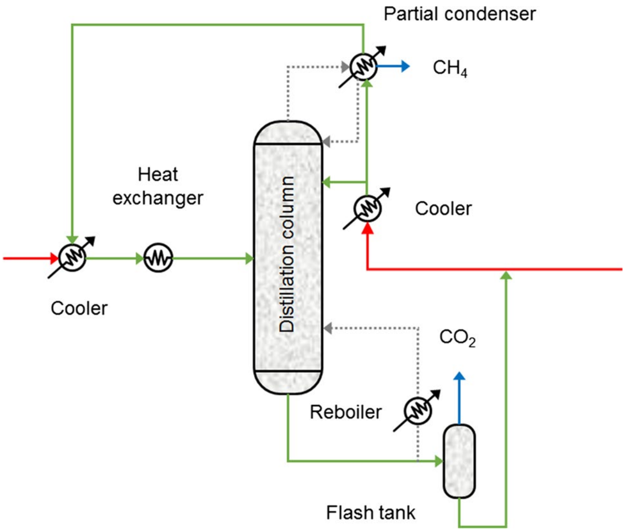


Fig. 2 Process scheme for natural gas purification. Adapted from Song et al. (2019)

Cryogenic packed bed

Currently, cryogenic packed beds are typically investigated to CO_2 capture from natural gas and/or high CO_2 content flue gas. Nevertheless, Tuinier and Van Sint Annaland (2012) applied their innovative system previously proposed in Tuinier et al. (2011b) to upgrade biogas. Their proposal based on numerical simulations proved to give better CH_4 purity and recovery than other techniques such as pressure swing adsorption and at the same time showed a higher productivity. As the main disadvantage, the H_2S removal needed of a – 150 °C initial temperature, which is a counteractive in energy demands. Figure 3 shows a simplified scheme of the process. Moreover, Figs. 4 and 5 show both the base and the improved scenario (named as reverse flow) proposed for biogas upgrading (Tuinier and Van Sint Annaland 2012). Energy requirements for cryogenic packed bed range from 263.4 kW in reversed flow scenarios and 390.7 kW in the basest case. In the process, about 20% of enhancement is obtained per kg of CH_4 versus pressure swing adsorption (2.9 MJ vs. 3.7 MJ). Nevertheless, thermal insulation needs improvement in order to reduce latent heat loss.

Further investigations in this technology to remove CO_2 from natural gas were studied by several authors. These configurations can be applied also for biogas upgrading, even though some of the proposed systems may need some changes. Ali et al. (2014) explored an experimental study for countercurrent-switched cryogenic packed beds by tuning different important parameters such as temperature profiles, feed composition, and feed flow rate. It is mainly concluded that reverse configurations can be of great interest for higher CO_2 contents in natural gas, which can be a major reason for testing in biogas upgrading. In a more recent study, Ali

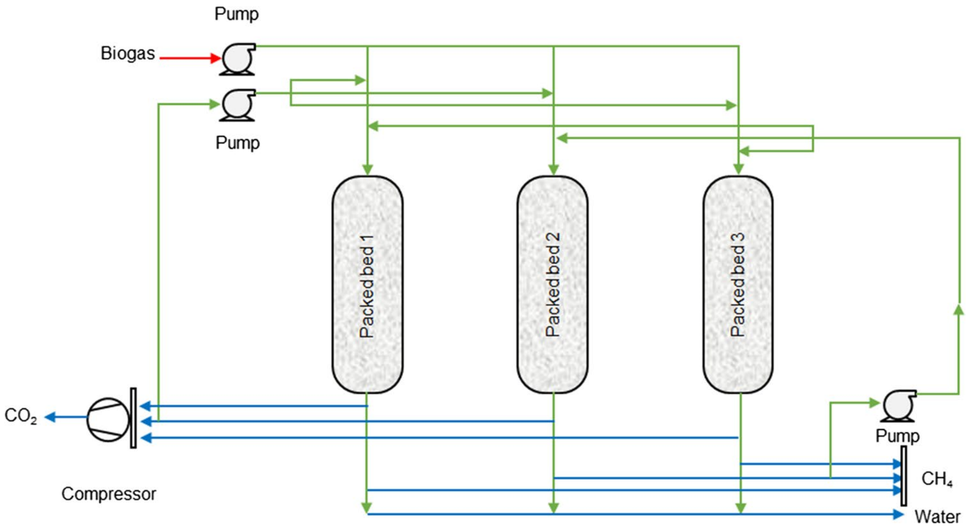


Fig. 3 Cryogenic packed bed scheme process for biogas upgrading. Adapted from Tuinier and Van Sint Annaland (2012)

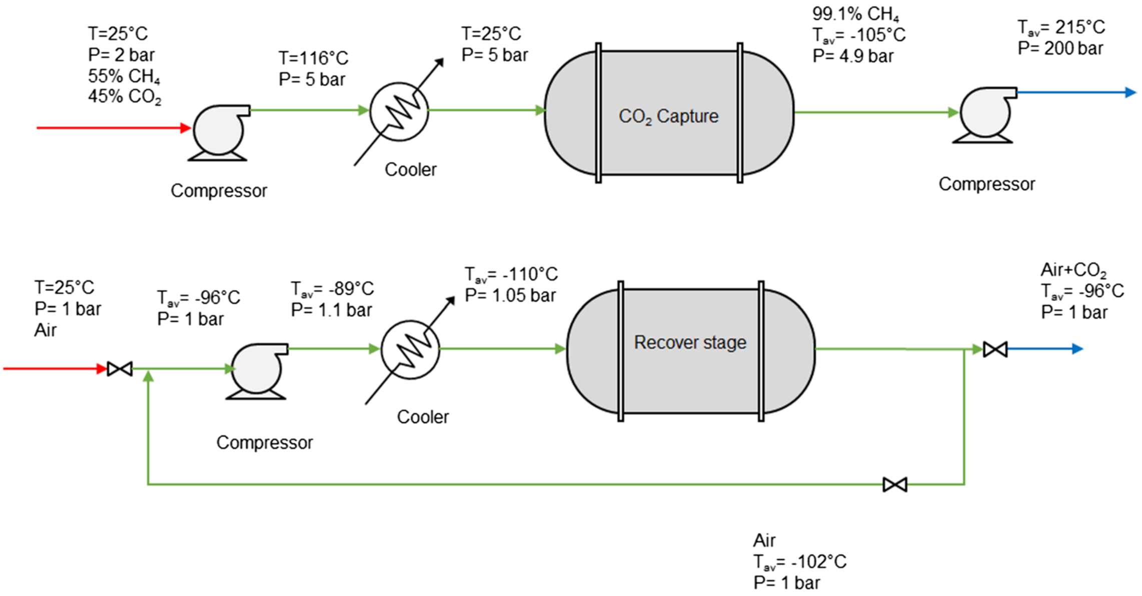


Fig. 4 Base case for cryogenic packed bed biogas upgrading. Adapted from Tuinier and Van Sint Annaland (2012)

et al. (2018) proposed a cryogenic packed bed network configuration based on a node-edge diagram in order to reduce hydrocarbon losses. In their study, a simulation for optimizing the energy consumption of the process by varying temperature, pressure, and raw gas composition was analyzed. In their optimal strategy, they achieved 94% product purity with 16% of hydrocarbon losses, demonstrating the potential commercial. Finally, they carried out a comparison between their previous experimental results and the new ones from the simulation, obtaining good concordance among them. DiMaria et al. (2015) simulated syngas purification in cryogenic packed beds, which could serve as an approach for biogas upgrading. Various syngas sources were analyzed and compared regarding energetic viability. Testing cryogenic packed bed technology in a coal-fired power plant supposed an energy consumption of 1.8 MJ/kg CO_2 (Tuinier et al. 2011b).

Anti-sublimation CO_2 capture process for biogas upgrading

The process called anti-sublimation consists of five stages in which CO_2 is obtained in liquid phase as final product. Even though there are no studies for biogas upgrading with this technique, it is considered a potential process to remove both minority elements and CO_2. The complete process is

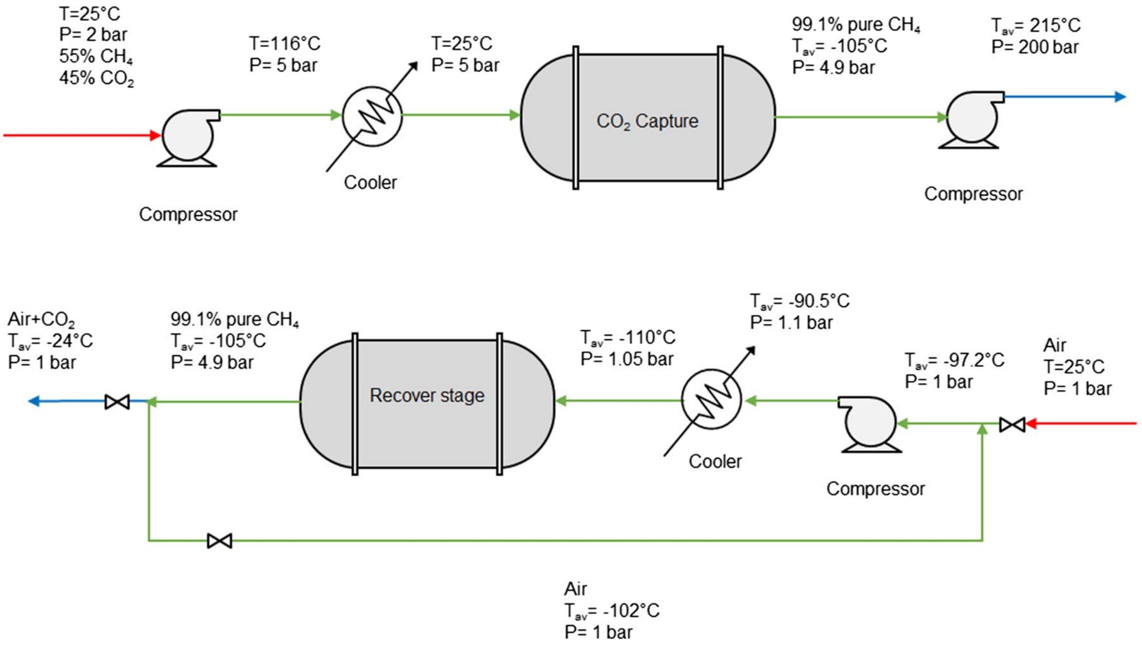


Fig. 5 Improved scenario for cryogenic packed bed biogas upgrading. Adapted from Tuinier and Van Sint Annaland (2012)

given in Fig. 6. According to Pan et al. (2013), the five stages of this CO_2 capture alternative are summarized in the next points:

- Flue gas clean-up and cooling down to − 40 °C with moisture removal, in which minority pollutants are removed. Furthermore, dehumidification for water removal is also carried out in this first stage.
- Heat exchanger between rich flue gases and poor flue gases.
- Refrigeration-integrated cascade, which was developed for liquefied natural gas applications by combining distillation and compression.
- CO_2 freezing heat exchanger. The mission of this stage is to control the defrosting process in order to consecutively sublimate and melt CO_2. As a consequence, at the end of this stage, both liquid and gas CO_2 are obtained.
- Final CO_2 recovery in liquid phase is with 99.9% purity.

Bench-scale pilot plants have served to test the anti-sublimation process in a 660-MW boiler (Clodic et al. 2005a, b). Under the conditions imposed of 15.47%, 60° C and 120 kPa, an energy penalty between 3.8 and 7.2% of the overall power efficiency was achieved, which are lower values comparing with other techniques (Romeo et al. 2008; Patiño-Echeverri and Hoppock 2012; Goto et al. 2013). More advantages of anti-sublimation process have been reported in the literature. For instance, the heat of fusion can be harnessed through its recovery on the surface of the heat exchanger in the defrosting stage, as well as the latent heat of fusion can be employed to quench quickly the liquid blend of refrigerants in a stage previous to evaporation. Nevertheless, it would be extremely necessary a deep study to corroborate the impact of the minority elements in case of biogas upgrading such as H_2S and siloxanes. These compounds could imply the modification of the general process and/or the addition of new stages. Economic studies were analyzed by Clodic et al. (2005b). They found that the overall cost to mitigate CO_2 emissions by means of anti-sublimation process was 34.5 €/ton. Regarding energy consumption, it was evaluated in a coal-fired power plant obtaining 1.18 $GJ_{electrical}$/ton CO_2, with a 90% of CO_2 recovery (Pan et al. 2013).

Advantages and handicaps of cryogenic technologies

The previous explained cryogenic technologies have some potential advantages which should be taken into account when exploring different options for biogas upgrading. Among these advantages, two of them clearly stand out: the high purity of the CO_2 obtained and the high pressure in which it is obtained; the potential of using the final CO_2 at low temperature as energy source. The first one mentioned above is the key point for either storage or utilization of CO_2. If the end of the CO_2 captured is the storage, the high pressure of the CO_2 product favors the transport along the pipeline systems. Additionally, for carbon capture and utilization, the high purity of the product obtained makes it

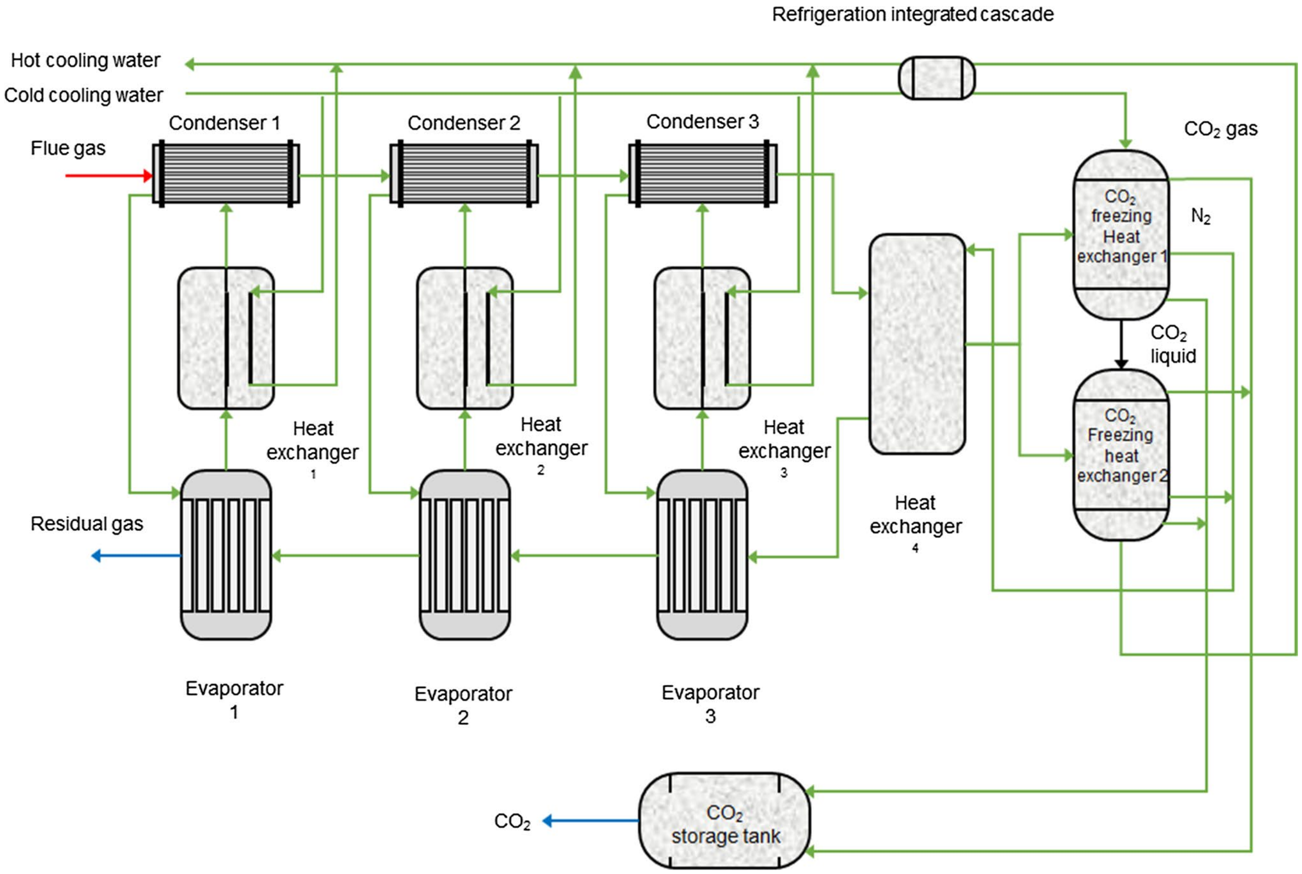


Fig. 6 Anti-sublimation process scheme. Adapted from Song et al. (2019)

usable for industries to produce chemicals and as feedstock (Baena-moreno et al. 2018a, b). As for the second advantage, Baxter's group has proposed an integrated system to take advantage of the cold energy source by their process energy storage for cryogenic carbon capture (Fazlollahi et al. 2015, 2016). The proposed process is capable of store CO_2 stored energy during non-peak demand periods for its reuse as refrigerant, achieving a 40% extra production when using this available energy. Nevertheless, there are still some challenges which need to be overcome by cryogenic technologies (Song et al. 2019). The disadvantages of using cryogenic technologies for biogas upgrading are summarized as follows:

- The availability and form of the cryogenic sources. In packed bed cryogenic technology, as the cold energy source is liquid nitrogen gas, the employment of this technology depends on the availability of it.
- The still high price of the CO_2 capture cost which is a common parameter with other capture technologies. For cryogenic distillation, need of compressors and coolers as cold energy sources make the overall installation cost higher than other biogas upgrading technologies.
- The overall efficiency depending on the operating temperature and environmental conditions of the location. Locating the plant in cold environments clearly benefit the efficiency of the process.
- The minority compounds are included in the raw biogas. As previously exposed, H_2S or siloxanes could damage the installations due to corrosion phenomena. To avoid this problem, specific building materials must be acquired and hence the capital cost is higher.

Cryogenic hybrid systems for low-temperature biogas upgrading

For carbon capture and storage technologies, compression is a necessary step after CO_2 capture. As for cryogenic techniques, CO_2 is already in high pressure to be transported through the pipeline, and some authors have proposed combined systems in which, generally, in the first section the majority of CO_2 is recovered and finally separated in the second stage. Moreover, hybrid technologies allow implementing a multi-objective optimization technique since the degree of freedom is higher. As the hybrid systems are composed by techniques which have proved to be valid for biogas upgrading, the overall hybrid system must be efficient in these terms. Another key point of cryogenic hybrid systems is the high-purity CO_2 product which is obtained for its utilization in other industries. Additionally, the absence of other kinds of solvents in the majority of these technologies favors the minimization of secondary pollution. Nevertheless, there are some challenges to be faced by hybrid-cryogenic technologies which are summarized in Table 3. These techniques are presented below as promising technologies for further investigation.

Cryogenic adsorption processes

Figure 7 shows a hybrid process which consists of an initial adsorption CO_2 recovery system followed by a cryogenic unit, allowing CO_2 to be obtained as liquid (Fong et al. 2016). This system gave as optimal result a reduction until 1.40 GJ/t CO_2 of energy consumption with an 88.9% of recover, according to the data reported. Additionally, the liquid CO_2 showed to be a high-purity stream and can be directly pumped for transportation. Other study tested the validity of adsorption processes in zeolite 4A at low temperatures to upgrade natural gas and hence the production of liquefied natural gas (Grande and Blom 2014). Authors found in their study that in the first stage CH_4 adsorption is controlled by kinetic, whereas CO_2 is controlled by equilibrium. Other study with the similar process showed that a 90.7% CH_4 recovery can be achieved with 41.8 ppm of CO_2 in the stream with a lower power consumption of 2.2 MW (Moreira et al. 2017).

Cryogenic membrane processes

This process arises as the most potential alternative which can join the advantages of cryogenic and membrane technologies in one hybrid system. The main advantage of the typical cryogenic membrane process (represented in Fig. 8) is its more cost-efficient economic analysis compared with traditional Monoethanolamine (MEA) absorption. It was reported a 9% of cost reduction per CO_2 ton with an 85% of CO_2 capture (Anantharaman et al. 2014). For biogas

Table 3 Main challenges to overcome by the hybrid cryogenic technologies treated in this work

Configuration	Main challenges	References
Cryogenic adsorption	Pretreatment for moisture removal High energy demand	Fong et al. (2016) and Song et al. (2019)
Cryogenic membrane	Necessity of an O_2 enrichment unit Membrane fouling	Scholes et al. (2013) and Song et al. (2019)
Cryogenic hydrate	Technology maturity Process optimization required	Song et al. (2019)
Cryogenic absorption	Toxicity Volatility	Hanak et al. (2015)

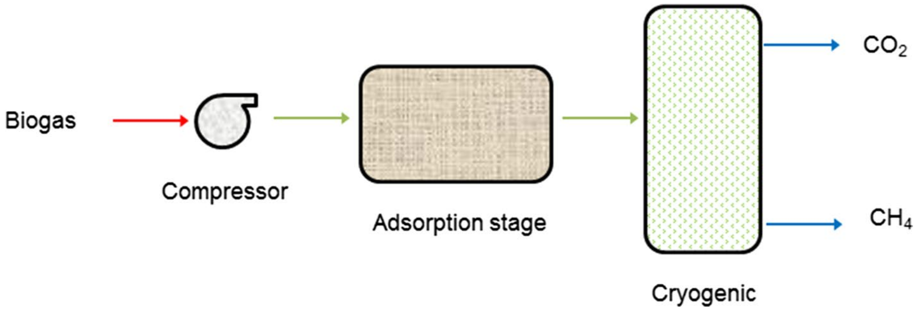


Fig. 7 Process diagram for cryogenic adsorption biogas upgrading. Adapted from Fong et al. (2016)

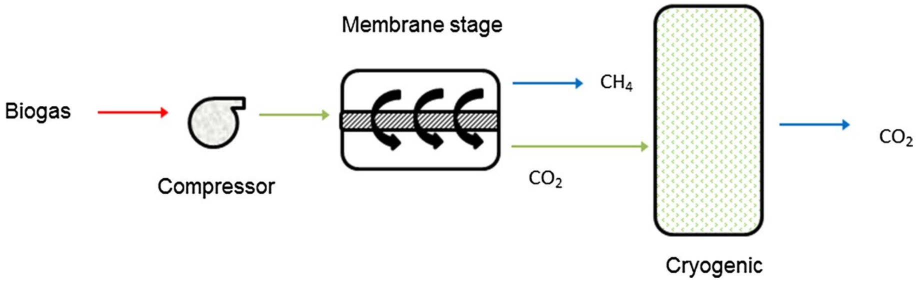


Fig. 8 Process diagram for cryogenic membrane biogas upgrading. Adapted from Maqsood et al. (2017)

upgrading, Scholz et al. (2013) reported that for the hybrid processes where high CH_4 recoveries are obtained, the cryogenic process has the lowest investment costs. Another process configuration was proposed by Song et al. (2017b) to optimize this hybrid process. They achieved a reduction of 1.7 MJ/kg CO_2 and a 54.4% of capital cost saving, and the proposed that operational cost can be reduced by 39.3–43–3%.

Cryogenic hydrate processes

Other potential future configurations for biogas upgrading hybrid cryogenic systems are cryogenic hydrate processes, previously tested for natural gas purifying. The merging between these two technologies born as the necessary conditions for its proper working are low temperature and high pressure. An approximate diagram of this technique is given in Fig. 9. For a proper performance of the process, firstly a cryogenic stage at − 55 °C is carried out where the CO_2 concentration decreases dramatically. Later in a second stage, the remaining CO_2 is captured by a hydrate phase which is accomplished at about 1 °C (Surovtseva et al. 2011). A CO_2 purity of 99% can be achieved by means of this process. Furthermore, this process allows removing impurities of biogas in the first stage (Sreenivasulu et al. 2015).

Cryogenic absorption processes

As competence for MEA as traditional and reference solvent for biogas upgrading, cryogenic absorption processes based on NH_3 can become a potential candidate for impurities removal of biogas, as shown in Fig. 10. The main reasons of this possible successful process are the low price, commercial availability, and low energy regeneration (Song et al. 2018). Another advantage of this low-temperature process is that NH_3 has been typically tested under temperature conditions of about 0–10 °C, which invites to think that good results will be obtained at lower temperatures. Nevertheless, further studies must be carried out to confirm the viability of cryogenic absorption hybrids processes, where the key point to confirm the validity of this process is the relationship between operating temperature and economic balance (Valenti et al. 2012).

Conclusion

In this paper, a brief analysis of current status and the development of cryogenic technologies and hybrid cryogenic technologies for biogas upgrading are reviewed. Both recognized technologies that have been applied for biogas

Fig. 9 Process diagram for cryogenic hydrate natural gas upgrading. Adapted from Hart and Gnanendran (2009)

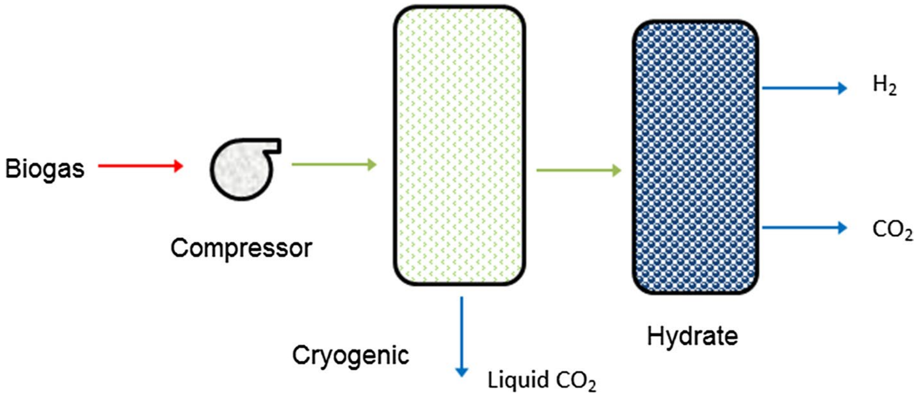
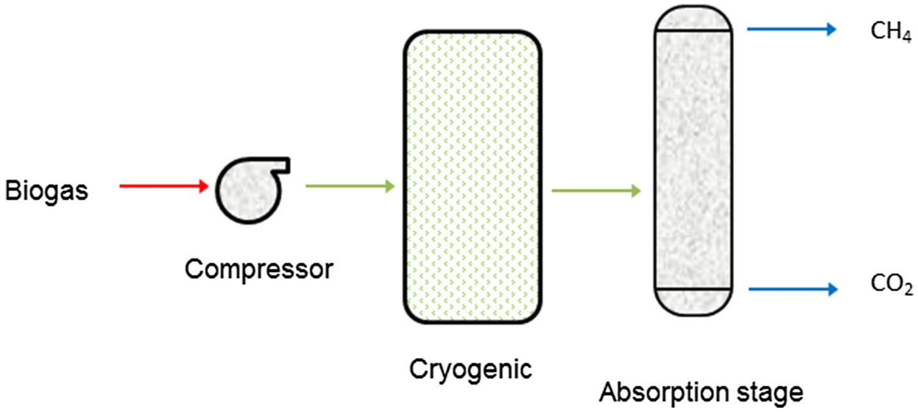


Fig. 10 Process diagram for cryogenic absorption natural gas upgrading. Adapted from Song et al. (2018)



upgrading and technologies which potentially could be attractive for this end are included. First, the three main technologies and configurations for cryogenic technologies are presented, analyzing in a subsequence section the main advantages and disadvantages of these techniques. Finally, hybrid systems which are considered for future potential solutions for cryogenic technologies handicaps are explained and divided into four different groups.

Regarding the standalone cryogenic technologies, it is a consensual among the belonging to the scientific community that the main advantage presented by this technique is the high product purity obtained. Among other advantages, the avoiding of solvents or sorbents and the compressed final product are highlighted. Nevertheless, the main handicap of this technology is its attractiveness only when low-cost energy sources surround the facilities, which make the overall more competitive in comparison with other biogas upgrading technologies. Against the problems presented by cryogenic techniques, cryogenic hybrid systems seem to be a solution in terms of energy penalty and installation investment. Presently, cryogenic membrane process has been the most intensely combination studied.

Future works should be led to further optimization of cryogenic upgrading processes as well as the development of new commercial configurations with less energy consumption. Concerning cryogenic hybrid systems, a long way should be covered to launch breaker commercial configurations into the market. However, the hopes in cryogenic membrane process are elevated and researchers from all over the world are focusing their efforts in this field.

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