

Journal of Soil Sciences and Agricultural Engineering

Journal homepage & Available online at: www.jssae.journals.ekb.eg

Analyzing Biogas Purification Using Low-Temperature Distillation For CO₂ Capture: Exergetic And Economic Evaluation

Dasith Ekanayaka^{1,2}; Tatiana Morosuk² and A. Jado^{3*}

¹ Faculty of Technology, Sabaragamuwa University of Sri Lanka, P.O. Box 02, Belihuloya, 70140, Sri Lanka.

² Institute for Energy Engineering, Technical University of Berlin, Berlin 10587, Germany.

³ Mansoura University, Department of Agricultural Engineering, Mansoura 35516, Egypt.



Cross Mark

ABSTRACT

As the world moves towards significant contributions to alternative energy sources, biogas and biomethane will play a significant role in the future. The biogas upgrading process to produce higher-purity biomethane while producing carbon dioxide as a by-product will find several applications. A low-temperature double-distillation method was developed and analyzed. Three biogas feed systems are discussed with typical biogas methane concentrations of 50%-mol, 60%-mol, and 75%-mol. The nitrogen refrigeration cycle provides the purification process at low temperatures. The design, simulation, sensitivity analysis were conducted using Aspen Plus software; energetic, exergetic, and economic analyses were performed accordingly, as well as improvement options suggested. The three systems with 1000 kmol/hr feed were designed to achieve 98.5%-mol methane purity while producing a higher-purity carbon dioxide stream. The simulations were conducted up to the level of a carbon dioxide-frozen-free environment. With methane content increasing from 50%-mol to 75%-mol in the biogas feed, the following can be achieved: reduction in the total power consumption from 22.97 MW to 20.77 MW and the specific energy consumption from 10.3 MJ/kgCH₄ to 6.21 MJ/kgCH₄; an increase in overall exergetic efficiency by 3%-point, and reduction in the total revenue requirements for the overall system (i.e., investment, operation and maintenance expenses, and fuel costs) from 259.68 million USD to 239.13 million USD.

Keywords: Biogas upgrading, low-temperature distillation, energy analysis, exergy analysis, economic analysis

INTRODUCTION

The world is moving towards renewable and sustainable energy sources due to significant concerns, such as global warming due to greenhouse gas emissions and the depletion of fossil fuel sources. According to the recently released "World Energy Outlook 2024" (International Energy Agency, 2024), fossil fuels accounted for only 60% of the global electricity supply last year. Biogas is a local energy source and can be upgraded, i.e., converted to biomethane as an alternative to natural gas. Among other low-emissions gases, the market of biogas and biomethane will grow, and by 2050, are expected to reach the following values: nearly 400 billion cubic meters equivalent (STEPS - Stated Policies Scenario) or 1.4 trillion cubic meters, which is 60% of total gas demand (NZE - Net Zero Emissions by 2050 Scenario). In the near future (by 2035), the combined demand for biogas and biomethane will double compared to 2023, as stated in the Stated Policies Scenario. The cost difference between natural gas and biomethane is around USD 10/MBtu (an average data that can be used as a reference for gas-consuming regions). The announced Pledges Scenario (APS) demonstrates the reduction of the difference to USD 4/MBtu because of the CO₂ pricing, national policy support, and market growth.

Biogas is an energy source for numerous applications due to its higher energy-dense methane components. Electricity generation, heat generation, vehicle fuel, and application for fuel cell operation are some practical uses of biogas (Sun *et al.*, 2015). The type of energy-related application depends on the composition of the raw biogas

mixture. Biogas composition depends on the biomass material and the operating conditions of the anaerobic digestion process. Carbohydrates, proteins, and fats are the three primary digestible raw substrates converted into biogas due to the biological treatment process in special operating conditions. Methane (CH₄) and carbon dioxide (CO₂) are the most significant components in biogas; among impurities are water vapor, hydrogen sulfide, nitrogen, dust particles, ammonia, and hydrogen (Christensen, 2010). The methane composition can vary from 40%-mol to 75%-mol, the carbon dioxide from 30%-mol to 47%-mol, and the lower heating value, correspondingly, can vary between 16 MJ/m³ and 28 MJ/m³ (Rasi *et al.*, 2011). Bioreactors with pure fat biomass substrates can produce a very high biogas yield of 1000-1250 l/kg of dry matter with a medium range methane content of 68-73%-vol, biomass substrate with pure protein provided the highest methane content of 70-75%-vol, but the lower gas yield of 600-700 l/kg of dry matter, the lowest methane content of 50-55%-vol in final biogas mixture is acquired with the feed of pure carbohydrate substrate (Christensen, 2010).

Biogas can be purified, upgraded, and separated into pure components. Impurities must be purified from the biogas mixture. Mainly to enhance the biogas methane content and to use separated carbon dioxide as a by-product. Biogas must be upgraded to at least 97%-mol methane to be suitable as a vehicle fuel or mixed with natural gas streams. Several techniques are used for biogas upgrading: pressure swing adsorption (PSA), water scrubbing, organic physical scrubbing, chemical scrubbing, biofiltration, membrane

* Corresponding author.

E-mail address: Jado@mans.edu.eg

DOI: 10.21608/jssae.2025.389083.1292

separation, and cryogenic upgrading. Considering the industrial-scale available biogas upgrading techniques, the PSA, water scrubbing, and physical scrubbing techniques can achieve more than 96%-mol methane content. The chemical scrubbing method gains the highest methane content of 99%-mol. Each technique has advantages and disadvantages based on energy consumption, purification and recovery capability, post-treatment and regeneration requirements, operational cost, and operation conditions (Burchell & Rogers, 2000; Kadam & Panwar, 2017).

One of the earlier publications in which biogas purification using cryogenic packed-bed technology was proposed was the paper by Tuinier and Van Sint Annaland (2012). The authors compared the developed technology to conventional vacuum pressure swing adsorption. The new technology requires a 22% lower energy consumption (2.9 MJ/kg_{CH₄} vs 3.7 MJ/kg_{CH₄}), although low temperatures around -150°C are required.

The cryogenic (better to use the term "low-temperature refrigeration") biogas upgrading system can be designed to have a higher methane purity (> 98%-mol) with lower methane loss and higher CO₂ recovery (Yousef *et al.*, 2018). Liquid CO₂ and liquid CH₄ can be produced from this technique at high pressure and transported directly into storage as valuable products. Even though the cryogenic biogas upgrading method provides numerous advantages over the other techniques, it is still the least common technique used on the industrial scale (Sun *et al.*, 2015). The technology is still under development because this method operates under low temperatures and higher pressure. Economic and environmental considerations are other essential factors in the evaluation of upgrading cryogenic biogas. Several researchers have conducted analyses based on the cost-effective cryogenic upgrade of biogas taken from anaerobic digestion reactors or landfill gas (Fan *et al.*, 2008). On the other hand, CO₂ solidification is another significant factor that needs to be considered in the cryogenic biogas upgrade. The solid CO₂ can damage the operational units, and special attention is needed on processes like distillation columns (Yousef *et al.*, 2018). The purity and the quality of the final products of CH₄ and CO₂ streams are limited by CO₂ solidification in low-temperature techniques (ZareNezhad, 2006).

A comprehensive evaluation of cryogenic technologies for biogas upgrading using energy, economic, and environmental criteria has been reported by Naquash *et al.* (2022). This study demonstrated that distillation-based biogas upgrading is more energy efficient, consuming 0.31–0.35 kWh/ Nm³, and strictly depends on the process configuration.

Exergy analysis has been applied in several studies. The approach "exergy in / exergy out" was selected; therefore, the exergy efficiency values are unrealistic for components and the overall systems. For example, in Naeiji *et al.* (2022), exergy analysis was applied and extended to thermoeconomic evaluation. The total exergy efficiency of cryogenic separation and chemical scrubbing was reported as 85% and 84%. Vilardi *et al.* (2020) reported an exergy efficiency of around 90% for three biogas upgrading processes. Nevertheless, all studies reported that cryogenic methods

could offer higher exergy efficiency under similar operation conditions.

The authors' research focuses on delivering a high-purity liquid methane stream with 98.5%-mol CH₄ that is feasible for commercial applications and mixed and used with natural gas. The proposed system was designed and simulated using Aspen Plus v.12 software. The three biogas feed streams with 1000 kmol/hr feed flow rates and 50%-mol, 60%-mol, and 75%-mol CH₄ content were simulated. Biogas feed is assumed to be pretreated and purified from contaminants, but a small amount of water exists. The flash drum unit was used to separate the remaining water from the feed stream, and two distillation column separation techniques were used to purify biogas. A refrigeration system with nitrogen as the working fluid (refrigerant) is designed and used to provide the refrigeration effect for the operation. Furthermore, CO₂ can be used as a by-product of the biogas purification process. Exergy analysis (in terms of "exergy of fuel / exergy of product") was conducted under different feed conditions to determine the components with the highest irreversibility in the system cause and, thus, which have a prominent improvement opportunity. Moreover, an economic analysis was performed to estimate the economic feasibility of the proposed systems.

This paper followed a comprehensive study on the undermentioned factors, optimized design to avoid the risk of CO₂ freezing while delivering a higher CH₄ purity product stream, separation of CH₄ and CO₂ as valuable liquid product streams, examine the impact of feed biogas compositions on operation conditions and product purity, and finally the detailed analysis of the proposed system results based on product specifications, energy and exergy evaluations, and economic considerations.

State-of-the-art

Biogas production and upgrading

Biogas is produced from the biological conversion of organic substances in an anaerobic environment, and this overall process is called anaerobic digestion. The overall process occurred without an outwardly supplied electron acceptor, such as oxygen, sulfate, or nitrate. The anaerobic digestion process or landfilled gas generation units are used to generate and collect biogas. The composition of the produced biogas mixture depends on the feedstock material type and the oxidation condition of the anaerobic digestion process (Christensen, 2010). CH₄ and CO₂ are the major components in the biogas mixture. Moreover, the biogas mixture contained water, hydrogen sulfide, ammonia, volatile components, and other impurities. Extracting CH₄ from biogas is the primary purpose of producing biogas.

Biomethane and carbon dioxide utilization

The biogas upgrading process has become vital since it produces highly concentrated CH₄ and CO₂ separately. The most substantial advantage is the capability of consuming the produced biomethane as a renewable energy source and replacing non-renewable natural gas. The consumption of biomethane as a product assisted in reducing GHG emissions and decarbonizing processes (Abd *et al.*, 2023; Burchell & Rogers, 2000). Furthermore, the low-temperature biogas upgrading process produced the biomethane and CO₂ in the liquid state, leading to troubleless storage and transportation compared to gaseous conditions.

Biomethane is the primary output product from the biogas upgrading process, which has a higher energy density of around 36 MJ/m³ (Christensen, 2010). Biogas can be used directly for direct water heating, cooking, or simple combustion applications. For example, the methane concentration needed to be over 97%-mol for introduction into the natural gas grid, and the concentration around 90-97%-mol range can be used as a transportation fuel. The pressure swing absorption biogas upgrading technique was mainly used to make a higher purity methane concentrated stream to mix with the natural gas stream (Abd *et al.*, 2023; Burchell & Rogers, 2000; Christensen, 2010). The low-temperature biogas upgrading technique discussed in this research is another ideal method to gain a higher methane purity stream to mix with the natural gas grid or use as a transportation fuel.

The use of natural gas can reduce carbon dioxide emissions by 30% compared to the emission from petroleum-fueled vehicles, and biomethane can reduce that value down to 90%. Natural gas as a fuel is stored, transported, and consumed worldwide with well-established infrastructure. Biomethane can also be introduced into this infrastructure without significant effort and large investments. Moreover, another notable benefit of biomethane is that it is possible to use in currently available conventional gas engines with a slight adjustment (Pavičić *et al.*, 2022). Ultimately, the most critical concern of energy sources for electricity production is the availability and balancing of the demanded power; henceforth, stored biomethane will be an ideal solution in the future as a renewable and sustainable energy source.

The CO₂ can be stored and consumed as a product for several applications. Such as chemical industries for the synthesis of chemicals, including polymers, fuel, and microalgae products, making materials for concrete buildings, producing biochar, and most significantly used for enhanced oil recovery (EOR) applications (Hepburn *et al.*, 2019). The EOR technology enhanced the oil recovery capacity significantly, and the CO₂ needed to be injected into the oil well at the liquid or supercritical stage to gain efficient oil recovery. Since CO₂ is a solvable component of oil, and in the EOR process, oil viscosity is reduced after CO₂ injection, which makes oil more transferable. The low-temperature biogas upgrading process provided CO₂ as a product stream in the liquid stage, and it is an ideal condition for the EOR. Moreover, there is a huge potential to store the separated CO₂ permanently in underground geological systems; also, the EOR provides additional benefits besides using CO₂ as a product, which assists in reducing global warming by reducing GHG emissions. Furthermore, compared with other EOR techniques, the use of CO₂ in the EOR process is cheaper, occupies little space, and is applicable for various kinds of oil reservoirs, including deeper heavy oils (Hepburn *et al.*, 2019).

Thermodynamic framework

Carbon dioxide and methane separation

The separation of CH₄ from the CO₂/CH₄ mixture in a distillation process is the most critical purification step in biogas upgrading. The selection of thermodynamic parameters, such as pressure and temperature, for the separation process highly depends on the CO₂ freezing conditions.

The purity of the final product directly depends on the carbon dioxide freezing conditions, especially in distillation unit operation (Yousef *et al.*, 2018). The operation condition can be adjusted to better separation while avoiding carbon

dioxide freezing. The freezing point of CO₂ in a biogas mixture varies with the operating pressure in a low-temperature biogas upgrading system. The freezing risk drops at elevated pressure (ZareNezhad, 2006). Moreover, additional components like heavy hydrocarbons can be used to depress the CO₂ freezing point in a biogas mixture. Hydrocarbon components like pentane and hexane are suited for this freeze-point depression process. Additional feed streams must be introduced to the system, such as into the distillation operation. This method will allow the system to operate without solid form at a lower temperature than the freezing point (Berstad *et al.*, 2012). However, the most suitable way to operate the system is without using any additional chemicals into the system.

Nitrogen as the working fluid

Nitrogen is used as the working fluid to generate the refrigeration capacity. The boiling point of nitrogen is -195.8 °C, the freezing point is -210 °C at 1 bar, and the critical point is -146.9 °C at 33.958 bar pressure. Furthermore, the boiling point increases with the pressure, and the freezing point is altered slightly. The operating temperature range of the refrigeration cycle is between 60 °C to -180 °C; henceforth, nitrogen is the most appropriate working fluid for the low-temperature refrigeration process. Moreover, nitrogen has several advantages as a refrigerant, such as being highly available in the market, being cheap compared to other working fluids, and having inert properties. Furthermore, due to the lower boiling point and higher refrigeration capacity, nitrogen is a better practical refrigerant for cryogenic and low-temperature operations (Beteta & Ivanova, 2015).

Thermodynamic framework

The biogas upgrading means the separation of CO₂ fraction from the biogas to gain higher methane content in the final mixture. Before upgrading, the pre-purification step is necessary to remove minor impurities such as dust, water vapor, hydrogen sulfide, oxygen, nitrogen, siloxanes, ammonia, and particles (Burchell & Rogers, 2000). The minor impurities are up to 1.5-2%-mol, and the separation of CO₂ from the biogas mixture is the fundamental separation step followed to upgrade the biogas to gain higher biomethane content (Christensen, 2010). Currently, industries use several chemical, mechanical, and biological techniques to upgrade biogas. The technique used depends on the purity requirement of biogas, availability of resources, energy requirements, and economic viability of the operation (Adnan *et al.*, 2019). Water scrubbing, Pressure swing adsorption, amine absorption, membrane separation, and biofiltration are widely used techniques on the current industrial scale. In the EU, water scrubbing is the most commonly used technique, and it follows PSA and amine absorption techniques (Kadam & Panwar, 2017). However, low-temperature biogas upgrading is an emerging technique that most researchers and industries are pursuing to apply biogas upgrading (Yousef *et al.*, 2018). Water scrubbing is the lowest energy-consuming method based on comparing currently used techniques.

Nevertheless, the amine absorption chemical treatment method provides the highest purification capability, i.e., >98%-mol methane purity in the final biogas mixture. This method can be operated at atmospheric pressure conditions; no additional pressurization is required. However, these techniques have high operational and maintenance costs and require the use of chemicals and regeneration operations, which can cause environmental concerns (Kadam & Panwar, 2017).

Compared to the commonly used purification methods, the low-temperature biogas purification techniques have several benefits. The key benefit is the capability of recovering higher CH₄ content from a biogas feed with minimum methane loss. Low-temperature distillation can result in up to 99.7%-mol methane recovery (Wong & Bioletti, 2002). Another significant advantage is the possibility of producing CO₂ with higher purity as a by-product of the purification process. However, this low-temperature biogas purification technique is still more energy-intensive and expensive than traditional upgrading methods. Also, compared with other commonly used techniques, the operating conditions needed more pressure to maintain optimum operation. However, an additional advantage is a technique that only needs electricity as the driving fuel for the operation.

Evaluation methods

Energy analysis

The total power consumption for the design was calculated as described in Equations (1) to (3), including specific values to compare with other biogas upgrading techniques reported in the literature:

Total Power consumption (MW) =

Sum of power consumption in all compressors – Power generation in the expander (1)

Specific power consumption per kg of biogas feed (MJ/kg) =

Total power consumption (MW) / Biogas feed (kg/s) (2)

Specific power consumption per kg of methane (MJ/kg_{CH4}) =

Total power consumption (MW) / Methane in the product stream (kg/s) (3)

Exergy analysis

The quality of energy is a vital factor for analyzing energy-conversion systems and energy-intensive chemical systems. The energy describes only the quantitative part, while the exergy analysis describes the quantity and quality of energy. The overall exergy of a system ($\dot{E}_{sys}$) is expressed using four contributors, chemical exergy ($\dot{E}^{CH}$), physical exergy ($\dot{E}^{PH}$), potential exergy ($\dot{E}^{PT}$), and kinetic exergy ($\dot{E}^{KE}$) (Bejan *et al.*, 1996)

$$\dot{E}_{sys} = \dot{E}^{CH} + \dot{E}^{PH} + \dot{E}^{PT} + \dot{E}^{KE} \quad (4)$$

In most applications, the potential and kinetic exergy effects are neglected. Therefore,

$$\dot{E}_{sys} = \dot{E}^{CH} + \dot{E}^{PH} \quad (5)$$

The physical exergy of a system at any given state j is expressed as,

$$\dot{E}_j^{PH} = (\dot{H}_j - \dot{H}_0) - T_0 (\dot{S}_j - \dot{S}_0) \quad (6)$$

where $\dot{H}$, $\dot{S}$, and T represent enthalpy rate, entropy rate, and temperature, respectively, and the property at "restricted dead state" (subscript 0).

Further, for some applications, the physical exergy of a stream is recommended to be split into two parts: the thermal exergy ($\dot{E}^T$) as the temperature-related part and the mechanical exergy ($\dot{E}^M$) as the pressure-related part

$$\dot{E}_j^{PH} = [(\dot{H}_j - \dot{H}_{j,A}) - T_0 (\dot{S}_j - \dot{S}_{j,A})]_{P=\text{constant}} + [(\dot{H}_{j,A} - \dot{H}_0) - T_0 (\dot{S}_{j,A} - \dot{S}_0)]_{T=\text{constant}} \quad (7)$$

Where the thermodynamic properties of state A are defined for each j material stream at operation pressure (p) and environmental temperature T_0

The chemical exergy of an ideal mixture of N ideal gases and the solution of liquids are represented in Eqs. (8) and (9), respectively. Here, e_i^{CH} is the standard molar chemical

exergy value of the i -th substance, x_i its molar fraction, and γ_i its activity coefficient (for the solution of liquids)

$$e^{CH}_{Ideal\ gas\ Mixture} = \sum_{i=1}^N x_i e_i^{CH} + \bar{R} T_0 \sum_{i=1}^N x_i \ln(x_i) \quad (8)$$

$$e^{CH}_{Solution} = \sum_{i=1}^N x_i e_i^{CH} + \bar{R} T_0 \sum_{i=1}^N x_i \ln(\gamma_i x_i) \quad (9)$$

The exergetic balance for an overall system (subscript 'tot') and the k -th component are expressed as follows:

$$\dot{E}_{F,tot} = \dot{E}_{P,tot} + \dot{E}_{D,tot} + \dot{E}_{L,tot} \quad (10)$$

$$\dot{E}_{F,k} = \dot{E}_{P,k} + \dot{E}_{D,k} \quad (11)$$

The exergy destruction value ($\dot{E}_D$) represents the irreversibility of a system, which occurred as a result of chemical reaction, mixing, friction, and heat transfer through finite temperature change. In addition to exergy destruction, the exergy loss term $\dot{E}_L$ describes the exergy transmission from the system to the environment. Considering the overall exergy balance equation, the $\dot{E}_P$, and $\dot{E}_F$ represent the exergy of product and exergy of fuel, respectively. The exergy of the product $\dot{E}_P$, is the desired product that can be obtained from a system/component. The exergy of fuel $\dot{E}_F$, is the exergy required to produce the exergy product (Bejan *et al.*, 1996). The exergetic efficiency is the evaluation variable:

$$\epsilon_{tot} = \dot{E}_{P,tot} / \dot{E}_{F,tot} \quad (12)$$

$$\epsilon_k = \dot{E}_{P,k} / \dot{E}_{F,k} \quad (13)$$

Exergetic efficiency is applicable to all system components except dissipative components. In addition to exergetic efficiency, the exergy destruction ratio is used for the exergy analysis

$$y_{D,k} = \dot{E}_{D,k} / \dot{E}_{D,tot} \quad (14)$$

Economic analysis

The estimation of the total capital investment was initiated by calculating the Purchase Equipment Cost (PEC). The PEC can be calculated using different techniques. The bare module cost C_B needs to be calculated, which is the deciding factor for PEC. The C_B is the cost of the base condition of the equipment, including commonly used materials, pressure, and temperature. Finally, with the help of the correction factors, this value will be adjusted (Bejan *et al.*, 1996; Cavin *et al.*, 2004).

$$C_M = C_B f_T f_P f_m f_B \quad (15)$$

where the meaning of each term can be found in the nomenclature.

The PEC of each component C_{cal} is usually calculated for previous years mentioned in the reference documents. Hence, the Chemical Engineering Plant Cost Index (CEPCI) should be used to adjust the PEC of the component for the reference year as C_{ref} .

$$C_{ref} = C_{calc} \times index_{ref} \times index_{cal} \quad (16)$$

For this research, all costs were adjusted to the year 2023.

The Fixed capital investment (FCI) was calculated as the summation of the total module costs of the equipment, plus service construction, service facilities, architectural work, and contingencies cost (Bejan *et al.*, 1996).

The Total Revenue Requirement (TRR) method was used in this thesis study to evaluate the cost of the evaluated system. The TRR was calculated yearly, considering the carrying costs and expenditures (Bejan *et al.*, 1996). Such as the carrying charges of investment returns and capital recovery, as well as O&M and fuel costs expenditures. All cost terms are not uniform throughout the economic life span of the project, and henceforth, the cost values were leveled to gain a constant annual rate. Levelized TRR is expressed as

$$TRR_L = CC_L + FCL + O\&M_L \quad (17)$$

After evaluating the equipment cost, the total capital investment (TCI) needed to be evaluated. The TCI was calculated by adding the FCI as the one-term investment cost plus the interest. The FCI included the direct and indirect costs of construction, design, and installation of the plant and other costs associated with plant site arrangement. Then the Levelized carrying charges CCL are calculated using the Capital recovery factor CRF. The calculation of CRF is expressed in Eq. (20)

$$\text{Interest for the investment} = FCI (1 + i_{eff})^{-1} \quad (18)$$

$$CC_L = TCI.CRF \quad (19)$$

$$CRF = i_{eff}(1+i_{eff})^n/(1+i_{eff})^{n+1} \quad (20)$$

The n is the plant's economic life, and i_{eff} is the effective annual interest rate. The expenditures of $O\&M_L$ and FC_L are calculated using the Constant Escalation Levelization Factor, CELF. The general CELF_{general} and fuel CELF_{fuel} values are used to evaluate $O\&M_L$ and FCL , respectively. The CELF is $CELF = K_{FC}(1-K_{FC})/(1-K_{FC}) \times CRF$, with $K_{FC} = 1+r$ $1+i_{eff}$ (21)

The r characterizes the average general inflation rate in $O\&M$ calculation, and r denotes the average annual escalation rate of fuel, which is the escalation rate of electricity in this Thesis study. Then, the $O\&M$ is calculated as

$$O\&M = 0.4 \times FCI \times CELF_{general} \quad (22)$$

The next step is to calculate the levelized fuel cost

Electricity Cost (\$/year) =

$$\text{Electricity Consumption (MWh/year)} \times \text{Electricity Price (\$/MWh)} \quad (23)$$

$$\text{Levelized annual electricity cost} = \text{Electricity cost} \times$$

$$CEFL_{fuel} \quad (24)$$

Finally, the levelized TRR can be calculated.

Simulation and evaluation

Property Method Selection

Based on the component and operation conditions, the Peng-Robinson (PR) equations of state were used for pure working fluids, and Soave-Redlich-Kwong (SRK) equations of state for mixtures. The CO_2/CH_4 binary mixture was simulated using Aspen Plus and compared with the ideal situation and NIST experiment results.

Biogas Upgrading Process Design

A series of processing units were designed in Aspen Plus to upgrade the biogas stream, and simulation was performed for three different systems. The feed stream was assumed to be pre-purified for the simulation, and CH_4 , CO_2 , and H_2O (vapor) were in the input feed flow. The water in the biogas feed needs to be removed before the low-temperature

distillation process. The Flash Drum was used to perform the water separation activity in the design. The vapor-liquid flash separation was designed at low temperatures and elevated pressure, which caused water vapor to liquefy and be extracted from the bottom of the flash drum. The CO_2 and CH_4 in the biogas stream remain at the vapor phase and are taken out from the top stream at the flash drum unit. The sensitive analysis was performed on the Flash drum unit with different temperature and pressure values to identify the optimum operating condition for the flash separation. The sensitive analysis was performed down to $-60^\circ C$ minimum temperature and up to 8 bar maximum pressure. The CO_2 stream needed to be in the vapor while all water in the feed was removed. The flash drum sensitive analysis value range was selected based on the CO_2 liquified conditions.

The required refrigeration capacity for the upgrading process can be reduced by precooling the biogas feed before entering the separation processes. The air streams, refrigeration cycle, and the low-temperature liquid CO_2 product stream, taken from the distillation column bottom streams, were used to precool the biogas stream, as shown in Figure 1. The countercurrent type of heat exchanger was used for the cooling load recovery process in the Aspen Plus simulation.

Figure 1 shows two series of distillation column operations that perform the critical separation process in the biogas upgrading process. The two series of distillation column operations are more energy efficient than one distillation column operation in achieving higher product purity (Hashemi *et al.*, 2019; Shafeeq *et al.*, 2010; Yousef *et al.*, 2018).

A set of parameters for the distillation process simulation decides the operation condition and capability of product purification, such as reflux ratio, distillate to feed ratio, number of stages, feed stage, feed temperature, heating, condensing duty, pressure gradient, temperature gradient, and so on. These parameters can be changed to gain the required output and achieve an efficient operation (Yousef *et al.*, 2018). The parameters such as the number of stages, Feed temperature, pressure gradient, and temperature gradient are decided based on the CO_2 frozen-free conditions. The feed input stage is determined by performing a sensitivity analysis by considering the targeted methane molar fraction in top streams in distillation columns 01 and 02.

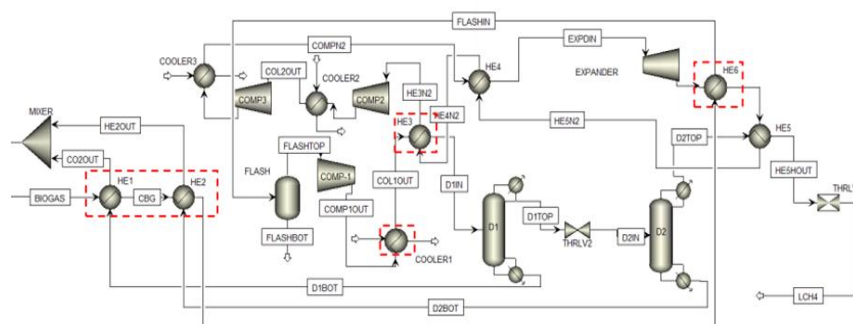


Fig.1. Biogas Upgrading Simulation (from Aspen Plus)

Biogas upgrading without CO_2 freezing

The most significant and critical concern in the low-temperature biogas upgrading process with a multi-distillation process is freezing the CO_2 within the process. All three systems need to be upgraded in the biogas without freezing the

CO_2 at any stage, especially the operation of the distillation columns. The pressure, temperature of each column, and parameters such as distillate-to-feed ratio and reflux ratio were decided based on the CO_2 freezing point at each stage in the distillation process.

The freezing point of CO₂ concerning each pressure and temperature of each tray in both columns was discovered based on references (Maqsood *et al.*, 2014; ZareNezhad, 2006; Zhang *et al.*, 2011).

The carbon dioxide and methane liquefaction

The bottom streams from the distillation columns, which were in the liquid phase, were combined and taken as the highly concentrated CO₂ product output stream, as shown in Figure 1. Additionally, the bottom streams from the distillation process were used to cool down the input biogas feed. Consequently, the temperature of the produced Carbon dioxide output stream was increased and reached the vapor-liquid stage. The mass liquid fraction at output Carbon dioxide product stream mixtures were 99.24%-mol, 96.23%-mol, and 92.76%-mol in systems 01, 02, and 03, respectively, with 50%-mol, 60%-mol, and 75%-mol in CH₄ feed inputs. The top stream of distillation column 02 was taken out as the highly concentrated biomethane product stream. The purity level was set up to reach 98.5%-mol in all three design simulations. The stream was at the vapor stage after the distillation process, and as shown in Figure 1, a heat exchanger was used to cool down the stream and produce liquid-phase high-concentrated biomethane product output. The nitrogen refrigeration cycle was used for the heat-exchanging operation, reducing the biomethane product's output stream temperature to -155 °C. Then, a throttling process was used to adjust the pressure of the produced biomethane stream to the optimum level.

The actual pressure level to store liquefied natural gas is 35 bar, corresponding to reduced storage vessel cost, creating a higher safety condition and less hazardous risk (Burchell & Rogers, 2000).

The nitrogen refrigeration cycle

A double-stage compressed, single-stage expanded, with nitrogen as the working fluid, a refrigeration cycle was used to generate the required refrigeration capacity for the low-temperature biogas upgrading process. The refrigeration cycle is shown in Figure 1.

Energy analysis

The power consumption was analyzed, and the results were compared for the three biogas feed conditions. The total

biogas feed rate was maintained at 1000 kmol/hr for each simulation, but the total mass flow rate varies with the concentration change in each design. 8.33 kg/s, 7.56 kg/s and 6.39 kg/s were the feed flow rate in 50%-mol, 60%-mol and 75%-mol methane feed systems, respectively.

The energy demand was compared to five different techniques: pressure swing adsorption, which uses electrical energy, especially for gas compression, wet scrubbing, which uses electrical energy (Paolini *et al.*, 2021), physical absorption, which uses electrical energy to feed the material and it depends on the solvent capacity, also the thermal energy consumption. Chemical absorption requires electrical energy; the stripping process requires thermal energy; membrane separation also uses electrical energy for the upgrading process. Since the physical and chemical absorption techniques used energy in electrical and heat forms, an assumption for the analysis is that 1 kWh of electrical energy equals 4 kWh of heat energy. The unit for the specific energy consumption for all techniques by kWh/Nm³ of purified stream volume.

Exergy analysis

The definitions for the exergy of fuel, exergy of the product, and exergy loss for the overall system are defined as follows,

$$\text{Exergy of Fuel } (\dot{E}_{F,tot}) =$$

$$\text{Exergy of biogas input stream into the system} + \text{Exergy input for the compressors} - \text{Exergy output from expander unit} \quad (25)$$

$$\text{Exergy of Product } (\dot{E}_{P,tot}) =$$

$$\text{Exergy of liquid methane stream} + \text{Exergy of liquid carbon dioxide stream} \quad (26)$$

$$\text{Exergy loss } (\dot{E}_{L,tot}) =$$

$$\text{Exergy loss from flash bottom water removing stream} \quad (27)$$

Finally, from the exergy balance,

$$\text{Exergy destruction } (\dot{E}_{D,tot}) =$$

$$\text{Total Exergy of Fuels} - \text{Total Exergy of Product} - \text{Total Exergy loss} \quad (28)$$

Table 1 shows the definition of the $\dot{E}_{F,k}$ and $\dot{E}_{P,k}$ for all productive components and the definition of $\dot{E}_{D,k}$ for all dissipative components (Fig. 1).

Table 1. The definitions for the exergetic analysis

Component	Stream Temperature	Exergy of Fuel	Exergy of Product
HE1 (Heat Exchanger 1)	$T_{hot,in} \leq T_{amb}$	$\dot{E}_{cold,in} - \dot{E}_{cold,out}$	$\dot{E}_{hot,out} - \dot{E}_{hot,in}$
HE2 (Heat Exchanger 2)	$T_{hot,in} \leq T_{amb}$	$\dot{E}_{cold,in} - \dot{E}_{cold,out}$	$\dot{E}_{hot,out} - \dot{E}_{hot,in}$
HE3 (Heat Exchanger 3)	$T_{cold,out} \quad T_{hot,out} \leq T_{amb} \leq T_{hot,in}$	$\dot{E}_{cold,in} - \dot{E}_{cold,out} + \dot{E}_{hot,in} - \dot{E}_{hot,out}^M$	$\dot{E}_{hot,out}^T$
HE4 (Heat Exchanger 4)	$T_{cold,out} \quad T_{hot,out} \leq T_{amb} \leq T_{hot,in}$	$\dot{E}_{cold,in} - \dot{E}_{cold,out} + \dot{E}_{hot,in} - \dot{E}_{hot,out}^M$	$\dot{E}_{hot,out}^T$
HE5 (Heat Exchanger 5)	$T_{hot,in} \leq T_{amb}$	$\dot{E}_{cold,in} - \dot{E}_{cold,out}$	$\dot{E}_{hot,out} - \dot{E}_{hot,in}$
Flash	$T_{in}, T_{out} \leq T_{amb}$	$\dot{E}_{in} - \dot{E}_{out,loss}$	$\dot{E}_{product}$
Comp1 (Compressor 1)	$T_{in}, T_{out} \geq T_{amb}$	$\dot{W}_{CM}$	$\dot{E}_{in} - \dot{E}_{out}$
Comp2 (Compressor 2)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{W}_{CM}$	$\dot{E}_{in} - \dot{E}_{out}$
Comp3 (Compressor 3)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{W}_{CM}$	$\dot{E}_{in} - \dot{E}_{out}$
Cooler1 (Air Cooler 1)	$T_{cool,in} \geq T_{amb}$	$\dot{E}_{hot,in} - \dot{E}_{hot,out}$	$\dot{E}_{cold,out} - \dot{E}_{cold,in}$
Cooler2 (Air Cooler 2)	$T_{cool,in} \geq T_{amb}$	$\dot{E}_{hot,in} - \dot{E}_{hot,out}$	$\dot{E}_{cold,out} - \dot{E}_{cold,in}$
Cooler3 (Air Cooler 3)	$T_{cool,in} \geq T_{amb}$	$\dot{E}_{hot,in} - \dot{E}_{hot,out}$	$\dot{E}_{cold,out} - \dot{E}_{cold,in}$
THRLV1 (Throttling valve 1)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{E}_{in}^M - \dot{E}_{out}^M$	$\dot{E}_{out}^T - \dot{E}_{in}^T$
THRLV2 (Throttling valve 2)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{E}_{in}^M - \dot{E}_{out}^M$	$\dot{E}_{out}^T - \dot{E}_{in}^T$
D1 (Distillation column 1)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{m}_{in} (e^{T,in} - e^{T,bot}) + \dot{m}_{in} (e^{M,in} - e^{M,bot})$	$\dot{m}_{top} (e^{CH,top} - e^{CH,in}) + \dot{m}_{top} (e^{M,top} - e^{M,in}) + \dot{m}_{bot} (e^{CH,bot} - e^{CH,in}) + \dot{m}_{top} (e^{T,top} - e^{T,in})$
D2 (Distillation column 2)	$T_{in}, T_{out} \leq T_{amb}$	$\dot{m}_{in} (e^{T,in} - e^{T,bot}) + \dot{m}_{in} (e^{M,in} - e^{M,bot})$	$\dot{m}_{top} (e^{CH,top} - e^{CH,in}) + \dot{m}_{top} (e^{M,top} - e^{M,in}) + \dot{m}_{bot} (e^{CH,bot} - e^{CH,in}) + \dot{m}_{top} (e^{T,top} - e^{T,in})$
Expander	$T_{cool,in} \leq T_{amb}$	$\dot{E}_{in}^M - \dot{E}_{out}^M$	$\dot{W}_{EX} + \dot{E}_{in}^T - \dot{E}_{out}^T$
Mixer		Dissipative component $\dot{E}_D = \dot{E}_{in1} - \dot{E}_{in2} - \dot{E}_{out}$	

Economic analysis

PEC calculation

The following assumptions were made for the PEC calculation for the different types of equipment.

Heat Exchangers:

The cost estimation graph (Ulrich *et al.*, 2004) was used to estimate the cost. The required heat transfer area for all heat exchangers was directly obtained from Aspen Plus simulations. The heat transfer area is less than 1000 m² for each heat exchanger. Shell-in-tube type of heat exchangers were assumed. The nickel mix carbon steel is assumed to be used for heat exchangers operated below the environmental temperature. The carbon steel material was assumed for the heat exchangers, which operated above the environmental temperature.

Compressors: The cost estimation equations reported by Xu *et al.* (2014) were used to estimate the cost of Compressor 02 and Compressor 03, which are operated below the environmental temperature. Compressor 01 operated above environmental temperature; the data from (Ulrich *et al.*, 2004) were used. **Flash separator:** The data reported by Xu *et al.* (2014) were applied.

Distillation columns:

The distillation columns are the most vital unit in the biogas upgrading system. Since these units were operated below the environmental temperature conditions, the data reported by Xu *et al.* (2014) are more appropriate. The cross-check was done using the data by Ulrich *et al.* (2004), and the average value was taken for the analysis.

Expander:

The expander operates below the environmental temperature. Stainless steel is the main material that should be used for low-temperature applications (with the corresponding material factor for economic analysis).

Two calculation methods were used to estimate the cost: reported by Hamdy *et al.* (2019) and by Li (2011). The average result was used for the analysis.

Mixtures, valves, etc.: The PEC of these units was not calculated separately; 25% of the total purchased equipment was added.

PEC calculation

A detailed cost calculation was performed for the overall biogas upgrading process using the TRR method, and the economic parameters mentioned in Table 2 were applied as the assumption.

Table 2. Assumptions used for economic analysis

Parameters	Value	Unit
Plant Economic life	25	Years
Effective interest rate	2.17%	%/year
Average general inflation rate(nominal)	2.60%	%/year
Average nominal escalation rate of electricity	12.00%	%/year
Electricity price (2022)	0.35	1000 \$/MWh
Total annual time of system operation	6000	Hours/Year
CRF	5	%/year
O&MC	5	% of FCL
Construction, Sering Facilities, and architectural work	25	% of Module Cost
Contingencies	10	% of Module Cost

RESULTS AND DISCUSSION

Properties of the working fluids

The SRK method graph shape and behavior are closer to the NIST experiment results, it was selected as the property

method for the simulation. As shown in Figure 2, the SRK property method is compatible with a system with nonpolar or mildly polar mixtures like biogas.

The biogas contains hydrocarbons like methane and light gases like carbon dioxide and hydrogen sulfide.

Moreover, the SRK is optimal for a high-pressure system and provides good results for a wide temperature range.

Considering all these factors, the SRK property method was used for the low-temperature biogas upgrade simulation.

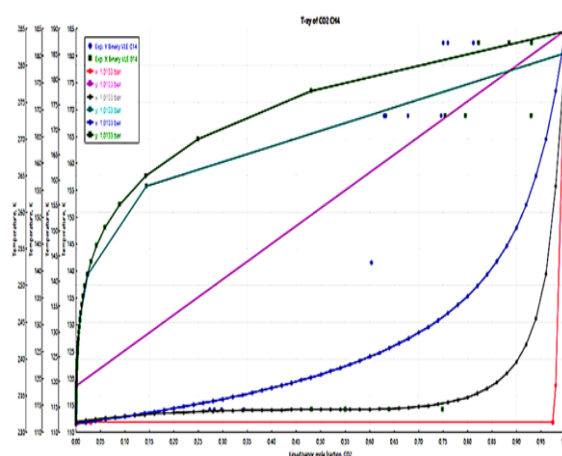


Fig.2. Carbon dioxide and Methane mixture binary data from Aspen Plus simulation (NIST experiment – Blue circles and Dark Green squares with y axis 1, PR – x (Red), y(Purple) with y axis 2. SRK – x(Brown), y(Light Green) with y axis 3. Ideal –x(Blue), y (Dark Green) with y axis 4

Design parameters and optimum operation condition

Flash Drum

The separation was increased with the temperature reducing and pressure increasing. However, according to the sensitivity analysis results shown in Figure 3, the maximum water removal can be achieved at around -35 °C at 8 bar pressure in the flash drum operation

Precooling and Distillation columns

The precooling conditions in HE1 and HE2 were based on the distillation column bottom stream temperature values. Based on the Aspen Plus simulation, the high methane content stream needed a lower heating duty than the low methane content biogas stream.

The total heating duty for the reboilers in both distillation column operations was 2.17 MW, 1.59 MW, and 1.15 MW from the higher CH₄ feed system to the lower methane feed system.

The low heating duty in higher methane feed makes a lower temperature level at the distillation bottom than the lower methane feed system distillation bottom, creating a low-temperature output stream from the bottom of the distillation column in the higher methane feed system.

Based on the distillation columns sensitivity analysis results shown in Figure 4, for column 01 and column 02 operation, with 91%-mol and 98.5%-mol methane molar concentration, respectively, the feed stage should be 5 in column 01 and stage 4 or 5 in column 2 operation.

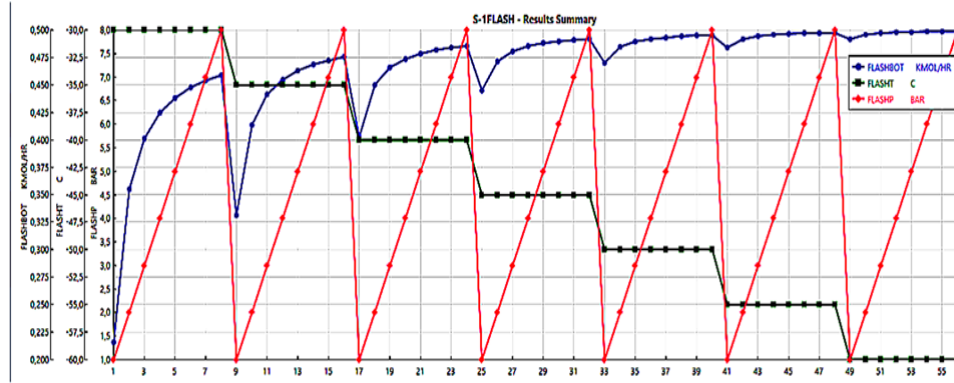


Fig.3. Sensitivity analysis results for the Flash drum unit Aspen plus simulation (Bottom water flow rate – Blue color graph with y axis 1. Flash temperature – Green color graph with y axis 2. Flash pressure – Red color graph with y axis 3), X axis is sensitivity analysis number

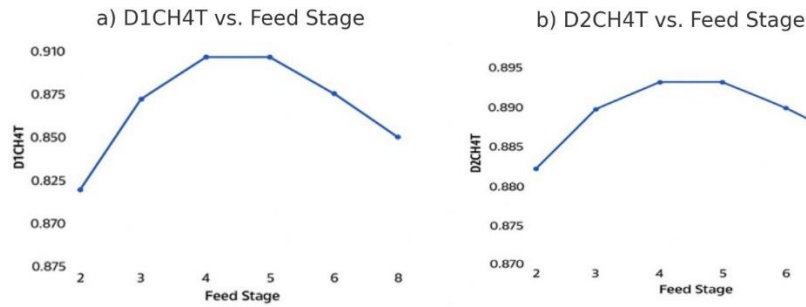


Fig.4. Feed Stage vs. Methane Purity in Distillation Columns (D1 & D2, 60% CH₄ Feed).

The results shown in Figure 5 demonstrate that vapor and liquid fractions temperatures did not cross the freezing CO₂. The safer side of operating the distillation process without CO₂ freezing in the biogas upgrading is that it maintains a temperature gap of at least over 1.5 K in vapor and liquid

fraction compared to the freezing point (Berstad *et al.*, 2012; Yousef *et al.*, 2016). As shown in the figures, all temperature lines at each stage are higher than 1.5 K compared to the CO₂ freezing points.

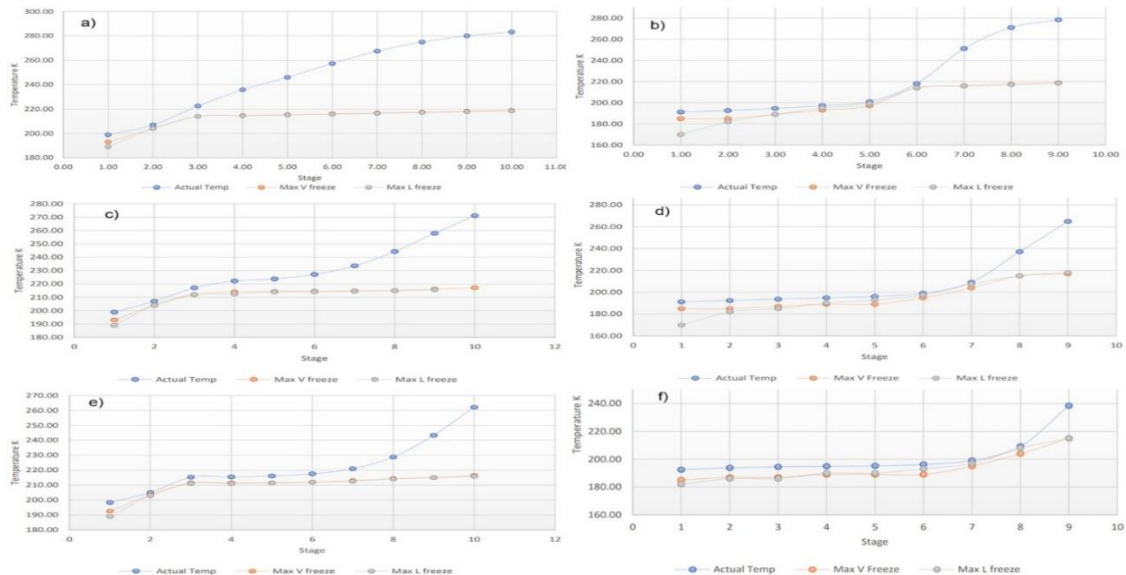


Fig.5. CO₂ freezing point comparison with stage-wise temperature profiles in the distillation process: (a) Column 01 in System 01, (b) Column 02 in System 01, (c) Column 01 in System 02, and (d) Column 02 in System 02, (e) Column 01 in System 03, and (f) Column 02 in System 03

Based on the simulation sensitivity analysis, the operating pressure for the distillation columns was adjusted to function at 49 bar for the first and 44 bar for the second distillation column as the optimum. Furthermore, another critical thermodynamic property in the biogas upgrading process is the feed temperature for the distillation columns. The

feed stream temperatures for Column 01 and Column 02 were maintained at -50 °C and -78 °C, respectively. The refrigeration cycle was used to keep the feed temperature and refrigeration load at the required level. Lower feed temperatures can be caused by frozen CO₂ in the distillation process. Considering these factors, it was determined that the feed temperatures

should be kept at -50 °C and -78 °C in distillation columns 01 and 02 to achieve the required CH₄ purity.

The process was designed to achieve higher biogas upgraded conditions with higher methane concentration at the output; the higher reflux ratio is more favored for the operation. The optimum reflux ratio that can be maintained in this thesis study was limited due to the CO₂-frozen environments. The optimum reflux ratio value was kept at 1 in the distillation columns 01 and 02 in all three simulations while changing the distillate-to-feed ratio as the variable parameter in the sensitive analysis to identify the optimum condition for the CO₂ frozen-free distillation process. The optimum distillate-to-feed ratio in the distillation column 01 was 0.54, 0.62 and 0.78, and for the distillation, column 02 was 0.922, 0.920, and 0.915, respectively, in systems 01, 02, and 03 with 50%, 60%, and 75% CH₄ methane feed biogas systems.

Low CH₄ Loss

Methane is the primary product output, and its "loss percentage" from the biogas upgrading process is a significant factor. One of the vital benefits of low-temperature biogas upgrading operation is the ability to keep lower CH₄ loss throughout the operation compared with other traditional biogas upgrading techniques. The product recovery of methane can be maintained at 99.53% for system 01 with a 50% methane feed, which means the CH₄ loss is 0.47%. This methane loss percentage is much lower than other traditional techniques.

The average methane loss value for traditional techniques is 1% in the water scrubbing technique, 1.8% in pressure swing adsorption, 0.5% in membrane technology, 1% in amine absorption, and 1.5% in genosorb scrubbing techniques. All these methane loss average percentage values are higher than the low-temperature upgrading process, resulting in value, and this competitive advantage assists in gaining energy-efficient and cost-effective operations.

Energy analysis results

The product stream conditions and power consumption results are shown in Figures 6 through 8. All three systems were designed with 1000 kmol/hr input feed flow rate and stetted output methane purity of 98.5%-mol in the methane product stream. The total energy requirement for the biogas upgrading process is getting reduced as the methane content increases; the system with 50%-mol CH₄ feed requires 22.97 MW energy input, and the system with 75%-mol/ system needs 20.77 MW.

System 1, with 50%-mol CH₄ content biogas feed, has the highest product recovery capacity compared with the two other systems, as shown in Figure 7. Even though it consumed an immense amount of energy, as shown in Figure 6, it produced the top total production rate of 981.71 kmol/hr with the highest product recovery of 99.53% CH₄ recovery and 99.30% CO₂ recovery. If carbon dioxide and methane are used as product streams, then system 1 provides better perspectives. However, system three has a higher CH₄ feed input and provides the highest liquid CH₄ product output of 717.26 kmol/hr, which has a better market demand as a renewable fuel.

The values of heating duty and refrigeration load at the decrease of CO₂ content in the raw biogas feed. Thermodynamic properties of the reboiler and condenser, the heating duty decreased to 0.55 MW from 1.43 MW in distillation column 01 and to 0.60 MW from 0.74 MW in distillation column 02, with a change of CO₂ concentration from 25%-mol to 50%-mol in the biogas feed. For the same, the refrigeration load in the

condenser unit changed to 1.43 MW from 1.57 MW in distillation column 01 and to 0.21 MW from 0.46 MW in distillation column 02. As shown in these results, the increased CO₂ content in the biogas feed caused more energy consumption for the biogas upgrading and separation in the distillation process. Figure 8 illustrates that System 01 (50% CH₄ feed) achieves the highest total product flow rate (CH₄ + CO₂), while System 03 (75% CH₄ feed) yields the highest liquid CH₄ flow rate but with reduced CO₂ output.

Based on the results shown in Figure 9, the proposed low-temperature biogas purification technique demands less energy than pressure swing adsorption, water scrubbing, and physical absorption techniques. The typical cryogenic method consumed 0.8 –1.4 kWh/Nm³ range energy in the biogas upgrading process (Adnan et al., 2019). Based on the results, the proposed method reflects better performance from an energy demand perspective. However, the chemical absorption and membrane separation have lower energy consumption than the proposed low-temperature upgrading technique. Based on the three-design system, the energy demand per Nm³ of purified gas is increased with the increased feed flow methane content.

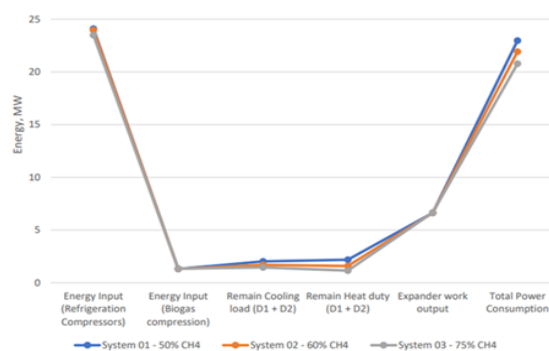


Fig.6. All Energy inputs and output (Expander) Results – MW.

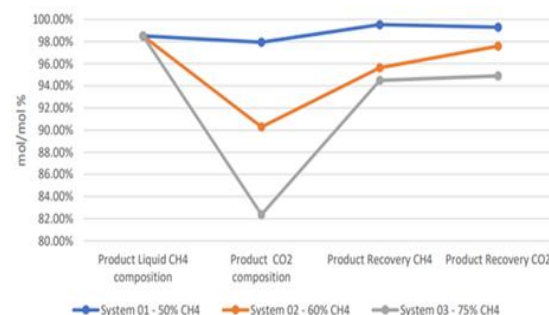


Fig.7. Product compositions and Product Recoveries.

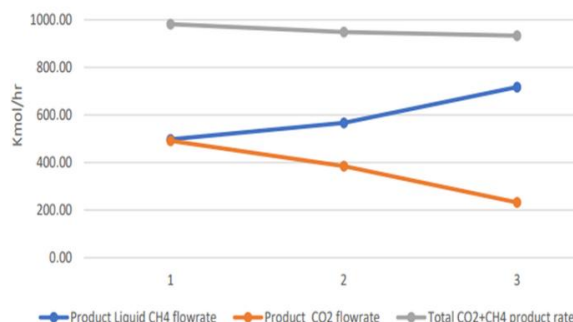


Fig.8. Product Flow rates (Kmol/hr), for system 01 (50% CH₄ feed), system 02 (60% CH₄ feed), and system 03 (75% CH₄ feed).

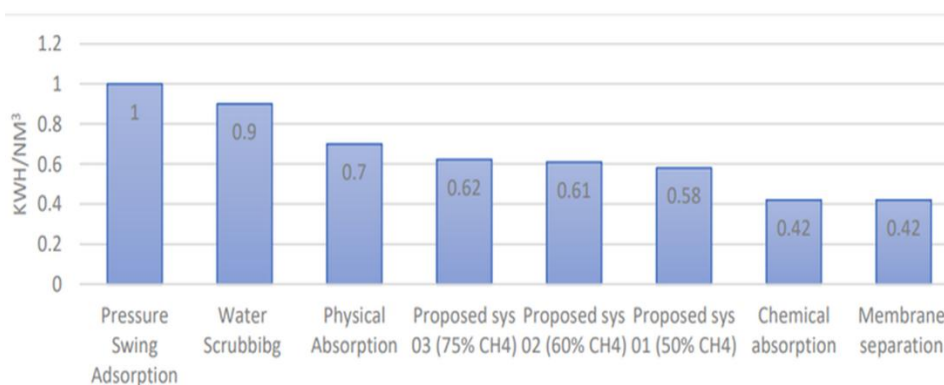


Fig.9. Energy demand for different technologies (kWh/Nm³).

Exergy analysis

The results obtained from the exergetic analysis for the three systems are shown in Figures 10 to 13. As for total exergetic efficiency, most of the components in system 03 with

a higher methane input stream displayed higher exergetic efficiency than the other systems, for example, distillation columns and some heat exchangers.

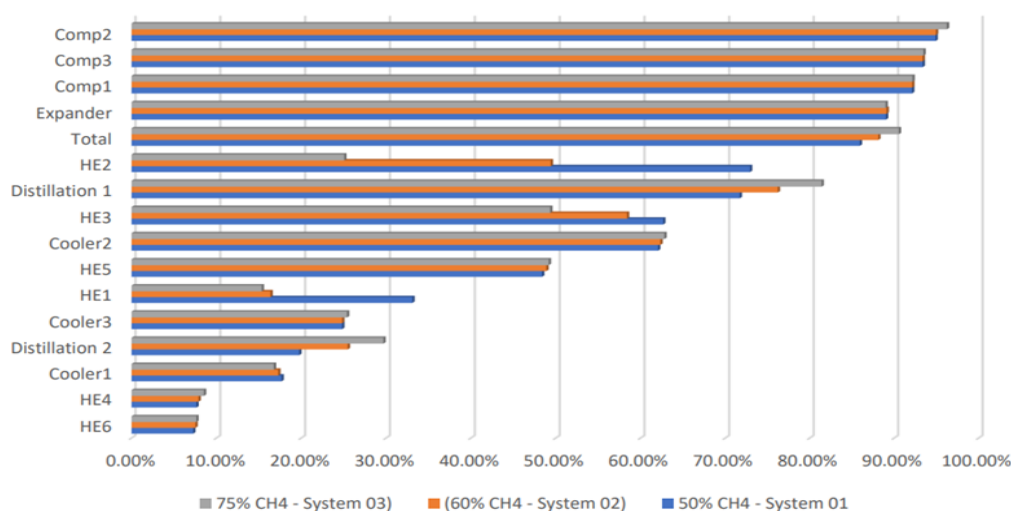


Fig.10. Exergetic efficiency for the most significant components in three simulated systems.

However, the precooling stage Heat exchanger 01, Heat exchanger 02, Heat exchanger 03, and air Coolers 01 showed opposing results with higher exergetic efficiency at system 01 with lower CH₄ feed conditions. The biogas input stream exchanged the heat with the bottom distillation column stream at the precooling stages. The available cooling load in system 01 bottom distillation column stream is lower, and the temperature is higher than in systems 02 and 03. All systems are designed to achieve the same temperature reduction at precooling stages. Hence, system 01, with lower methane content, showed a higher exergetic efficient heat exchanger than the other two systems.

Based on Figure 17 results and considering the overall system, the biogas exergetic efficiencies were recorded as 85.94%, 88.13%, and 90.56% of System 01 (50%-mol CH₄), System 02 (60%-mol CH₄), and System 03 (75%-mol CH₄), respectively. Based on the results of the energy analysis, the specific power consumption per kg of biogas feed, system 01 performed better than systems 02 and 03. However, from energy demand for the biogas upgrading process and an exergy analysis perspective, system 03, with higher methane content biogas feed, showed better results with higher exergetic efficiency than systems 01 and 02.

Similar to the exergy efficiency, the exergy destruction value of each component in the systems was calculated, and the

results can be analyzed to make decisions and conclusions. Figures 18-20 show the exergy destruction of each component as a percentage of overall and exergy destruction for three simulated systems for the most significant components only. The distillation columns, Heat exchangers such as HE4, HE5, cooler 2, and the Expander unit enclosed the highest exergy destruction in all three systems. The distillation process is generally because the distillation operation is greatly energy-demanding and thermodynamically lower efficient. Moreover, significant exergy destruction occurred within the distillation process because of exergy differences in input and output streams. Also, the temperature gradient between the reboiler and condenser caused the degradation of thermal energy and exergy of the distillation process (Javed et al., 2022). Furthermore, with the methane content increased in the feed stream, the amount of energy required for the overall separation is reduced, and the exergy destruction value and exergy destruction shared percentage from total have reduced from system 01 to 03. Considering the two-distillation column operation and based on the Aspen Plus simulation results, system 03, with the highest methane content, has a lower energy consumption of 1.15 MW and 1.45 MW compared to the other two systems, for the heating duty and condenser work respectively, and recorded the lowest exergy destruction of 9.25 MW among three systems for the overall distillation process as

mentioned in Figure 20. However, based on the exergetic efficiency values shown in Figure 17, the distillation column 01 showed over 65% exergetic efficiency in all three designs, which is a good performance value from an exergy perspective. Although the distillation column 02 recorded exergetic efficiency lower than 30% in all three simulations, the performance needed to be improved by reducing the exergy destructions.

The heat exchanger 04 and the expander unit recorded higher exergy destruction in all three designs. After the distillation in column 02, the HE4 noted higher exergy destruction in all three simulations. Design 01 and 02 recorded 17%, and Design 03 recorded 16% share from total exergy destruction, and the exergetic efficiency value is meager in all three simulations. The heat exchanger occurred at the vapor phase in both cold and hot streams in HE4. The required heat transfer area is also high, and the unit is more expensive. However, this heat exchanger serves one of the significant tasks in the system by reducing the temperature of the nitrogen working fluid up to the required level before the expanding process in the refrigeration cycle. Furthermore, like the distillation column 02 operation, even with higher exergy destruction, the HE4 performs a critical task in the design. Therefore, the performance of this unit needs to be improved by considering exergoeconomic analysis.

A simple refrigeration process was used in the simulation to obtain the cooling effect, and the pressure drops through the expander are very high in the refrigeration cycle. The pressure was reduced from 80 bar to 2 bar. As a result, the exergy destruction is higher in the expander unit in all three simulations, but the exergetic efficiency is over 88%, and the unit operates in good condition. Based on the exergy analysis perspective, the distillation column 02, HE1, HE4, HE5, HE6, and Air coolers of Cooler 1 and Cooler3, have lower exergetic efficiencies and need to improve operation condition and exergetic performance. Among these units, the distillation column 02 and HE4 have the highest exergy destruction value in all three simulations, which will be the most significant units to improve from the exergy perspective. The summary of exergy analysis is shown in Table 3.

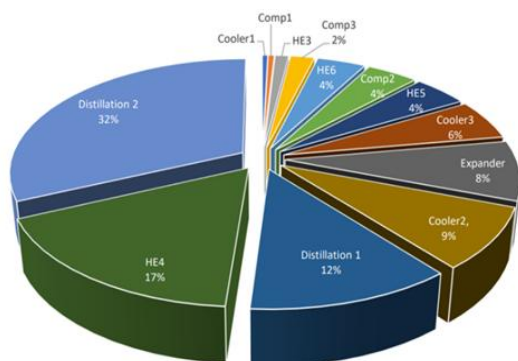


Fig.11. Distribution of the exergy destructions among the components in system 01.

Table 4. Total Module cost of the three simulated systems

	System 01 (50% CH ₄)	System 01 (50% CH ₄)	System 01 (50% CH ₄)
Total Module cost M\$ 2023	30.34	30.03	29.79

The cost estimation results are summarized in Figures 14–18. The component-wise cost comparisons for the three simulation designs are shown in Figures 19–21. Most

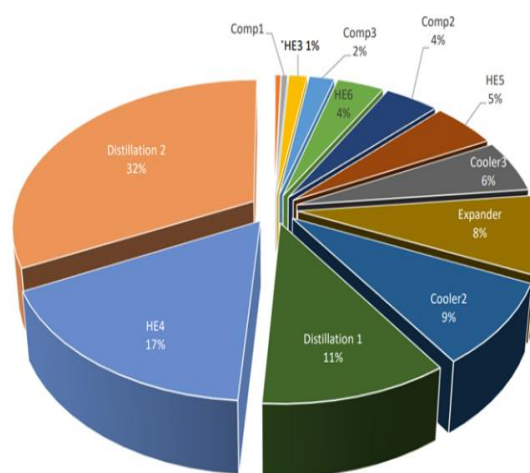


Fig.12. Distribution of the exergy destructions among the components in system 02.

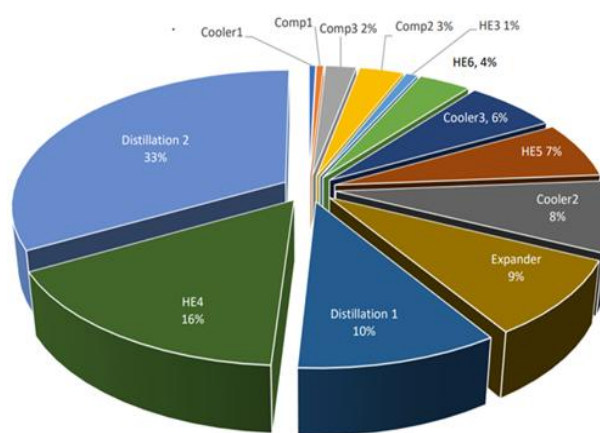


Fig.13. Distribution of the exergy destructions among the components in system 03.

Table 3. The results of the exergy analysis for the overall system at different feed conditions

	50% CH ₄ - system 01	60% CH ₄ - system 01	75% CH ₄ - system 03
$\dot{E}_{f,tot}$ (MW)	163.2264	185.3560	218.8680
$\dot{E}_{p,tot}$ (MW)	140.2750	163.3528	198.1999
$\dot{E}_{L,tot}$ (MW)	0.01	0.01	0.01
$\dot{E}_{D,tot}$ (MW)	22.9427	21.9944	20.6593
ε (%)	85.94%	88.13%	90.56%
$\gamma_{D,tot}$ (%)	14.06%	11.87%	9.44%

Economic analysis

The total module cost of three simulated systems was evaluated by summing up all module cost values for all equipment. Table 4 displays the results. Similar to exergy analysis, system 03 shows better results with low purchase equipment cost estimation. That means the required cost for purchasing equipment to produce a 98.5%-mol CH₄ purity system is lower in the higher methane-concentrated biogas feed system. The PEC has increased by 1.86% from 50%-mol CH₄ (system 01) compared to the 75%-mol CH₄ feed (system 03).

components in system 03 with higher CH₄ content recorded the lowest cost, and system 01 with lower CH₄ content showed the highest cost for most components. Because, with the same

molar flow rates in all three systems, the mass flow rates were lower with the higher CH₄ content due to the lower molecular weight of CH₄ compared to CO₂, and the required refrigeration load and compressor power in the refrigeration cycle are lower in the system 03. The following is observed for the turbomachinery in three designs: Compressor 2 is almost 45% of the total purchase cost, Compressor 3 is almost 23%, and Expander is almost 14%. Reflecting all three designs, system 01, with lower CH₄ content, demonstrates the highest equipment cost compared to the two other systems. Based on the result, the levelized carrying charges, the levelized operational and maintenance cost, and the levelized fuel cost are reduced from system 03 to system 01. As the purchase equipment cost is higher in system 01 with a lower CH₄ content feed, the O&M and FCL are higher in lower methane content system 01. That means the system with higher CH₄ content has lower operational and maintenance costs and lower "fuel" (=electricity) costs compared with higher CH₄ feed systems.

According to the total cost (Figure 17), the overall cost has been reduced by 20.55 M\$/year (which is 7.9%) from system 01 to system 03. The average biogas upgrading cost is around USD 2/MBTU. The upgrading process includes several steps, but this study assumed the feed is pre-purified and removed some impurities before feeding into the system. In this study, the cost of pre-purification from dust, nitrogen, hydrogen sulfide, and minor impurities is not included in the analysis. The average cost for the biogas upgrading process is less than the average value, as shown in Figure 18. The average cost per biogas feed is reduced with the CH₄ content increase in the biogas feed. The average cost for the biogas upgrading process without pre-purification steps is USD 0.98/MBTU in the 50%-mol CH₄ feed system, and the value is USD 0.61/MBTU in the 75%-mol CH₄ feed system.

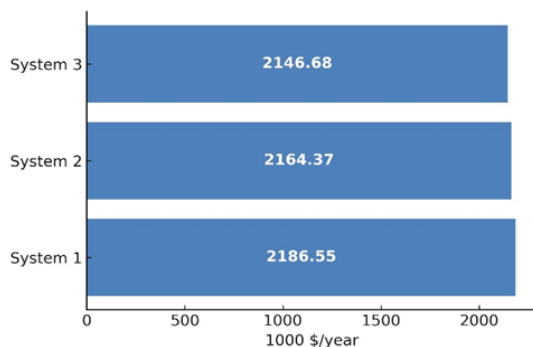


Fig.14. Levelized carrying charges CCL 1000\$/year in three systems.

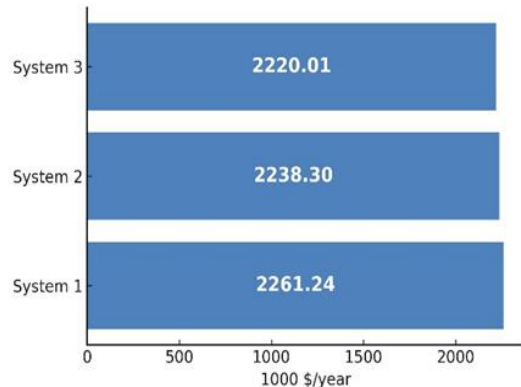


Fig.15. Levelized OMCL 1000\$/year in three systems.

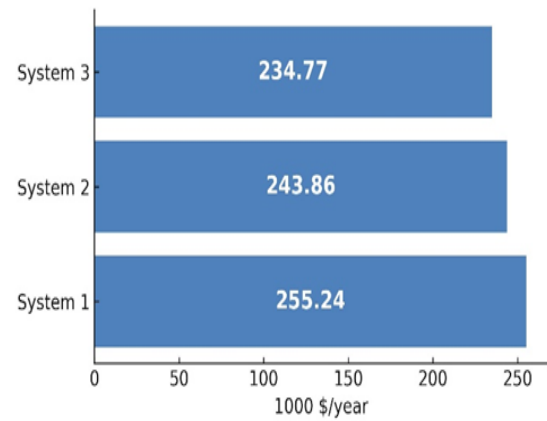


Fig.16. Levelized electricity cost ECL 1000\$/year in three systems.

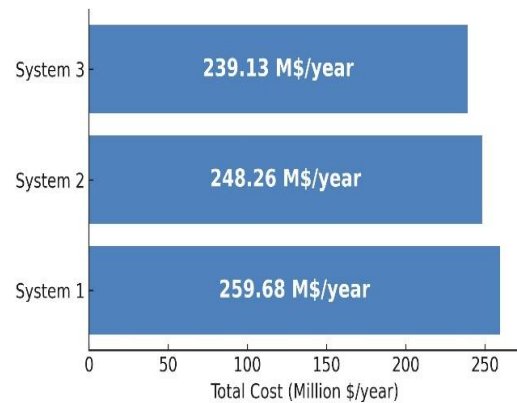


Fig.17. Total Annual Cost In Three Systems.

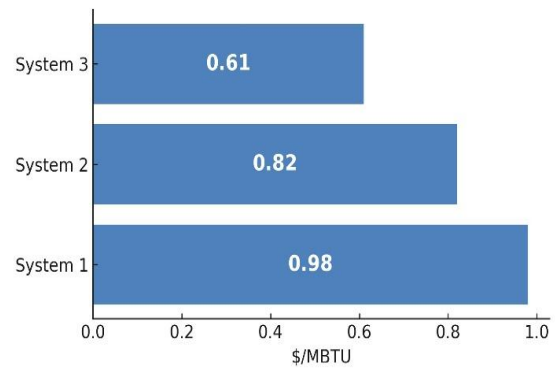


Fig.18. Average Biogas upgrading cost \$/MBTU in three systems.

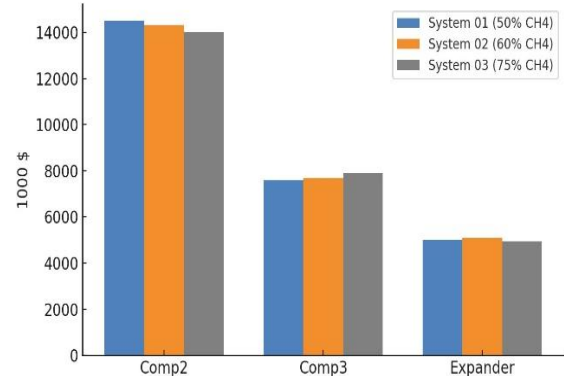


Fig. 19. Highest PEC components in three simulation designs.

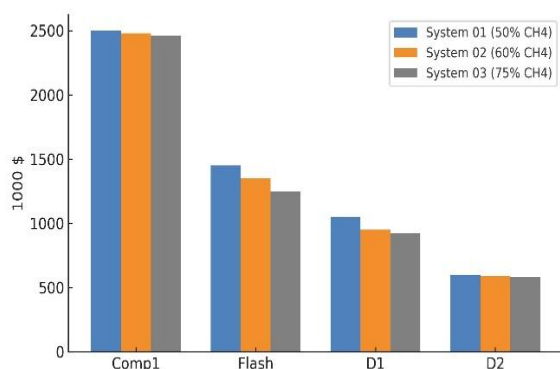


Fig.20. Middle-range PEC components in three simulation designs.

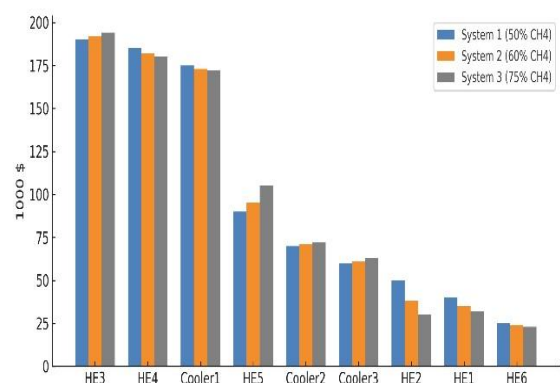


Fig. 21. Lowest-range PEC components in three simulation designs.

The specific operational and maintenance cost is highly dependent on the biogas feed rate, and with a higher feed flow rate, the average O&M is shown to have a lower value. The O&M of different biogas upgrading techniques is shown in Table 5, with different feed flow rate conditions. The design systems in these design simulations showed low O&M value compared with other traditional biogas upgrading techniques due to the lower contaminant biogas feed with higher feed flow rate conditions. Furthermore, the water-scrubbing and chemical absorption methods displayed a lower O&M than simulation results in 2000 m³/h and 137 m³/h feed flow rates, respectively.

Table 5. Operational and management cost comparison for different technologies and at different feed conditions

Method	O&M Cost (Cent \$/KWh)	Flowrate (m ³ /h)
Chemical absorption	1.92	137
Pressure Swing Adsorption	6.5	600
Physical Absorption	7.1	600
Typical Cryogenic separation	6.1	600
Membrane separation	6.7	600
Water Scrubbing	0.48	2000
Proposed sys 03 (75% CH ₄)	1.38	2909
Proposed sys 02 (60% CH ₄)	1.81	3028
Proposed sys 01 (50% CH ₄)	2.08	3039

CONCLUSION

A low-temperature biogas upgrading process was performed by producing two significant product outputs of CH₄-rich stream and CO₂-rich stream. Three biogas feed conditions were analyzed with 50%-mol, 60%-mol, and 75%-mol CH₄ concentrations. The distillation process was

used as the critical separation step while addressing the biogas' purification conditions without freezing out the CO₂. The low-temperature biogas upgrading method reveals its unique advantage of producing high-purity biomethane with 98.5%-mol and generating valuable by-products of CO₂ with 97.94%-mol in a 50%-mol CH₄ feed biogas system. The higher distillation column pressure reduces the risk of CO₂ freezing. The optimum distillation pressure is 49 bar for the first and 44 bar for the second distillation column. The higher reflux ratio and distillate-to-feed ratio are favored to gain higher methane concentration at the top product stream, but the value was limited due to the CO₂-frozen environments. The optimum distillate-to-feed ratio value that can be kept in the distillation column 01 was 0.54, 0.62 and 0.78, and for the distillation, column 02 was 0.922, 0.920, and 0.915, respectively, in systems 01, 02, and 03 with 50%-mol, 60%-mol, and 75%-mol CH₄ feed biogas systems. The optimum value for the number of stages was maintained at 10 and 9 for the distillation columns 01 and 02 for all three designs to perform a smooth distillation process without freezing the carbon dioxide at any stage.

Furthermore, the energy, exergy, and economic analysis was performed on the three biogas feed systems. The system performed better and moved positively with the increase of CH₄ content in the biogas feed. The total power consumption was reduced from 22.97 to 20.77 MW, and the specific energy consumption per kg of CH₄ was reduced from 10.30 to 6.21 MJ/kg from system 01 to system 03, with CH₄ content increasing from 50%-mol to 75%-mol. The exergetic efficiency of the overall system was increased from 85.94% to 90.56%. The highest exergy destruction was observed at the distillation column 02, and the heat exchanger that was used to cool down the refrigerant after the compression process. Then, the expander, distillation columns 01, and air coolers follow the highest exergy destructions in all three design simulations. Similar to energy and exergy analysis, system 03, with the highest CH₄ feed, showed better results in economic analysis. The total cost for the overall system, including investment, O&M, and fuel costs, was reduced from 259.68 to 239.13 M\$ from system 03 to system 01.

The energy demand for the biogas upgrading process varies from 0.58 kWh/Nm³ to 0.62 kWh/Nm³ of biogas feed from system 01 to system 03. The average biogas upgrading cost was reduced from USD 0.9/MBTU 8 to USD 0.61/MBTU from system 01 to system 03. These are decent values compared with other average figures in alternative technologies. Moreover, low-temperature biogas upgrading techniques have additional advantages, such as producing CH₄ at a higher purity while producing higher-purity CO₂ as a by-product. The product streams have higher pressure values and can contribute to reducing transportation and storage costs; additional chemicals for purification are not required.

The economic viability of cryogenic biogas purification systems can vary based on specific operation parameters, including scale, energy prices, and regional factors. Detailed feasibility studies will be considered for accurate assessments. The more complex biogas feed with more impurities can also be evaluated in the future to gain more realistic results.

Nomenclature

C_B	Bare module cost, USD	Subscripts	
C_M	Module cost, USD	0	Ambient, restricted dead state, reference
f_T	Temperature factor	A	State point at T_0 and p
f_p	Pressure factor	el	Electricity
f_m	Material factor	D	Destruction
f_d	Design factor	F	Fuel
f_{BM}	Bare module factor	i, j	Running index
E	Exergy, J	k	k-th component
$\dot{E}$	Exergy rate, W	L	Loss (exergetic analysis), levelized cost (economic analysis)
e	Specific exergy, kJ/kg	l	Liquid
H	Enthalpy, J	P	Product
h	Specific enthalpy, kJ/kg	tot	Total, overall
LHV	Lower Heating Value, J/kg	Abbreviations	
M	Mass, kg	DM	Dry Matter
n	Economic life Years	PSA	Pressure Swing Absorption
p	Pressure, bar	HE	Heat Exchanger
S	Entropy, kJ/K	CEPCI	Chemical Engineering Plant Cost Index
s	Specific entropy, kJ/kg K	CRF	Capital Recovery Factor
T	Temperature, K, °C	FCI	Fixed Capital Investment
ε	Exergetic efficiency, %	CC -	Carrying Chargers
Superscripts		FC -	Fuel Cost
CH	Chemical	CELF	Constant Escalation Levelization Factor
KN	Kinetic	CAPEX	Capital expenditure
M	Mechanical	Mtoe	Metric tons of oil equivalent
PH	Physical	O&MC	Operational and Maintenance Cost
PT	Potential	PEC	Purchase Equipment cost
T	Thermal	TCI	Total Capital Investment
		TRR	Total revenue requirement

REFERENCES

- Abd, A. A., Othman, M. R., Shamsudin, I. K., Helwani, Z., & Idris, I. (2023). Biogas upgrading to natural gas pipeline quality using pressure swing adsorption for CO₂ separation over uio-66: Experimental and dynamic modelling assessment. *Chemical Engineering Journal*, 453, 139774. <https://doi.org/10.1016/j.cej.2022.139774>
- Adnan, A. I., Ong, M. Y., Nomanbhay, S., Chew, K. W., & Show, P. L. (2019). Technologies for biogas upgrading to Biomethane: A review. *Bioengineering*, 6(4), 92. <https://doi.org/10.3390/bioengineering6040092>
- (n.d.). Bejan, A., Tsatsaronis, G., Moran M. (1996) *Thermal Design and Optimization*-John Wiley & Sons.
- Berstad, D., Neksa, P., & Anantharaman, R. (2012). Low-temperature CO₂ removal from natural gas. *Energy Procedia*, 26, 41-48. <https://doi.org/10.1016/j.egypro.2012.06.008>
- Beteta, O., & Ivanova, S. (2015, September). Cool down with liquid nitrogen. *Chemical Engineering Progress*, 111(9), 30–35. <https://www.aiche.org/cep>
- Burchell, T., & Rogers, M. (2000). Low pressure storage of natural gas for vehicular applications. *SAE Technical Paper Series*. <https://doi.org/10.4271/2000-01-2205>
- Cavin, L., Mošat, A., Fischer, U., & Hungerbühler, K. (2004). Identifying the optimal process design for a chemical process to be implemented in an existing multipurpose batch plant. *Computer Aided Chemical Engineering*, 367-372. [https://doi.org/10.1016/s1570-7946\(04\)80127-5](https://doi.org/10.1016/s1570-7946(04)80127-5)
- Christensen, T. H. (2010). Introduction to waste management. *Solid Waste Technology & Management*, 1-16. <https://doi.org/10.1002/9780470666883.ch1>
- Fan, Q. H., Li, H. Y., Yin, Q. S., Jia, L. X., Weisend, J. G., Barclay, J., Breon, S., Demko, J., DiPirro, M., Kelley, J. P., Kittel, P., Klebaner, A., Zeller, A., Zagarola, M., Van Sciver, S., Rowe, A., Pfotenhauer, J., Peterson, T., & Lock, J. (2008). Design and analysis of a small-scale biogas liquefaction cycle. *AIP Conference Proceedings*, 985, 1166-1174. <https://doi.org/10.1063/1.2908468>
- Hamdy, S., Moser, F., Morosuk, T., & Tsatsaronis, G. (2019). Exergy-based and economic evaluation of liquefaction processes for cryogenics energy storage. *Energies*, 12(3), 493. <https://doi.org/10.3390/en12030493>
- Hashemi, S. E., Sarker, S., Lien, K. M., Schnell, S. K., & Austbø, B. (2019). Cryogenic vs. absorption biogas upgrading in liquefied biomethane production – An energy efficiency analysis. *Fuel*, 245, 294-304. <https://doi.org/10.1016/j.fuel.2019.01.172>
- Hepburn, C., Adlen, E., Beddington, J., Carter, E. A., Fuss, S., Mac Dowell, N., Minx, J. C., Smith, P., & Williams, C. K. (2019). The technological and economic prospects for CO₂ utilization and removal. *Nature*, 575(7781), 87–97. <https://doi.org/10.1038/s41586-019-1681-6>
- International Energy Agency. (2024, October 18). *World energy outlook 2024*. <https://www.iea.org>
- Javed, A., Hassan, A., Babar, M., Azhar, U., Riaz, A., Mujahid, R., Ahmad, T., Mubashir, M., Lim, H. R., Show, P. L., & Khoo, K. S. (2022). A comparison of the exergy efficiencies of various heat-integrated distillation columns. *Energies*, 15(18), 6498. <https://doi.org/10.3390/en15186498>
- Kadam, R., & Panwar, N. (2017). Recent advancement in biogas enrichment and its applications. *Renewable and Sustainable Energy Reviews*, 73, 892-903. <https://doi.org/10.1016/j.rser.2017.01.167>
- Li, Y. (2011). Cryogen based energy storage: Process modelling and optimisation. <https://api.semanticscholar.org/CorpusID:108405169>
- Maqsood, K., Mullick, A., Ali, A., Kargupta, K., & Ganguly, S. (2014). Cryogenic carbon dioxide separation from natural gas: A review based on conventional and novel emerging technologies. *Reviews in Chemical Engineering*, 30(5). <https://doi.org/10.1515/revce-2014-0009>
- Morosuk, T., & Tsatsaronis, G. (2019). Splitting physical exergy: Theory and application. *Energy*, 167, 698-707. <https://doi.org/10.1016/j.energy.2018.10.090>

- Naeeji, E., Noorpoor, A., & Ghanavati, H. (2022). Energy, Exergy, and economic analysis of cryogenic distillation and chemical scrubbing for biogas upgrading and hydrogen production. *Sustainability*, 14(6), 3686. <https://doi.org/10.3390/su14063686>
- Naquash, A., Qyyum, M. A., Haider, J., Bokhari, A., Lim, H., & Lee, M. (2022). State-of-the-art assessment of cryogenic technologies for biogas upgrading: Energy, economic, and environmental perspectives. *Renewable and Sustainable Energy Reviews*, 154, 111826. <https://doi.org/10.1016/j.rser.2021.111826>
- Paolini, V., Tratzi, P., Torre, M., Tomassetti, L., Segreto, M., & Petraccini, F. (2021). Water scrubbing for biogas upgrading: Developments and innovations. *Emerging Technologies and Biological Systems for Biogas Upgrading*, 57-71. <https://doi.org/10.1016/b978-0-12-822808-1.00001-5>
- Pavičić, J., Novak Mavar, K., Brkić, V., & Simon, K. (2022). Biogas and Biomethane production and usage: Technology development, advantages and challenges in Europe. *Energies*, 15(8), 2940. <https://doi.org/10.3390/en15082940>
- Rasi, S., Lätelä, J., & Rintala, J. (2011). Trace compounds affecting biogas energy utilisation – A review. *Energy Conversion and Management*, 52(12), 3369-3375. <https://doi.org/10.1016/j.enconman.2011.07.005>
- Shafeeq, A., Daood, S. S., Muhammad, A., & Ijaz, A. (2010). Effect of variable reflux ratio on binary distillation in a laboratory scale distillation column. 2010 2nd International Conference on Chemical, Biological and Environmental Engineering, 35-38. <https://doi.org/10.1109/icbee.2010.5650925>
- Sun, Q., Li, H., Yan, J., Liu, L., Yu, Z., & Yu, X. (2015). Selection of appropriate biogas upgrading technology-a review of biogas cleaning, upgrading and utilisation. *Renewable and Sustainable Energy Reviews*, 51, 521-532. <https://doi.org/10.1016/j.rser.2015.06.029>
- Tuinier, M. J., & Van Sint Annaland, M. (2012). Biogas purification using cryogenic packed-bed technology. *Industrial & Engineering Chemistry Research*, 51(15), 5552-5558. <https://doi.org/10.1021/ie202606g>
- Ulrich, G. D., & Vasudevan, P. T. (2004). Chemical engineering process design and economics: A practical guide (2nd ed.). Process Publishing.
- Vilardi, G., Bassano, C., Deiana, P., & Verdone, N. (2020). Exergy and energy analysis of three biogas upgrading processes. *Energy Conversion and Management*, 224, 113323. <https://doi.org/10.1016/j.enconman.2020.113323>
- Wong, S., & Bioletti, R. (2002). Carbon dioxide separation technologies (Technical Report). Carbon & Energy Management, Alberta Research Council. <http://www.ipcc.ch/>
- Xu, G., Liang, F., Yang, Y., Hu, Y., Zhang, K., & Liu, W. (2014). An improved CO₂ separation and purification system based on cryogenic separation and distillation theory. *Energies*, 7(5), 3484-3502. <https://doi.org/10.3390/en7053484>
- Yousef, A. M., Eldrainy, Y. A., El-Maghlany, W. M., & Attia, A. (2016). Upgrading biogas by a low-temperature CO₂ removal technique. *Alexandria Engineering Journal*, 55(2), 1143-1150. <https://doi.org/10.1016/j.aej.2016.03.026>
- Yousef, A. M., El-Maghlany, W. M., Eldrainy, Y. A., & Attia, A. (2018). New approach for biogas purification using cryogenic separation and distillation process for CO₂ capture. *Energy*, 156, 328-351. <https://doi.org/10.1016/j.energy.2018.05.106>
- ZareNezhad, B. (2006). Prediction of CO₂ freezing points for the mixtures of CO₂-CH₄ at cryogenic conditions of NGL extraction plants. *Korean Journal of Chemical Engineering*, 23(5), 827-831. <https://doi.org/10.1007/bf02705935>
- Zhang, L., Burgass, R., Chapoy, A., Tohidi, B., & Solbraa, E. (2011). Measurement and Modeling of CO₂ Frost Points in the CO₂–Methane Systems. *Journal of Chemical & Engineering Data*, 56(6), 2971-2975. <https://doi.org/10.1021/je200261a>

تحليل تنقية الغاز الحيوي باستخدام التقطير منخفض الحرارة لاحتجاز ثاني أكسيد الكربون: تقييم إكسيري واقتصادي

داسيث إكاتاياكا^١، تتيانا موروزوق^٢ وأحمد جادو^٣

^١كلية التكنولوجيا - جامعة سابرا اغلوا في سريلانكا - بيليهولوي - سريلانكا

^٢معهد هندسة الطاقة - جامعة برلين للتقنية - برلين - ألمانيا

^٣قسم الهندسة الزراعية والنظم الحيوية - كلية الزراعة - جامعة المنصورة - مصر

المخلص

مع التوجه العالمي المتزايد نحو مصادر الطاقة البديلة، يتوقع أن يلعب الغاز الحيوي والبيوغاز المكرر (الميثان الحيوي) دوراً محورياً في مستقبل الطاقة. تهدف عملية ترقية الغاز الحيوي إلى إنتاج ميثان عالي النقاء، مع فصل ثاني أكسيد الكربون كمنتج ثانوي، وهي عملية قابلة للتطبيق في عدة مجالات. في هذا البحث، تم تطوير وتحليل طريقة تقطير مزدوج منخفضة الحرارة لتنقية الغاز الحيوي. تمت دراسة ثلاثة أنظمة تغذية مختلفة للغاز الحيوي تحتوي على تراكيز نموذجية للميثان تبلغ ٥٠٪، ٦٠٪، و ٧٥٪ مولية. وقد تم تنفيذ عملية التنقية عند درجات حرارة منخفضة باستخدام دورة تبريد قائمة على النيتروجين. شملت الدراسة التصميم والمحاكاة وتحليل الحساسية وخيارات التحسين باستخدام برنامج Aspen Plus، بالإضافة إلى إجراء تحليلات طاقة، إكسيريجية، واقتصادية. صُممت الأنظمة الثلاثة لمعالجة تدفق غازي يبلغ ١٠٠٠ كيلومتر/ساعة، بهدف الوصول إلى نقاء ميثان بنسبة ٩٨.٥٪ مولية، مع إنتاج تيار ثاني أكسيد الكربون بنقاء مرتفع. وقد أجريت المحاكاة حتى الوصول إلى بيئة خالية من تجمد ثاني أكسيد الكربون أظهرت النتائج أنه مع زيادة نسبة الميثان في التغذية من ٥٠٪ إلى ٧٥٪، يمكن تحقيق ما يلي: تقليل استهلاك الطاقة الكلي من ٢٢,٩٧ ميغا واط إلى ٢٠,٧٧ ميغا واط، وخفض استهلاك الطاقة النوعي من ١٠,٣ ميغا جول/كجم CH₄ إلى ٦,٢١ ميغا جول/كجم CH₄، وزيادة الكفاءة الإكسيريجية الإجمالية بمقدار ٣ نقاط مئوية، إلى جانب خفض إجمالي التكاليف المطلوبة للنظام (بما يشمل الاستثمار، التشغيل، والصيانة، وتكاليف الوقود) من ٢٥٩,٦٨ مليون دولار أمريكي إلى ٢٣٩,١٣ مليون دولار أمريكي.