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Comparative Analysis of Thermodynamic Performance of Two Cryogenic Systems for Biogas Upgrading Combined with Carbon Capture

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ABSTRACT

Cryogenic systems have been recognized as a biogas upgrading technology that is more environmentally friendly and less energy-intensive by capturing carbon dioxide in either liquid or solid form with negligible entrained methane concentration. Cryogenic distillation (D-SYS) and Liquefaction combined with desublimation (LD-SYS) are identified as cryogenic systems for accomplishing these functions. In this work, these alternatives of equal capacity were investigated and compared from a thermodynamic point of view, using standard measures: gas purity, removal efficiency, methane slip, energy consumption and exergy analysis. The simulation models of the two systems were performed using Aspen HYSYS 2006. The simulation results were in accordance with previous researches. It was shown that D-SYS displays better results in gas purity, methane loss, removal efficiency and energy consumption than LD-SYS; Moreover, the exergy efficiency of D-SYS and LD-SYS were found to be 95.59% and 92.80% respectively. It was also found that more that 80% of internal exergy loss occurs during cooling and refrigeration process. This work is useful for efficient thermodynamic design of cryogenic systems.

1. INTRODUCTION

The indiscriminate emission of greenhouse gases into the atmosphere through combustion of fossil fuels needs to be avoided in order to reduce the detrimental effect of global warming and air quality. For the 2℃ target set by the Paris climate accord to be achieved, some important mitigation technologies have to be adopted (Tan *et al.,* 2016). One of the ways of overcoming this problem is renewable energy production accompanied with carbon capture (Eric *et al.*, 2009). The availability of raw materials and environmental benefit of biogas production has attracted the interest of the public as sustainable energy to mitigate the negative impact of these gases.

Biogas can be produced from manure, sewage sludge, organic solid wastes and energy crop. Digester chamber and landfills are the two sources of biogas production (Johansson, 2008). The raw material used and the process design adopted determine the composition of the gases leaving the anaerobic digester (Rasi *et al.*, 2007). Apart from methane and carbon dioxide being the main components, other species such as water vapour, hydrogen sulphide, nitrogen, oxygen and volatile organic compounds can be found in trace amount (Chmielewski *et al.,* 2013). The need to use biogas for power generation and as vehicle fuel has prompted the need for its cleaning and upgrading (Margareta *et al.*, 2006). The main reason behind the upgrading is to increase the methane concentration in the biogas. The energy cost for upgrading the gas is typically 3-6% of the calorific value of upgraded gas, depending on the techniques used (Persson, 2003). The technologies involved in biogas upgrading are based on four principles: cryogenics, adsorption, membrane and absorption (Bauer *et al.,* 2013). Biogas upgrading can be achieved by using different technologies: Pressure swing adsorption, chemical solvent scrubbing, Pressurized water scrubbing, Physical solvent scrubbing, membrane separation, cryogenic distillation, supersonic separation, industrial lungs. Some of these techniques are available commercially while some are still under development. (Peterson and Wellinger, 2009). Cryogenics is a science of very low temperature that uses difference in condensing temperature as the basis of separation. With the massive release of CO_2 associated with rapid expansion of biogas, CO_2 capture of upgraded biogas attracts great attention (Li *et al., 2017*). Cryogenics has been recognized as a technique that is environmentally and economically beneficial due to its capability of producing methane with high purity along with pure CO_2 as side product. However, the process is still under development although few plants are now available at commercial level in Europe (Bauer *et al.,* 2013).

Researchers have carried out studies on process design, optimization and performance assessment (energy efficiency) of cryogenic plants. Yousef *et al.* (2016) proposed a cryogenic system for biogas upgrading. The process was simulated in *Aspen HYSYS* without allowing solid CO_2 formation within the distillation column. They concluded that, the proposed upgrading system is energy efficient and environmentally beneficial as it captures both CH_4 and CO_2 with a very good quality. Maiziriwan *et al.* (2018) presented work on simulation of biogas liquefaction in *Aspen HYSYS* using cryogenic process with the aim of assessing the technology to obtain liquefied biomethane and CO_2 as by-product. The review work presented by Yuting *et al.* (2017) was based on different cryogenic systems combined with carbon capture and property impact (Phase equilibrium) in cryogenic systems. They recommended that, thermodynamic optimization and comparison of different cryogenic systems combined with carbon capture should be evaluated. Yang *et al.* (2015) proposed and analysed a new liquefaction method together with desublimation for CO_2 separation based on N_2/CO_2 phase equilibrium. They concluded that Peng-Robinson together with van der Waals mixing rule is more accurate than other cubic equations of state. It was also reported in their work that total recovery and CO_2 purity of the proposed system were satisfactory. Even though, the simulated biogas upgrading combined with carbon capture using cryogenic process have already been examined in some aspects, studies on detailed exergy analysis of this system are very scarce.

Exergy analysis is a thermodynamic analysis technique, resulting from combination of first and second laws of thermodynamics. It provides better results when used for assessing and comparing processes because both the quantity and quality of energy are put into consideration (Ahamed *et al., 2011*). Considering the processing steps involved in cryogenic process for upgrading biogas such as compression and refrigeration, large amount of energy is required for the successive operations (Tuinier *et al.*, 2012). Therefore, comparative assessment of different cryogenic systems combined with carbon capture is necessary.

This paper is aimed at analyzing thermodynamically two types of cryogenic systems combined with carbon capture: D-SYS and LD-SYS. Their performances were evaluated and compared on the basis of gas purity, removal efficiency, methane slip, power consumption and exergy analysis.

2. MATERIAL AND METHODS

2.1. Model Development and Process Simulation

Produced biogas from a digester contains methane, carbon dioxide, water vapour and other impurities in trace amounts. This work models raw biogas processing facility that uses refrigerants to condense CO_2 from dry biogas after the water vapour removal. The process simulation of the systems was carried out using *Aspen HYSYS version 2006* using the Peng Robinson equation of state. The model consists of compressors, heat exchangers, distillation column, flash drums for vapour-liquid separation, pump, adsorber and cascade refrigeration cycle using ethylene/propylene as refrigerants. A linear model of cascade cycle developed by Etoumi *et al.;* (2015) and validated by rigorous HYSYS simulations given in Equation (1) was used in conjunction with Equations (2) and (3) to predict the power requirement and heat output in the cycles for streams 10 and 13 in LD-SYS and 10 and condenser (distillation column) in D-SYS.

$$COP_{act} = 0.596COP_{ideal} - 0.213 \quad (1)$$

$$COP_{ideal} = \frac{T_{evap}}{T_{cond} - T_{cond}} \quad (2)$$

$$COP_{act} = \frac{Q_{evap}}{W_{in}} \quad (3)$$

Where COP_{act}, the actual coefficient of performance; COP_{ideal}, the ideal coefficient of performance; Q_{evap} the heat removed by the refrigeration cycle; W_{in}, the work input in the refrigeration cycle, T_{evap}, the temperature of the evaporator; T_{cond} the temperature of the condenser.

Two different configurations were considered for modeling the cryogenic system combined carbon capture for biogas upgrading: D-SYS and LD-SYS. For justifiable comparison, the feed streams to the two systems were made to be of the same property. The properties of the feed raw biogas are shown in Table 1. A standard operating pressure range for transmission system is 2758 to 9928 kPa. Pressure of the upgraded biogas of the two systems is targeted at 3590 kPa. The tendency of the biogas to form liquid in the transmission line is examined by comparing the dew point temperature with the temperature of the upgraded biogas. The dew point temperature at this pressure is obtained from dew point curve for upgraded biogas generated in *Aspen HYSYS*.

The process model was developed based on the following assumptions:

- Kinetic and potential exergy are neglected.
- The reference temperature and pressure are 25 °C and 101.3 kPa respectively.
- The feeds conditions are at a temperature of 35 °C and a pressure 101.3 kPa.
- All gases behave ideally.
- The pressure drop in coolers used in the simulation is taken to be zero.
- The CO_2 in D-SYS is captured in liquid form at a pressure of 112 bar.
- The effect of refrigeration cycle is taken into consideration by emphasizing on heat absorbed by the cycle and work input on the processing plant.
- The condenser temperature in the refrigeration cycle is assumed to be 25 °C.
- The chemical exergy of CO_2 in any state is considered the same.

Table 1: Operating conditions of the feed raw biogas (Rasi, 2009; Nizami, 2012; Asadullah, 2014)

Temperature (°C)	Pressure (kPa)	Molar flow (kgmol/hr)	% mole composition		
			CH_4	CO_2	H_2O
35	85	1000	60	36	04

2.2. Distillation System (D-SYS)

The flow diagram of distillation system for biogas upgrading combined with carbon capture is shown in Figure 1. The feed raw biogas at 35 °C and 101.3 kPa was pressurized and then cooled to temperature 70 °C before entering the first separator (SEP 1) where part of the water vapour (3) is removed. The overhead stream (4) was made to pass through the adsorber (ADS) where the remaining water vapour (5) was removed. The stream from ADS (6) was further pressurized to 5000 kPa and subsequently pre-cooled in gas/gas heat exchanger (LNG) by already refrigerated gas (11) at -88 °C. The cooled stream (8) was then fed to the chiller (CO2) where further cooling was accomplished. The outgoing stream from the chiller (9) at temperature -18.9 °C was then refrigerated to -62 °C before being processed in a distillation column (DIST) to produce the upgraded biogas (UBG) as the top product and liquid CO_2 (LC) as the bottom product (12). It should be noted that the cooling duty of condenser in the distillation column is provided by the refrigeration cycle.

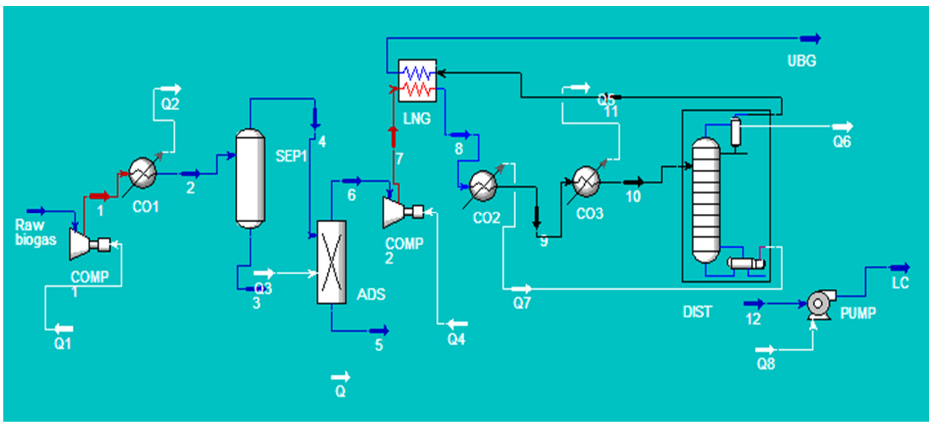

Figure 1: Process flow diagram of Distillation system (D-SYS)

The data considered for the successive running of the column is presented in Table 2. With the current operating conditions, assumed values for column feed and condenser temperatures are the best values for maximum purity of the products subject to CO_2 freeze-out prevention within the column. The assumed exit pressure of CO_2 from the processing plant is considered export pressure for transport which is very close to what was assumed by Berstad *et al.* (2011).

Table 2: Distillation column data of the process simulation model

Parameter	Value
Feed stage	3
Number of stages	8
Condenser pressure(kPa)	3600
Reboiler pressure(kPa)	4050
Condenser temperature (°C)	-88
Reflux ratio	2.5
Reboiler duty (kW)	2181
Condenser duty (kW)	1798

2.3. Liquefaction Combined with Desublimation System (LD-SYS)

The process flow diagram of the liquefaction combined with desublimation system for biogas upgrading is shown in Figure 2. The dry gas (6) obtained after the raw biogas had been passed through SEP1 and ADS was compressed to a pressure 6000 kPa. The high pressure stream (8) was cooled before it entered SEP2 where partially condensed CO_2 was separated. The overhead stream (12) in SEP2 was made to pass through the chiller (CO4) for further cooling. The cooled stream (13) was then allowed to undergo expansion through a Joule-Thompson nozzle into an expansion vessel at pressure 500 kPa and temperature -106.3 ºC. The solid CO_2 (SC1) was separated in SEP3. The upgraded biogas (UBG) obtained in SEP3 (15) was pressurized and cooled in order to meet up with pipeline flowing pressure. The bottom stream from SEP1 (11) was processed further by reducing its pressure before it finally enters SEP4 where dissolved CH_4 is separated. The overhead product from SEP4 (17) was recycled back to the system because of the high concentration of CO_2 while the bottom product was collected as desublimated CO_2 (SC2).

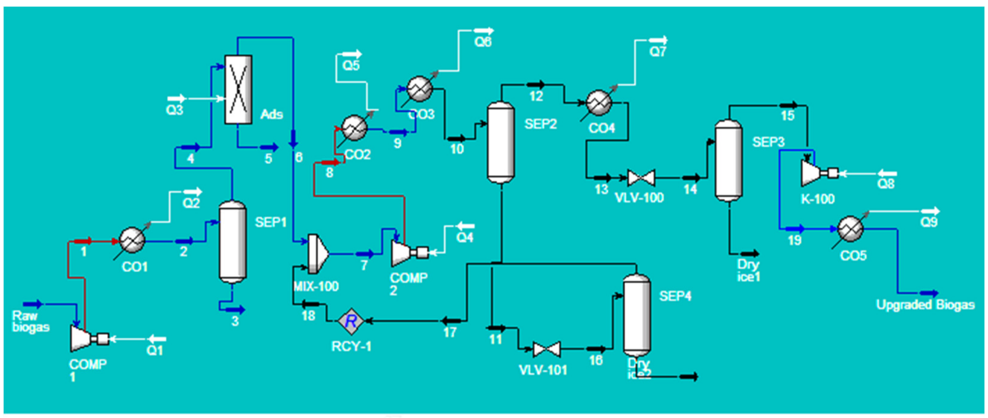

Figure 2: Process flow diagram of liquefaction and desublimation system (LD-SYS)

2.4. Exergy Analysis

Thermodynamic analysis of separation process is required for sensible utilization of energy. The entropies and enthalpies of every stream at both stream and environmental conditions needed in calculating exergy of steams were estimated using *Aspen HYSYS version 2006* software. Mass and energy data from the simulation model were transferred to spreadsheet to compute the exergy of the streams and thermodynamic losses of the process under study.

The physical exergy B^{ph} of a stream is calculated using Equation (4).

$$B^{ph} = M\{(H - H_o) - T_o(S - S_o)\} \quad (4)$$

Where:

M is the molar flow rate of a stream; H and S , the specific enthalpy and entropy calculated at the stream condition; H_o and S_o are the specific enthalpy and entropy calculated at the atmospheric temperature(T_o) and pressure (P_o).

The specific chemical exergy at STP derived by Szargut (2007) are considered for all the species involved in the process simulation. The chemical exergy (B_s^{ch})of a stream containing more than one component k of mole fraction X can be determined using Equation (5).

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$$B_s^{ch} = \sum_k^1 X_k B^{chk} + RT_o \sum_k^1 X_k \ln(X_k) \quad (5)$$

The total exergy associated with a particular stream is calculated by adding the physical and chemical exergy of that stream. The exergy transfer, EX_q by heat or cold is estimated using Equation (6).

$$EX_q = \left(1 - \frac{T_o}{T}\right)Q \quad (6)$$

Where T is the temperature of the stream and Q is the quantity of heat transferred.

The internal exergy destruction (I) for all the equipment in the systems is calculated by using Equation (7).

$$I = \sum B_{in} - \sum B_{out} \quad (7)$$

Where:

B_{in} is the exergy flow into the system and B_{out} is the exergy flow out of the system.

It is noticed that some equipment operate above the atmospheric temperature while some operate below the atmospheric temperature, hence, the total internal exergy losses in the systems was calculated as the sum of internal exergy losses of all the equipment that make up the system. The output streams from the systems are classified into waste and useful streams. In this case, all the output streams are considered waste streams except UBG and CO_2 since they are released directly into the environment. Hence, external exergy loss was then taken as total exergy of waste streams ($\sum B_{ow}$).

The realistic thermodynamic performance of a process unit can be assessed by exergy efficiency. Fratzscher *et al.* (1986) and Grassmann (1950) defined exergy efficiency (η_b) as the ratio of exergy flow of useful products ($\sum B_{ou}$) to total exergy flow into the system ($\sum B_{in}$). This can be expressed mathematically as shown in Equation (8).

$$\eta_b = \frac{\sum B_{ou}}{\sum B_{in}} = 1 - \frac{I}{\sum B_{in}} - \frac{\sum B_{ow}}{\sum B_{in}} \quad (8)$$

$$\sum B_{in} = \sum B_{feed} + \sum B_w \quad (9)$$

Where:

$\sum B_w$ and $\sum B_{feed}$ represent total exergy flow accompanying work and feed into the system respectively.

3. RESULTS AND DISCUSSION

3.1. Gas Purity

The properties and chemical compositions of product streams are presented in Table 3. It is worth noting that the UBG from the systems is made to be at the same temperature and pressure. For UBG in D-SYS, the percentage purity of 97.67% with CO_2 concentration of 2.33% was attained. This is a better result compared to the simulation results obtained by Yousef *et al.* (2016). The percentage purity of UBG through LD-SYS in the simulated results is 94.10%. The quality of UBG from both configurations is in accordance with the European standard for gas grid injection. The maximum concentration of CO_2 in UBG that can be used for gas grid injection is 1-8 mol% in Europe and 2-3 mol% in the USA (Echterhoff and Mckee, 1991).

The chemical compositions of worthily by-product (CO_2 streams) are also examined. In the case of D-SYS where the CO_2 stream is in liquid form, the percentage purity is a little bit higher than that of LD-SYS. The higher percentage purity of UBG and LC in D-SYS could be ascribed to distillation column considered instead of flash drum for CH_4-CO_2 separation. The production of CO_2 in either solid or liquid form makes the systems better than other biogas upgrading technologies which require additional cost if CO_2 would be considered as a valuable by-product.

Table 3: Results of process simulation

Properties	D-SYS		LD-SYS		
	UBG	LC	UBG	SC1	SC2
Temperature (°C)	30	15.3	30	-106.3	-77.5
Pressure (kPa)	3590	11200	3590	500	500
Molar flow (kgmole/hr)	614	345.7	629.4	140.3	190
Composition (%)					
CH_4	0.9767	0.0001	0.9406	0.0326	0.0164
CO_2	0.0230	0.9999	0.0594	0.9674	0.9836
H_2O	0.0000	0.0000	0,0000	0.0000	0.0000

3.2. Removal Efficiency and Energy Consumption

The simulated results presented in Table 4 shows that 96.02% of CO_2 in raw biogas feed is removed in D-SYS. Considering the components molar flow rate of CO_2 streams (SC1 and SC2) in the two outlets of LD-SYS, the removal efficiency was estimated to be 89.62%. The comparison of removal efficiencies of the two alternatives shows the more effective system to be D-SYS. An overview of energy consumption of the two alternatives is also shown in Table 4. In the case of D-SYS, the total power consumed was 6877.71 kW with 68.06% being expended in raw biogas pre-compression while 31.28% and 0.66% were used in auxiliary refrigeration cycle and LC pumping respectively. The specific energy consumed was 0.312 kWh/Nm^3 raw biogas. This value is greater than the value reported by Yousef *et al.;*(2016). This might be as a result of higher percentage purity of both UBG and LC attained as well as water being one of the components of the feed in this work which was not considered in their work. With LD-SYS, the total power consumed (8379.82 kW) is greater than that of D-SYS. The need for the process to be operated at higher pressure and compression of UBG in order to meet up with pipeline flowing pressure might be responsible for the higher value. The amount of power consumed in raw biogas pre-compression, auxiliary refrigeration cycle and UBG compression were 65.49%, 15.33% and 19.18% respectively. The specific energy consumption was estimated to be 0.38 kWh/Nm^3 raw biogas. It is noted from this analysis that the highest percentage of power consumed in compression process in both cases may be attributed to the proposed process routes made to be operating at high pressure. The power consumption in refrigeration cycle can be minimized by improving its coefficient of performance. It can be concluded that the better alternative using power consumption and removal efficiency as a factor is D-SYS.

3.3. Methane Slip

The contribution of methane gas to global warming is so enormous that it requires serious attention when choosing a biogas upgrading technology (Sun *et al.*, 2015). Methane losses are specified by gas treatment service to be less than 2% and can expected to be below 0.5% in an optimized plant (Bauer *et al.,* 2013). As shown in Table 4, the methane loss in D-SYS and LD-SYS were calculated to be 0.01% and 1.28% respectively which are well below the specified limit. However, there might be need to further lower methane loss especially in LD-SYS due to stricter environmental regulation. This can be accomplished by thermal or catalytic oxidation of methane.

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Table 4: Removal efficiency and power consumption of the systems

	D-SYS	LD-SYS
% CO_2 capture	96.02	89.62
% CH_4 loss	0.01	1.28
Power consumption		
Auxiliary refrigeration cycle (kW)	2151.58	1607.82
Raw biogas pre-compression (kW)	4681	5487
Liquid CO_2 pumping (KW)	45.13	-
Upgraded biogas compression (kW)	-	1285
Total power consumed (kW)	6877.71	8379.82
Specific energy consumption (kWh/Nm^3)		
Raw biogas	0.312	0.38
Upgraded biogas	0.508	0.604

3.4. Exergy Analysis

The exergy analysis of the simulation models was performed in order to investigate the efficiency of the models and then make comparison between the two alternatives. Tables 5 and 6 summarize the results of exergy analysis of D-SYS and LD-SYS respectively. The largest portion of exergy input into the systems is contributed by raw biogas as shown in Tables 5 and 6. This could be ascribed to presence of hydrocarbon (CH_4) in the feed. The exergy efficiencies of D-SYS and LD-SYS were calculated to be 95.59% and 92.80% respectively. The high values of efficiency could be attributed to chemical exergy of CH_4 included in the formulation of exergy efficiency. However, the exergy efficiency of the systems could still be improved by reducing exergy losses. It is indicated in the tables that external exergy losses is less than internal exergy losses. This could be interpreted as exergy efficiency improvement could be accomplished by modification of the flow sheets rather than recycling the output streams. It is also indicated in the Tables that 4.44% of total exergy input was lost in D-SYS while 7.22% was lost in LD-SYS. This is due to greater number of coolers in LD-SYS which is one of the major sources of inefficiency in the systems. The comparison of the two systems using exergy analysis shows that D-SYS is more efficient and environmentally friendly than LD-SYS.

Table 5: Exergy analysis of D-SYS

	Exergy flow rate (kW)	% of total Exergy inlet (kW)
Raw biogas (kW)	140337.63	95.36
Compression of raw biogas (kW)	4681	3.18
Refrigeration cycle (kW)	2151.58	1.46
LC pumping (kW)	45.13	0.03
Compression of UBG (kW)	-	-
Total exergy in (kW)	147170.21	-
Internal exergy loss (kW)	5076.57	3.45
External exergy loss (kW)	1451.56	0.99
Total exergy loss (kW)	6528.13	4.44
Exergy Efficiency	95.59	-

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Table 6: Exergy analysis of LD-SYS

	Exergy flow rate (kW)	% of total Exergy inlet (kW)
Raw biogas (kW)	140337.63	94.37
Compression of raw biogas (kW)	5487	3.69
Refrigeration cycle (kW)	1607.82	1.08
LC pumping (kW)	-	-
Compression of UBG (kW)	1285	0.86
Total exergy in (kW)	148717.45	-
Internal exergy loss (kW)	7739.08	5.2
External exergy loss (kW)	3000.79	2.01
Total exergy loss (kW)	10739.87	7.22
Exergy Efficiency	92.80	-

Furthermore, identifying the main source of internal exergy losses is a prerequisite for efficiency improvement. To achieve this, a detailed exergy analysis of equipment in the flow sheets was carried out. The equipment was classified into groups according to their functions: separation, cooling, refrigeration, mixing and compression. It should be noted that pump and valves were not considered for the classification. The contribution of these processes to total internal exergy losses in D-SYS and LD-SYS is demonstrated in Figures 3 and 4. As shown in Figures 3 and 4, noticeable irreversibilities take place during cooling and refrigeration process. The losses are purely physical exergy because chemical exergy is not affected in any of these processes except separation. The great exergy loss might be as a result of wide temperature difference. It is also revealed in the Figures that physical exergy accounts for more than 80% of the irreversibilities in the two systems and these losses can be minimized by technology development.

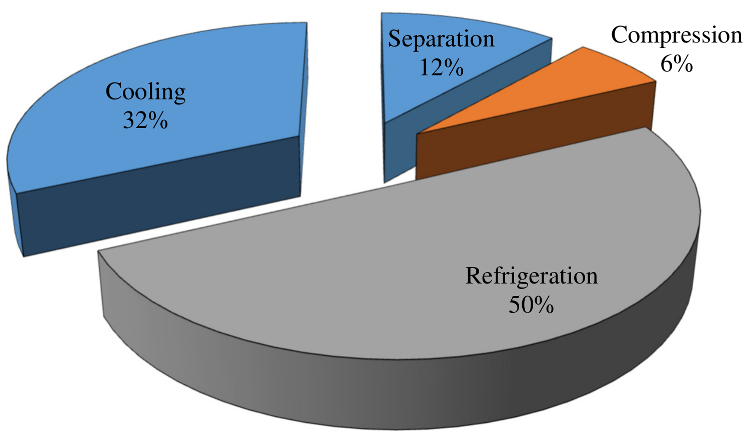

Figure 3: Internal exergy losses distribution in D-SYS

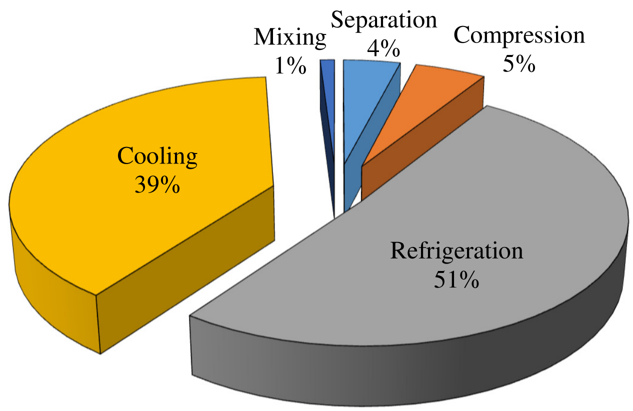

Figure 4: Internal exergy losses distribution in LD-SYS

4. CONCLUSION

Both the distillation system and liquefaction combined with desublimation system are suitable for biogas upgrading combined with carbon capture. To know which one is a better alternative, simulation models with equal capacity of the two systems were carried out using *Aspen HYSYS*. The product streams are in accordance with the previous researches. To compare the two alternatives, gas purity, energy consumption and removal efficiency, methane slip and exergy analysis were used as standard measures. Having realized from the simulation results that D-SYS displays higher gas purity, higher removal efficiency, lower energy consumption, lower methane loss and higher exergy efficiency, D-SYS is considered as a better alternative. Moreover, detailed exergy analysis performed on various equipment in the flow sheet shows that more than 80% of total irreversibilities occurs during cooling and refrigeration processes and these losses can be minimized by technology development. It is recommended that a more efficient thermodynamic system be designed by reducing exergy losses in each of the component considered in the model. Future work should consider cost estimation as one of the yardsticks of comparison.

5. CONFLICT OF INTEREST

There is no conflict of interest associated with this work.

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