



UNIVERSITI
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**Optimal Operation Of Multiple Cryogenic Bed Network For
Purification Of Malaysian Natural Gas**

By

Lam Pei Shin

Dissertation submitted in partial fulfillment of the requirements for the

Bachelor of Engineering (Hons)

(Chemical Engineering)

August 2014

Universiti Teknologi PETRONAS

Bandar Seri Iskandar

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CERTIFICATION OF APPROVAL

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Approved by,

(Prof. Dr. Saibal Ganguly)

UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

May 2014

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

Lam Pei Shin

ABSTRACT

Natural gas processing consists of separating all of the various hydrocarbons and impurities from the wellhead gas, to produce what is known as 'pipeline quality' dry natural gas. The present study investigates the optimal conditions for the multiple cryogenic packed beds network which separates water, carbon dioxide and heavy hydrocarbons from high pressure natural gas. The working principle of multiple cryogenic packed beds network is based on the difference in desublimation temperature of each component in natural gas. In general, the separation process involves three cycles, namely cooling cycle, capture cycle and regeneration cycle. During cooling cycle, refrigerant is used to cool the packed bed. When the packed bed is cooled to a desired temperature, which is below the desublimation temperature of the component to be removed, for instance carbon dioxide, natural gas feed is introduced into the packed bed and the capture cycle began. When the bed reaches its saturation point and is no longer efficient in capturing carbon dioxide, the refrigerant flow is cut off in order to regenerate the packed bed by utilizing hot air or hot carbon dioxide flow to vaporize the solid carbon dioxide that formed on the solid packing surface. In present study the optimal temperature and pressure conditions for natural gas processing are explored. For natural gas feed of 40% carbon dioxide, multiple cryogenic packed beds network with optimal pressure and temperature combinations is able to produce methane gas with 92% purity.

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CHAPTER 1

INTRODUCTION

1.1. Background of Study

Natural gas is a fossil fuel that is well-known for being one of the cleanest energy resources and plays an important role in contributing to the world's energy supply. With the rise of demand for clean energy, high impurity wells are explored worldwide so as to supply the increasing market demand. The wellhead natural gas contains many impurities, in this study only water and carbon dioxide (CO₂) is considered. The water content in natural gas not only causes the corrosion but also forms solid hydrocarbon which can plug the transmission line. Therefore, the allowable water content in natural gas ranges from 65 to 110 mg per standard m³. The presence of CO₂ in natural gas decreases the calorific value and also cause the corrosion in pipelines (Rufford et al., 2012; Darman & Harun, 2006). Due to this reason, the CO₂ content in natural gas pipeline transmission must be reduced to less than 3% (Hubbard, 2010).

According to Burgers et al. (2011), gas resources with CO₂ composition between 15%-80% is consider as sour gas resources that has high CO₂ content. Some of the Malaysian natural gas reserve has CO₂ content that can goes up to 80%.and therefore is considered as sour gas resource. The following tables summarize all the high CO₂ gas fields in Malaysia (Darman & Harun, 2006).

TABLE 1. Penisular Malaysia gas fields CO₂ contents

Penisular Malaysia		
Holder	Field	CO ₂ content
PETRONAS	Bujang	66%
PETRONAS	Sepat	60%
PETRONAS	Noring	60%
PETRONAS	Inas	60%
PETRONAS	Tangga Barat	32%
PCSB	Ular	50%
PCSB	Gajah	50%
PCSB	Bergading	40%
PCSB	Palas NAG	46%

TABLE 2. Sarawak gas fields CO₂ contents

Sarawak		
Holder	Field	CO ₂ content
PETRONAS	K5	70%
PETRONAS	J5	87%
PETRONAS	J1	59%
PETRONAS	T3	62%
PETRONAS	Tenggiri Mm.	47%

There are several dehydration processes namely absorption, adsorption, gas permeation and refrigeration. Dehydration processes which are widely used are absorption and adsorption. On the other hand, CO₂ could be removed using methods such as adsorption, absorption, membrane separation and cryogenic separation. However, cryogenic separation is not comprehensively investigated, due to the perception of high energy cost. The advantage of cryogenic separation is that no chemical reaction is involved as well as has minimum footprint for offshore application. Recent finding shows that cryogenic separation is capable to separate water and CO₂ from natural gas at optimum energy requirements (Abulhassan et. al, 2014).

1.2. Problem Statement

Natural gas must undergo processing to remove impurities in order to be used as a fuel. With the depletion of sweet gas reserves, Malaysian natural gas reserves that have high contents of CO₂ (up to 80%) need to be explored in order to meet the global demand. The presences of water contents and CO₂ in natural gas not only reduce the heating value but also cause pipelines corrosion and plugging.

Multiple cryogenic packed beds that remove both water and CO₂ from raw natural gas through desublimation and freezing at atmospheric pressure has been proposed by Abulhassan et al. (2014). However, the optimal performance which is closely related to the operating pressure and temperature of multiple cryogenic beds network at high pressure is yet to be investigated in details.

The optimal operating conditions for simultaneous dehydration and CO₂ removal are investigated in present study by using multiple cryogenic packed beds.

1.3. Objectives

- i) To identify the sequence of multiple packed beds used for the separation of CO₂ and heavy hydrocarbons from natural gas
- ii) To identify optimal operating conditions with
 - Minimum methane loss
 - Minimum energy usage
 - Maximum separation efficiency

1.4 Scope of Study

- i) Simulation of high pressure flow line from natural gas pipeline at cryogenic condition
- ii) Investigate optimal operating conditions using branch and bound logic

CHAPTER 2

LITERATURE REVIEW

Cryogenic separation process can be classified into three categories, namely conventional, non-conventional and hybrid cryogenic separation process. Non-conventional cryogenic separation process encourages the formation of solid CO₂ while conventional cryogenic separation process avoids the formation of solid CO₂. The hybrid cryogenic separation process includes both conventional and non-conventional cryogenic separation process.

The conventional cryogenic separation process, developed by Ryan/Holmes, is an extractive cryogenic distillation network by adding n-Butane in the condenser of the distillation column to avoid carbon dioxide solidification (Holmes et al., 1982). Liquid carbon dioxide can be obtained in this separation process. It is considered as one of the most capable method of high content CO₂ separation from CH₄. However, cryogenic distillation is a process that requires huge amount of energy and the solid CO₂ formed in the distillation column due to the vapour liquid equilibrium need to be handled carefully.

One example for the non-conventional cryogenic separation process is Controlled Freeze ZoneTM (CFZTM). CFZTM is a technology invented by ExxonMobil in 1983 that separates CO₂ and H₂S from natural gas through controlled freezing and re-melting of CO₂. With this technology, gases with wide range of CO₂ and H₂S content can be managed easily while sales quality gas could be produced at a lower cost.

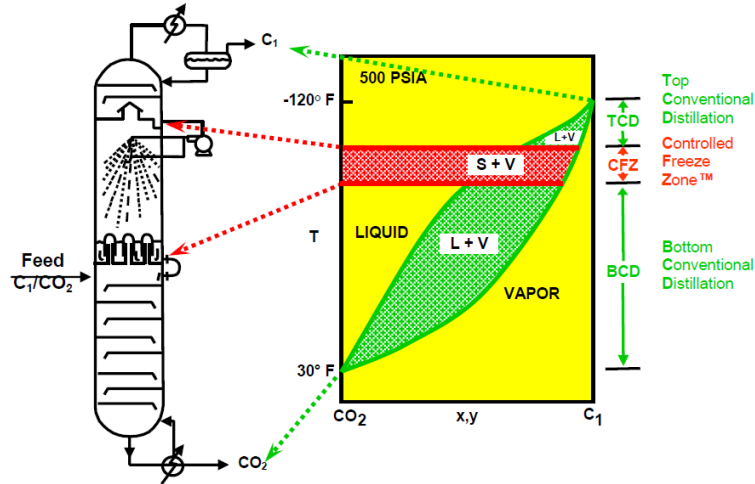


FIGURE 1. CFZ™ Process (Northrop & Valencia, 2009)

FIGURE 1 shows that the distillation column is divided into three sections, upper rectification section, CFZ™ chamber and a lower stripping section. The fed natural gas vapor flows up the cryogenic distillation column and contacts with the cold liquid sprayed through the nozzle. In order to prevent plugging problem, cold liquid sprayed through the nozzle is close to solidification condition so that the solidification process does not take place outside of the freezing zone. Light components such as methane vaporize during the liquid droplets fall as temperature gets higher when going down the column. As a result, the concentration of residual CO₂ in the liquid droplets increases. CO₂ freezes out and form pure CO₂ solid when the residual concentration in the liquid droplets is high enough. Solidified CO₂ falls onto a melt tray which is kept above the solidification temperature leads to the formation of liquid CO₂ that will then be sent to the stripping section. This is done in order to further recover the valuable light components of natural gas. At the end of the process, the removal of pure solid CO₂ enables the production of methane-rich vapor. It is concluded that CFZ™ is able to reduce capital cost, sale gas recompression cost, acid gas injection cost as well as eliminate the need to use solvents or additives (Kelley, Valencia, Northrop, & Mart, 2011; Northrop & Valencia, 2009; Valencia & Mentzer, 2008). Nevertheless, this technology requires huge size equipment and there is operational issue due to the solidification of CO₂.

The purification of natural gas which is of high pressure and high content of CO₂ is simulated by Maqsood et. al. (2014). In this hybrid cryogenic separation process, Maqsood et. al. use both cryogenic packed beds and distillation columns to purify high pressure natural gas with high content of CO₂. It is also reported that the use of multiple cryogenic packed bed that has been proposed by Abulhassan et al. (2014) allows the equipment size to be reduced as well as abolishes the use of butane as an extractive distillation additive.

Another example of non-conventional cryogenic separation involves the separation of CO₂ from flue process. Clodic and Younes (2002, 2005) reported that CO₂ was captured through anti-sublimation, by forming solid on the surface of heat exchangers. The solid CO₂ is then removed from the heat exchanges in the form of liquid by raising the pressure during regeneration cycle. Although the energy requirement reported in this process is comparatively lower than the CO₂ absorption process, however due to the accumulation of solid CO₂ on the surfaces of heat exchangers, there is a decrease in process efficiency. Also, there is a limitation of this process, which is the feed gas must not contain water contents so as to avoid plugging problem.

Tuinier et. al. (2010, 2011) used dynamic packed bed to improve the cryogenic CO₂ capture process from flue gases. This process consists of three cycles, namely cooling cycle, capture cycle and recovery cycle. Packed bed temperature is cooled down to below the desublimation temperature of CO₂ during cooling cycle and the capture cycle is started once the flue gas is introduced into the packed bed. Although Tuinier et al. reported that the difference in both dew and desublimation temperature of the flue gas components allows the refrigerated packed bed to separate water, CO₂, and permanent gases effectively at atmospheric pressure. However, the process have not been applied to natural gas which is of high CO₂ content and high pressure.

Cryogenic separation of natural gas using dynamic packed bed has been studied by Syahera (2012). In the study, Syahera concluded that the dynamic packed bed has high potential to capture CO₂ from natural gas with high CO₂ concentration, as the simulation result shows that higher CO₂ concentration allows more solid CO₂ to be deposited.

Abulhassan et. al (2014) reported that optimal separation and energy efficiencies could be achieved using a counter current switched packed bed. Energy requirement of the cryogenic packed bed is significantly lower than the cryogenic distillation process, as the former requires 660 to 810 kJ/kg CO₂ energy while the latter needs 1472 kJ/kg CO₂ to purify natural gas with 70% of CO₂ content. However, both cryogenic packed bed and cryogenic distillation process have similar energy requirements when the feed has low CO₂ concentration. This observation proves that the cryogenic packed bed is efficient for the purification of natural gas with high contents CO₂.

With the dynamic packed bed proven to be suitable to remove CO₂ from natural gas, Karen (2013) introduced the multiple beds concept for simultaneous dehydration and CO₂ removal. The effect of several parameters, including packed bed length, initial bed temperature and feed flow rate to the dehydration and CO₂ removal processes were studied using multiple cryogenic packed beds. Karen concluded that longer packed bed, lower initial bed temperature and higher flow rate allow more water to be removed during the dehydration and CO₂ removal process. Ali (2013) extended the previous work of Karen by introducing hydrocarbon in the feed. Both simulation and experimental studies were conducted to investigate the effect of bed temperature, feed flow rate and inlet CO₂ concentration on CO₂ removal process using multiple cryogenic packed beds. Ali reported that lower bed temperature and higher feed flow rate allow more CO₂ to be deposited as solid, while higher inlet CO₂ concentration results in longer bed saturation time.

Most of the CO₂ removal process involved flue gas that has very low CO₂ content. Although dynamic packed bed and multiple cryogenic packed beds have been proposed to purify Malaysian natural gas that has high CO₂ concentration, however, the optimal conditions for high feed gas pressure is yet to be investigated. Present study attempts to bridge the gap in existing literature on cryogenic packed beds by investigating the optimal condition of multiple cryogenic packed beds that purify Malaysian natural gas at high pressure.

CHAPTER 3

METHODOLOGY

3.1 Project Flow Chart

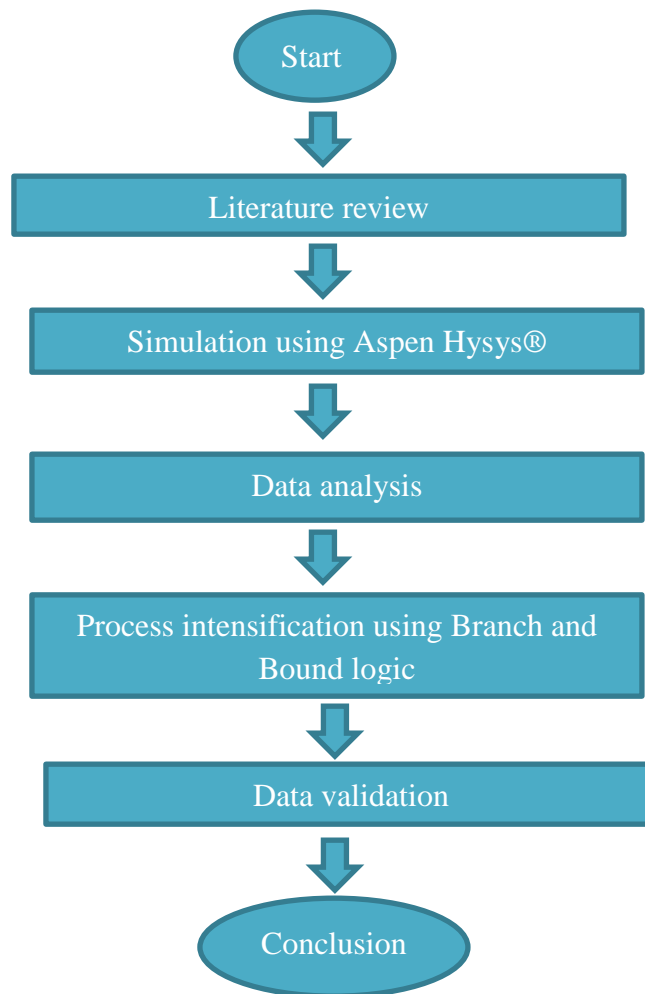


FIGURE 2. Frame Work.

3.2 Gantt Chart And Key Milestone

Final Year First Semester

TABLE 3. Gantt Chart and Key Milestone for Final Year First Semester

No.	Details/Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14	Study week
1	First meeting with supervisor		■													
2	Preliminary research work and preparing proposal		■	■	■	■	■	■								
3	Submission of extended proposal to supervisor							■								
4	Proposal defense								■							
5	Commencement of experimental work							■	■	■	■	■	■			
6	Submission of Interim Draft Report													■		
7	Submission of Final Interim Report														■	
8	Submission of marks by supervisor															■

Final Year Second Semester

TABLE 4. Gantt Chart and Key Milestone for Final Year Second Semester

No.	Details/Week	1	2	3	4	5	6	7	8	9	10	11	12	13	14	Study week
1	Project Work Continues	■	■	■	■	■	■	■								
2	Submission of Progress Report							■								
3	Project Work Continues								■	■	■	■	■			
4	Pre-SEDEX										■					
5	Submission of Draft Final Report											■				

6	Submission of Dissertation (soft bound)																			
7	Submission of Technical Paper																			
8	Viva																			
9	Submission of Project Dissertation (Hard Bound)																			

Process

Suggested Milestone

3.3 Methodology

3.3.1 Process Concept

Single Packed Bed

The cryogenic separation involves component separation that is based on the difference of freezing and desublimation points. Three different cycles, namely cooling cycle, capture cycle and recovery cycle are involved during the separation process. Refrigerant is used in the cooling cycle to bring the packing material temperature below the freezing point of the component to be separated. The refrigerant is either in direct contact with the packing material or being introduced into the jacket. When the packing reaches desire temperature, cooling cycle is completed and the capture cycle starts once the feed gas is introduced into the packed bed. Component with higher freezing point freezes on the surface of the packing while other components with lower freezing temperature flow through the packed bed without any phase change. When the bed reaches its saturation point, the feed supply is cut off and the bed undergoes recovery cycle to remove the frost components.

Multiple Packed Bed

The multiple packed bed system is a series of packed beds operating at different pressure and temperature to remove both water and CO₂ in different beds. As the presence of water in raw natural gas causes plugging problem in the transportation

pipelines, therefore it is utmost important to remove water before the CO₂ removal. Thus, in the multiple cryogenic packed bed network, the first focus is the water removal until the pipeline specifications is reached, while the other packed beds will focus on CO₂ removal. Based on this component removal concept, a general schematic diagram is illustrated in FIGURE 3. Stream outlets on top of the packed beds such as 3, 6, 8, 20 and 22 signify that the product is vapor whereas stream outlets below the packed beds such as 4, 7, 10, 19 and 21 signify that the product is in liquid phase. As for the stream outlets below the packed beds, for example stream 2, 5, 9, 11 and 12, these are the components that will be separated from the natural gas in the form of solid phase.

Proposed Multiple Packed Bed Schematics

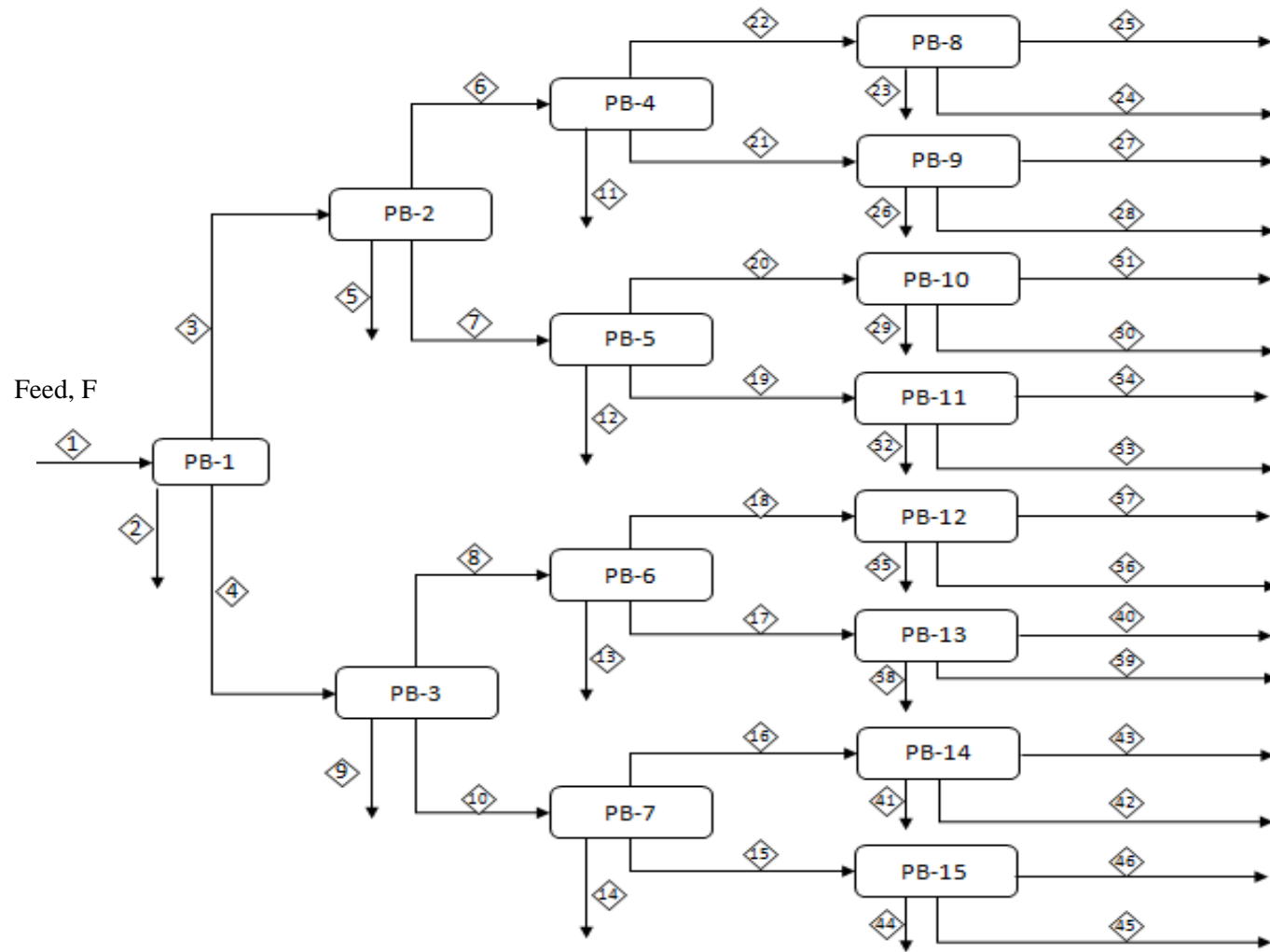


FIGURE 3. Multiple Cryogenic Packed Beds Network Synthesis

As aforementioned, the first step in natural gas processing is dehydration to prevent the pipelines plugging problem and hydrates formation. Thus, in the first packed bed, the pressure and temperature of the packed bed need to be operated in a way so that only water vapor in the raw natural gas is solidified. Meanwhile, the low temperature also leads to the condensation of heavy hydrocarbons into liquid phase. Through this solid-liquid-vapor separation, dry natural gas with higher purity leaves the packed bed in the form of vapor. From the general schematic diagram, it is clearly shown that the vapor and liquid products from the first packed bed separation will then become the feed for the next packed beds, packed bed 2 and packed bed 3 respectively. Since water has been removed in packed bed 1, the operating pressure and temperature of packed bed 3 whom feed consists of mostly heavy hydrocarbon will be adjusted in such a way that only CO₂ desublimates. Packed bed 2 operating conditions will depend on the composition of packed bed 1 vapor stream. If the stream meets the pipeline specification of water contents than the bed 2 will act as CO₂ removal unit otherwise dehydration unit.

Thermodynamic Representation of Multiple Packed Beds

Pressure and temperature are the two variables that need to be handled in order to achieve desired separation. For example, packed bed 1 focuses on water removal and therefore the operating pressure and temperature for packed bed 1 needs to be adjusted in such a way that maximum water removal with minimum methane loss. As for packed beds that focus on CO₂ removal, the operating pressure and temperature need to be adjusted in such a way that maximum CO₂ is removed as solid with minimum methane losses.

The pressure-temperature diagram (PT diagram) as shown in FIGURE 4 shows the freezing point of individual components in the natural gas. From FIGURE 4, it is also elucidated that at atmospheric pressure, CO₂ has high freezing point (-78 °C) while hexane that has the highest freezing point among other hydrocarbons starts to desublimates at -100 °C. In order to have effective separation and minimum hydrocarbon loss, the study is conducted in the temperature range between -100 °C to 0 °C and pressure range of 0 bar to 80bar.

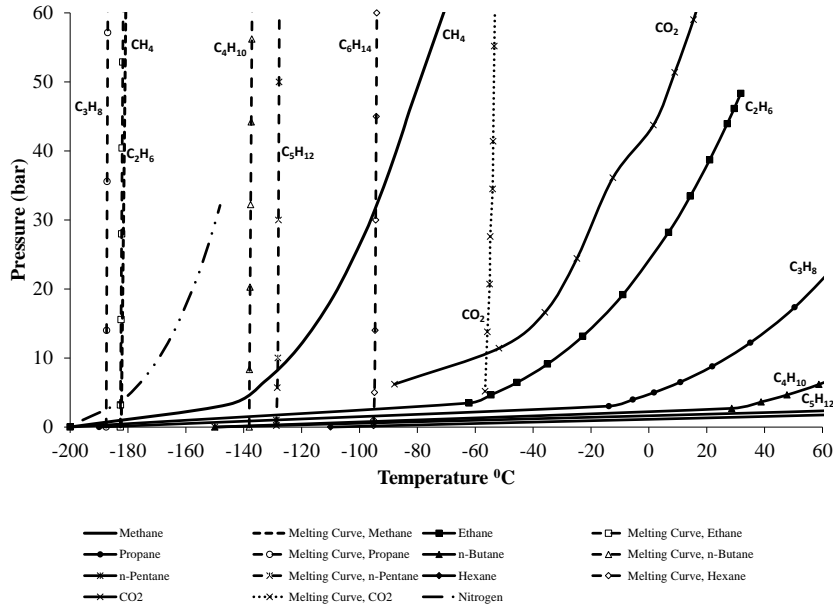


FIGURE 4. PT Diagram for Natural Gas Components

3.3.2 Strategy for Selection of Optimal Operating Conditions for Multiple Packed Beds

The investigation of multiple packed beds' optimal operating conditions consists of two steps, namely the simulation process and process optimization. In the simulation process, all possible operating conditions were simulated using Aspen HYSYS® and the simulation results obtained were tabulated. After the simulation process, node-edge diagrams are developed before sensitivity studies were carried out to optimize the multiple packed beds' operating conditions. The details of these two steps are given in nest sections.

Simulation Process

In order to investigate how the pressure and temperature combination affect the separation efficiency, process analysis has been carried out using Aspen HYSYS® with Peng-Robinson fluid package. Simulations results were obtained based on the general schematic diagram. Vapor and liquid streams compositions of each pressure and temperature combinations are recorded in a process analysis table. This process analysis table is then analyzed and utilized in order to pre-determine the suitable pressure-temperature combination for each packed bed.

Aspen HYSYS® provides good estimation for the vapor and liquid stream compositions while FIGURE 5 is used to predict the solid formation of carbon dioxide during the cryogenic separation. The natural gas composition used for the present study is given in TABLE 5.

TABLE 5. Composition of Feed Natural Gas (Engineer, 2004).

Components	Mole Fraction
CH ₄	0.440
C ₂ H ₆	0.050
C ₃ H ₈	0.027
i-C ₄ H ₁₀	0.010
n-C ₄ H ₁₀	0.010
i-C ₅ H ₁₂	0.010
n-C ₅ H ₁₂	0.001
C ₆ H ₁₄	0.001
C ₇ H ₁₆	0.001
C ₈ H ₁₈	0.001
H ₂ O	0.040
CO ₂	0.400
N ₂	0.010

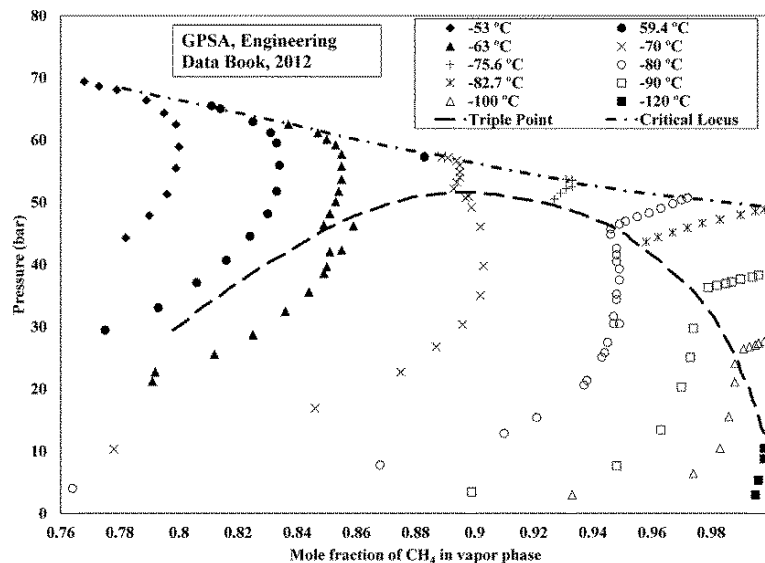


FIGURE 5. Dew Point and Frost Data for CO₂ and CH₄

FIGURE 5 shows the region where solid CO₂ formation is possible, which is the region below the parabolic curve. The region covers an operating pressure

that ranges from approximately 1 bar to 55 bar and operating temperature range that is between 60°C and -120°C. The FIGURE 5 also provides an insight of how decreasing temperature increases the purity of methane in vapor phase. The sample simulation results for dehydration with different operation conditions are shown in TABLE 6. Similar tables were generated for CO₂ removal with varying operating conditions are as shown in the Appendix.

TABLE 6. Sample process analysis table for dehydration of natural gas

Feed		Gas					
Pressure (bar)	80.00	80.00					
Temp (°C)	25.00	0.00	-2.00	-5.00	-10.00	-15.00	-20.00
CH ₄	440.000	386.06	367.57	332.41	253.11	145.87	1.98
C ₂ H ₆	50.000	39.37	36.41	31.29	21.53	11.05	0.13
C ₃ H ₈	27.000	18.79	16.93	13.95	8.93	4.29	0.05
i-C ₄ H ₁₀	10.000	6.18	5.46	4.36	2.67	1.23	0.01
n-C ₄ H ₁₀	10.000	5.84	5.11	4.03	2.42	1.11	0.01
i-C ₅ H ₁₂	10.000	4.98	4.27	3.29	1.90	0.85	0.01
n- C ₅ H ₁₂	1.000	0.47	0.40	0.30	0.17	0.08	0.00
C ₆ H ₁₄	1.000	0.36	0.30	0.22	0.12	0.05	0.00
C ₇ H ₁₆	0.500	0.13	0.11	0.08	0.04	0.02	0.00
C ₈ H ₁₈	0.500	0.10	0.08	0.06	0.03	0.01	0.00
H ₂ O	40.000	0.23	0.19	0.14	0.07	0.03	0.00
CO ₂	400.000	321.63	298.16	256.91	176.99	90.75	1.08
N ₂	10.000	9.14	8.81	8.15	6.54	4.04	0.06
Feed		Liquid					
Pressure (bar)	80.00	80.00					
Temp (°C)	25.00	53.94	72.43	107.59	186.89	294.13	438.02
CH ₄	440.000	10.63	13.59	18.71	28.47	38.95	49.87
C ₂ H ₆	50.000	8.21	10.07	13.05	18.07	22.71	26.95
C ₃ H ₈	27.000	3.82	4.54	5.64	7.33	8.77	9.99
i-C ₄ H ₁₀	10.000	4.16	4.89	5.97	7.58	8.89	9.99
n-C ₄ H ₁₀	10.000	5.02	5.73	6.71	8.10	9.15	9.99
i-C ₅ H ₁₂	10.000	0.53	0.60	0.70	0.83	0.92	1.00

n- C ₅ H ₁₂	1.000	0.64	0.70	0.78	0.88	0.95	1.00
C ₆ H ₁₄	1.000	0.37	0.39	0.42	0.46	0.48	0.50
C ₇ H ₁₆	0.500	0.40	0.42	0.44	0.47	0.49	0.50
C ₈ H ₁₈	0.500	39.77	39.81	39.86	39.93	39.97	40.00
H ₂ O	40.000	78.37	101.84	143.09	223.01	309.25	398.92
CO ₂	400.000	0.86	1.19	1.85	3.46	5.96	9.94
N ₂	10.000	53.94	72.43	107.59	186.89	294.13	438.02
cooling duty (MW)		0.762655	0.849286	0.985026	1.220813	1.460335	1.698447

Process Optimization

i) Development of Node-Edge Diagram

Based on the pre-determined suitable pressure-temperature combinations obtained from the process analysis table, a node-edge diagram as shown in FIGURE 6 is prepared so as to optimize the temperature and pressure variables using depth-first branch and bound method.

In the context of branch and bound optimization method, packed bed 1 as shown in the node-edge diagram serves as a root node of the solution tree. As the path going down, two terminal nodes are created for each node through the branching steps.

Meanwhile, the numbers shown on the node-edge diagram also symbolizes the packed bed number, however with a few exceptions. For example, number 0 that refers to raw natural gas storage tank, number 16 that symbolizes water storage tank, number 17 that is being referred as CO₂ storage tank, number 18 that is denoted as storage tank for methane with high purity, storage tank number 19 that is used to store methane with small amount of CO₂ and last but not least, number 20 that symbolizes storage tank for methane with low purity.

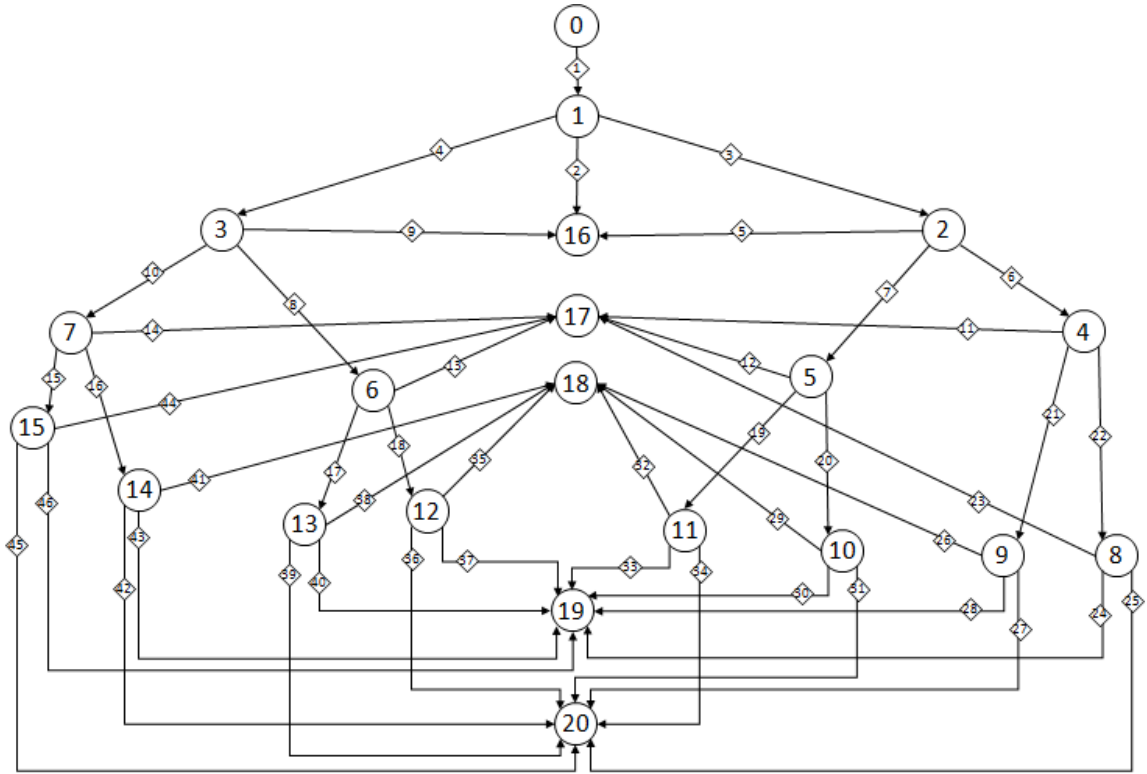


FIGURE 6. Node-Edge Diagram of Multiple Packed Beds for Dehydration and CO₂ Removal from Natural Gas

In this node-edge diagram, it is also shown that feed natural gas coming from the feed storage tank is introduced into packed bed 1, where cryogenic separation takes place and the solid ice formed will be recovered and sent to water storage tank numbered 16. Thus, in this case packed bed 1 is known as the dehydration bed. Meanwhile, the heavy hydrocarbons that have condensed into liquid state will be sent to packed bed 3 while vapor product of packed bed 1 will be undergo another stage of cryogenic separation at packed bed 2. If there is water vapor remaining in the vapor product of packed bed 1, dehydration process will again take place in packed bed 2 and the ice will again be recovered and sent to water storage tank numbered 16. However, if water is completely removed in packed bed 1, the subsequent packed beds will be known as CO₂ removal bed that removes solid CO₂ to the CO₂ storage tank numbered 7. The vapor and liquid product will then become feed for the next two packed beds until there is no more proposed packed bed for separation. The pure methane product will be sent to storage tank numbered 18 whereas the impure methane product will be sent to storage tank numbered 20.

ii) Development of Sensitivity Analysis

With the suitable pressure-temperature combinations obtained from the process analysis table being used as nodes in the node-edge diagram where depth-first branch and bound optimization method applies, sensitivity analysis is also carried out so as to provide an insight of how a small change in pressure and temperature affects the cryogenic separation efficiency of water and CO₂. Therefore, this sensitivity analysis is essential and aids in decision making. Apart from that, this sensitivity analysis also helps in the investigation of relationship between the pressure-temperature combination and energy required for the refrigeration.

The sensitivity analysis is carried out by manipulating the either the operating pressure or temperature of the selected suitable pressure-temperature combination. For example, -5 °C is the selected temperature for packed bed 1 at an operating pressure of 80 bar. The separation efficiency at 80 bar and -5 °C is investigated. The investigation is then repeated for 80 bar and -10 °C as well as 80 bar and -15 °C if temperature is the variable to be investigated. With this strategy, the effect of change in temperature on the separation efficiency is known and decision such as whether to maintain or lower the operating pressure so as to achieve the desired separation could be made. Similarly, if pressure is the variable to be investigated in the sensitivity analysis, the investigation will be repeated for 70 bar and -5 °C as well as 60 bar and -5 °C.

Calculation of Objective Function

By choosing different suitable pressure-temperature combinations obtained from the process analysis table for each packed bed, many schemes can be formed. In order to determine which scheme provides the optimum pressure-temperature combination for all packed beds, an objective function that serves to be a comparison value is required. The formula below shows the calculation for the objective function.

Objective Function

$$\text{Profit, } \phi \text{ (\$/cycle)} \phi = (\phi_1 + \phi_2) - (\phi_3 + \phi_4) - \phi_5$$

Where ϕ_1 =Methane product with high purity

ϕ_2 = Low grade methane product which is of low purity

ϕ_3 = Cost of energy required

ϕ_4 = Cost of methane loss

ϕ_5 = equipment cost

TABLE 7 shows the price of each component in the natural gas that is involved in the revenue calculation.

TABLE 7. Price of Components

Component	Price (\$/kg)
Methane	0.246
Ethane	0.25
Propane	0.6208
i-Butane	1.0231
n-Butane	1.0231
i-Pentane	1.1206
n- Pentane	1.1206
Hexane	1.1206
Heptane	1.1206
Octane	1.1206
Water	
Carbon dioxide	0.04
Nitrogen	

Flow sheet

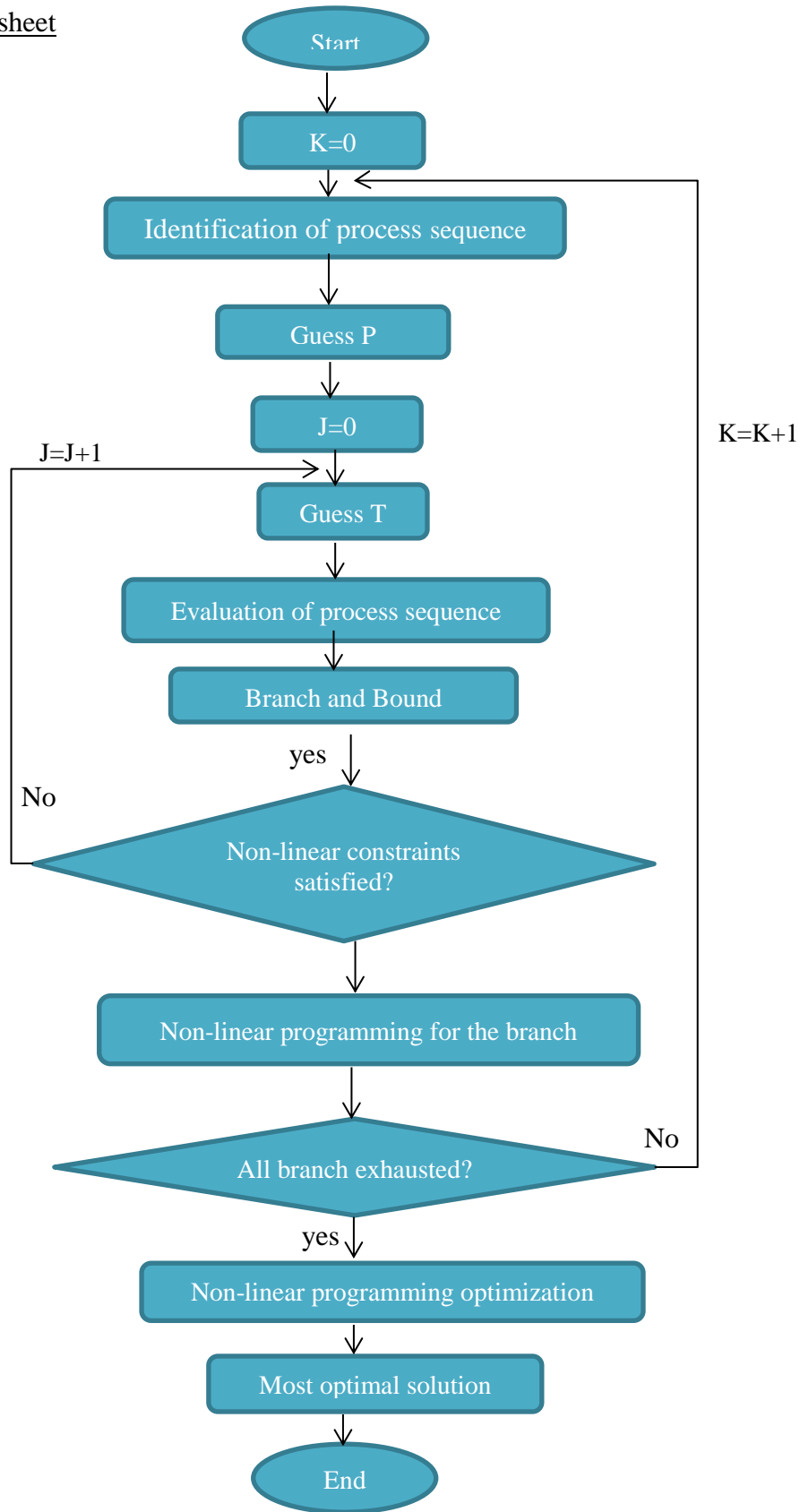


FIGURE 7. Flow sheet of Project 21

CHAPTER 4

RESULTS AND DISCUSSION

4.1 Simulation Results

The simulation results for both dehydration and CO₂ removal are presented in this section for natural gas feed. The simulation results can be used to estimate the composition of vapor, liquid and solid streams after the cryogenic separation process. However, the solidification of components were further investigated by using thermodynamic data. The composition of vapor stream after the cryogenic separation in packed bed 1 is as shown in TABLE 8 while TABLE 9 shows the composition of liquid stream after the cryogenic separation in packed bed 1. It is elucidated from all tables that 40% CO₂ (TABLE 5) containing natural gas at 80 bar and 25 °C is fed into packed bed 1 which has an operating pressure of 80 bar and different initial bed temperatures, ranging from -20 °C to 0 °C. With a lower initial bed temperature, it was observed that by decreasing bed temperature more water is removed in form of solid. The second effect of lower bed temperature was observed of methane liquefaction, as the bed temperature decrease more methane goes in liquid stream which further effect the product purity. TABLE 8 shows that the cooling duty is inversely proportional to the initial bed temperature. As the initial bed temperature decreases, the cooling duty of the cooler increases as more energy is required to further cool down the packed bed.

TABLE 8. Composition of Vapor Stream after Cryogenic Separation in Packed Bed 1

Feed		Vapor					
Pressure (bar)	80.00	80.00					
Temp (°C)	25.00	0.00	-2.00	-5.00	-10.00	-15.00	-20.00
CH ₄	440.00	386.06	367.57	332.41	253.11	145.87	1.98
C ₂ H ₆	50.00	39.37	36.41	31.29	21.53	11.05	0.13
C ₃ H ₈	27.00	18.79	16.93	13.95	8.93	4.29	0.05
i-C ₄ H ₁₀	10.00	6.18	5.46	4.36	2.67	1.23	0.01
n-C ₄ H ₁₀	10.00	5.84	5.11	4.03	2.42	1.11	0.01
i-C ₅ H ₁₂	10.00	4.98	4.27	3.29	1.90	0.85	0.01
n- C ₅ H ₁₂	1.00	0.47	0.40	0.30	0.17	0.08	0.00
C ₆ H ₁₄	1.00	0.36	0.30	0.22	0.12	0.05	0.00
C ₇ H ₁₆	0.50	0.13	0.11	0.08	0.04	0.02	0.00
C ₈ H ₁₈	0.50	0.10	0.08	0.06	0.03	0.01	0.00
H ₂ O	40.00	0.23	0.19	0.14	0.07	0.03	0.00
CO ₂	400.00	321.63	298.16	256.91	176.99	90.75	1.08
N ₂	10.00	9.14	8.81	8.15	6.54	4.04	0.06
Total flow (mole/h)	1000.00	793.28	743.79	655.20	474.52	259.37	3.33
cooling duty (MW)		0.76	0.85	0.99	1.22	1.46	1.70

TABLE 9. Composition of Liquid Stream after Cryogenic Separation in Packed Bed 1

Feed		Liquid					
Pressure (bar)	80.00	80.00					
Temp (°C)	25.00	0.00	-2.00	-5.00	-10.00	-15.00	-20.00
CH ₄	440.0	53.94	72.43	107.59	186.89	294.13	438.02
C ₂ H ₆	50.000	10.63	13.59	18.71	28.47	38.95	49.87
C ₃ H ₈	27.000	8.21	10.07	13.05	18.07	22.71	26.95
i-C ₄ H ₁₀	10.000	3.82	4.54	5.64	7.33	8.77	9.99
n-C ₄ H ₁₀	10.000	4.16	4.89	5.97	7.58	8.89	9.99
i-C ₅ H ₁₂	10.000	5.02	5.73	6.71	8.10	9.15	9.99
n- C ₅ H ₁₂	1.000	0.53	0.60	0.70	0.83	0.92	1.00
C ₆ H ₁₄	1.000	0.64	0.70	0.78	0.88	0.95	1.00
C ₇ H ₁₆	0.500	0.37	0.39	0.42	0.46	0.48	0.50
C ₈ H ₁₈	0.500	0.40	0.42	0.44	0.47	0.49	0.50
H ₂ O	40.00	0.00	0.00	0.00	0.00	0.00	0.00
CO ₂	400.00	78.37	101.84	143.09	223.01	309.25	398.92
N ₂	10.00	0.86	1.19	1.85	3.46	5.96	9.94
Total flow	1000.0	206.71	256.21	344.80	525.47	740.62	996.66

TABLE 10. Composition of Solid Stream after Cryogenic Separation in Packed Bed 1

Feed		solid					
Pressure (bar)	80.00	80.00					
Temp (°C)	25.00	0.00	-2.00	-5.00	-10.00	-15.00	-20.00
CH ₄	440.000	0.00	0.00	0.00	0.00	0.00	0.00
C ₂ H ₆	50.000	0.00	0.00	0.00	0.00	0.00	0.00
C ₃ H ₈	27.000	0.00	0.00	0.00	0.00	0.00	0.00
i-C ₄ H ₁₀	10.000	0.00	0.00	0.00	0.00	0.00	0.00
n-C ₄ H ₁₀	10.000	0.00	0.00	0.00	0.00	0.00	0.00
i-C ₅ H ₁₂	10.000	0.00	0.00	0.00	0.00	0.00	0.00
n- C ₅ H ₁₂	1.000	0.00	0.00	0.00	0.00	0.00	0.00
C ₆ H ₁₄	1.000	0.00	0.00	0.00	0.00	0.00	0.00
C ₇ H ₁₆	0.500	0.00	0.00	0.00	0.00	0.00	0.00
C ₈ H ₁₈	0.500	0.00	0.00	0.00	0.00	0.00	0.00
H ₂ O	40.00	39.77	39.81	39.86	39.93	39.97	40.00
CO ₂	400.00	0.00	0.00	0.00	0.00	0.00	0.00
N ₂	10.00	0.00	0.00	0.00	0.00	0.00	0.00

At the operating pressure of 80 bar, $-5\text{ }^{\circ}\text{C}$ is selected as the optimum initial bed temperature for packed bed 1. It is elucidated from tables, at 80 bar and $-5\text{ }^{\circ}\text{C}$, most of the water presents in the natural gas has been removed in the form ice while there is comparatively little methane condense to liquid state and furthermore the cooling duty is not significantly high. The same elimination technique has been applied on other operating pressures, ranging from 70 bar to 40 bar, in order to select an optimum initial bed temperature at each operating pressure as shown in the Appendix.

The selected optimum pressure and temperature then became the feed conditions for packed bed 2 and packed bed 3. The simulation process and the elimination technique repeated again in a cycle, until there is no more proposed packed beds to be simulated.

After simulation process for all the packed beds, process analysis is done so as to compare the pressure and temperature combinations based on the performance objectives. The performance objectives include water separation, CO_2 separation, and heavy hydrocarbon separation. The production cost, which is better known as revenue in this case, is calculated based on the price of each component, which is shown in the methodology section earlier. Since the process analysis is done for all packed beds and in each bed there are a numbers of selected temperatures for every operating pressure, there are many schemes for this cryogenic separation system, which can be identified using the node-edge diagram as shown in the methodology section.

4.2 Sensitivity analysis for optimal temperature and pressure based on Node-edge diagrams

The effect of temperature and pressure on separation in multiple beds is discussed in previous section. In this section the optimal condition for maximum separation, minimum methane losses and minimum energy consumption is explored. For this objective a series of node-edge diagrams were used. The temperature and pressure for both dehydration and CO_2 removal beds varied from feed condition in order to investigate the optimal conditions.

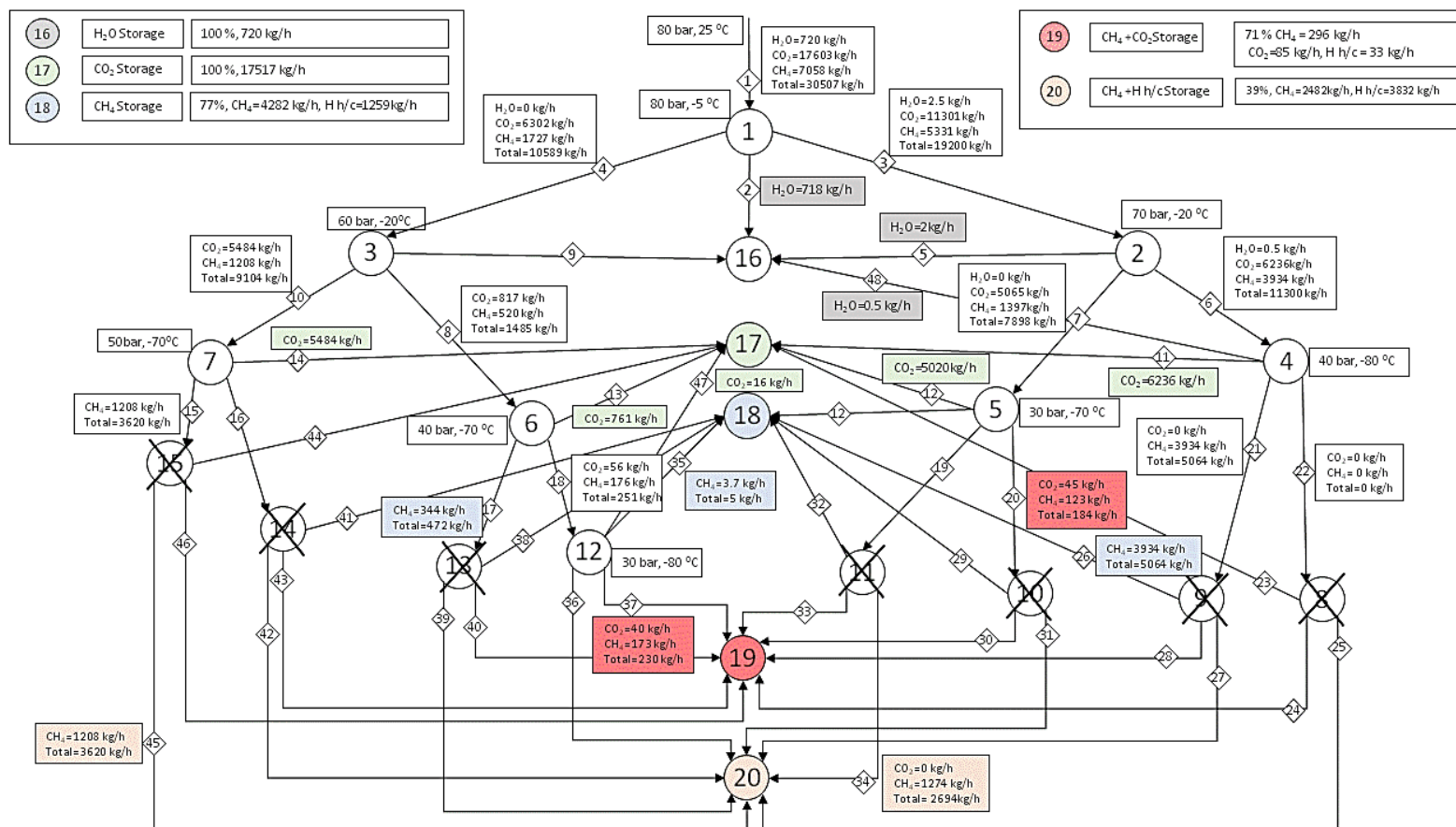
4.2.1 Analysis PB-1

It is illustrated from FIGURE 8 that the raw natural gas is fed into packed bed 1 at a feed condition of 80 bar and 25 °C. Initially, the separation efficiency of packed bed 1 is investigated at 80 bar and -5 °C, where the operating temperature is below the freezing point of water.

When associated water has been separated from the natural gas in solid form, the vapor phase product fed into packed bed 2 while the water-free condensed liquid phase product fed into packed bed 3. Due to small amount of water vapor still remains in the vapor phase stream in packed bed 1, the natural gas undergoes dehydration process again in packed bed 2. Meanwhile, the water-free liquid product that has been directed into packed bed 3 for CO₂ removal. As the operating pressure of packed bed 3 reduces, some of the liquid feed vaporizes and separates some of the heavy hydrocarbons from the methane. Both the vapor and liquid product of packed bed 3 are then fed into packed 6 and 7 respectively to undergo CO₂ removal, by operating the bed below the desublimation temperature of CO₂.

The forth layer of packed beds in node edge diagram, namely packed bed 8, 9, 10, 11, 12, 13, 14, 15 are used if the CO₂ content of the vapor phase or liquid product do not meet the pipeline specifications. For instance, in the diagram above, it is shown that only packed bed 12 is in use to remove the huge amount of CO₂ that is contained in the feed stream, while other beds are not in use and act as pipelines that transport the product to respective product storage tank

After the separation of water and CO₂ the purity of the products are calculated. As shown in FIGURE 8, grade 1 product which is stored in storage tank 18 is 77% pure while grade 2 product that is stored in storage tank 19 has a purity value of 71%. Grade 3 which consists of mainly heavy hydrocarbons contains 39% of methane.



Analysis	Manipulated Pressure & Temperature combination of Packed Bed	Total product Cost								ϕ_1 , Product Cost (\$/cycle)	Energy (MW/hr)	ϕ_2 , Energy Cost (\$/cycle)	ϕ_4 , equipment cost (\$/cycle)	objective function (\$/cycle)
		kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)					
0	(Packed Bed 1) 80 bar, -5 °C	17534	701.36	5877	1160.33	1397.50	246.46	4978	593.48	233421199	3.00	28550016	497979	204373204

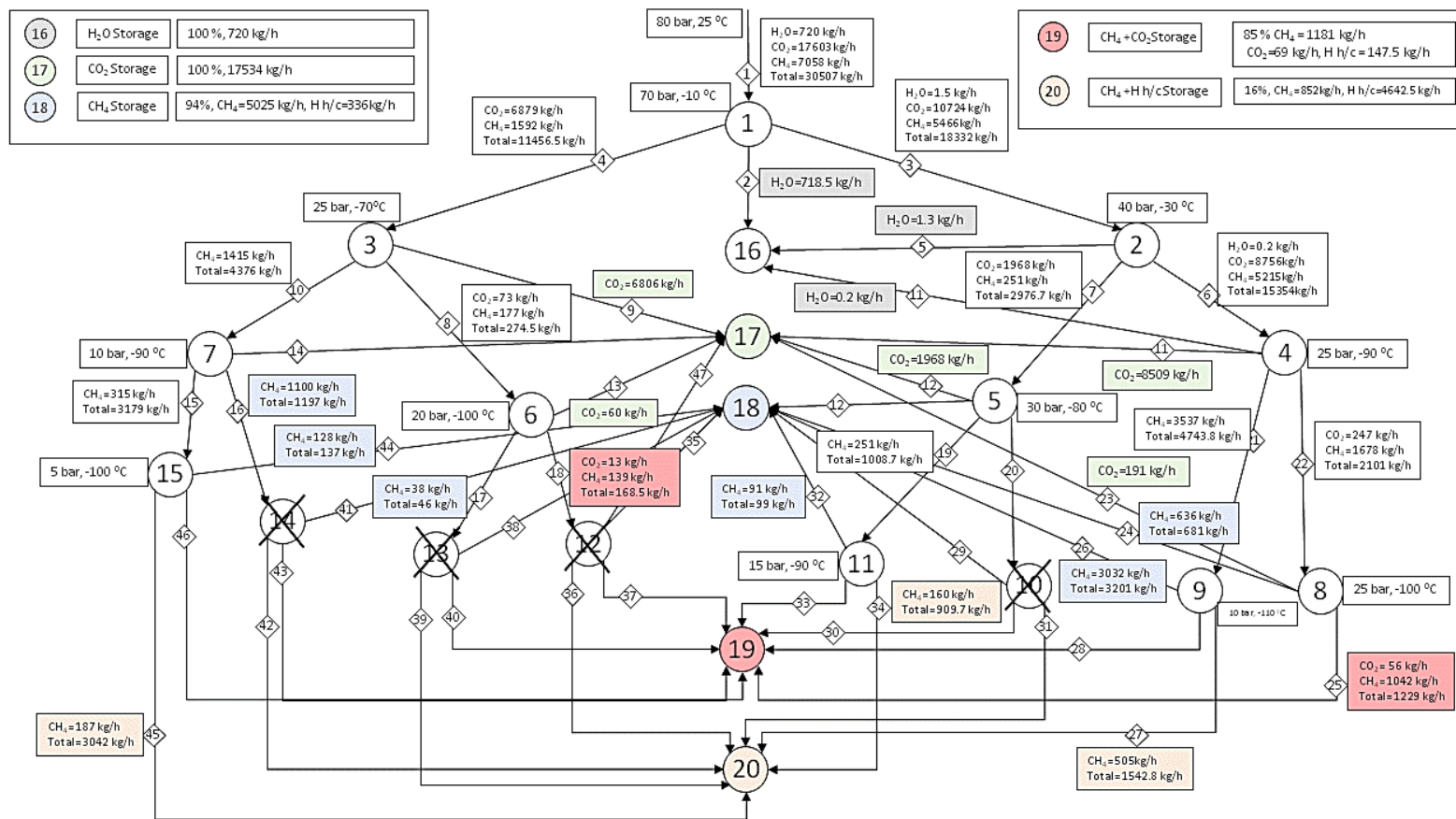
FIGURE 8. Node-edge Diagram for PB-1

4.2.2 Analysis PB-2

In order to investigate how the temperature and pressure affects the separation, several temperature and pressure combinations were used namely; 80 bar and -10°C , 80 bar and -15°C , 70 bar and -10°C , 70 bar and -15°C . The sensitivity analysis shows that the maximum separation is achieved with minimum energy requirement when packed bed 1 is operated at 70 bar and -10°C .

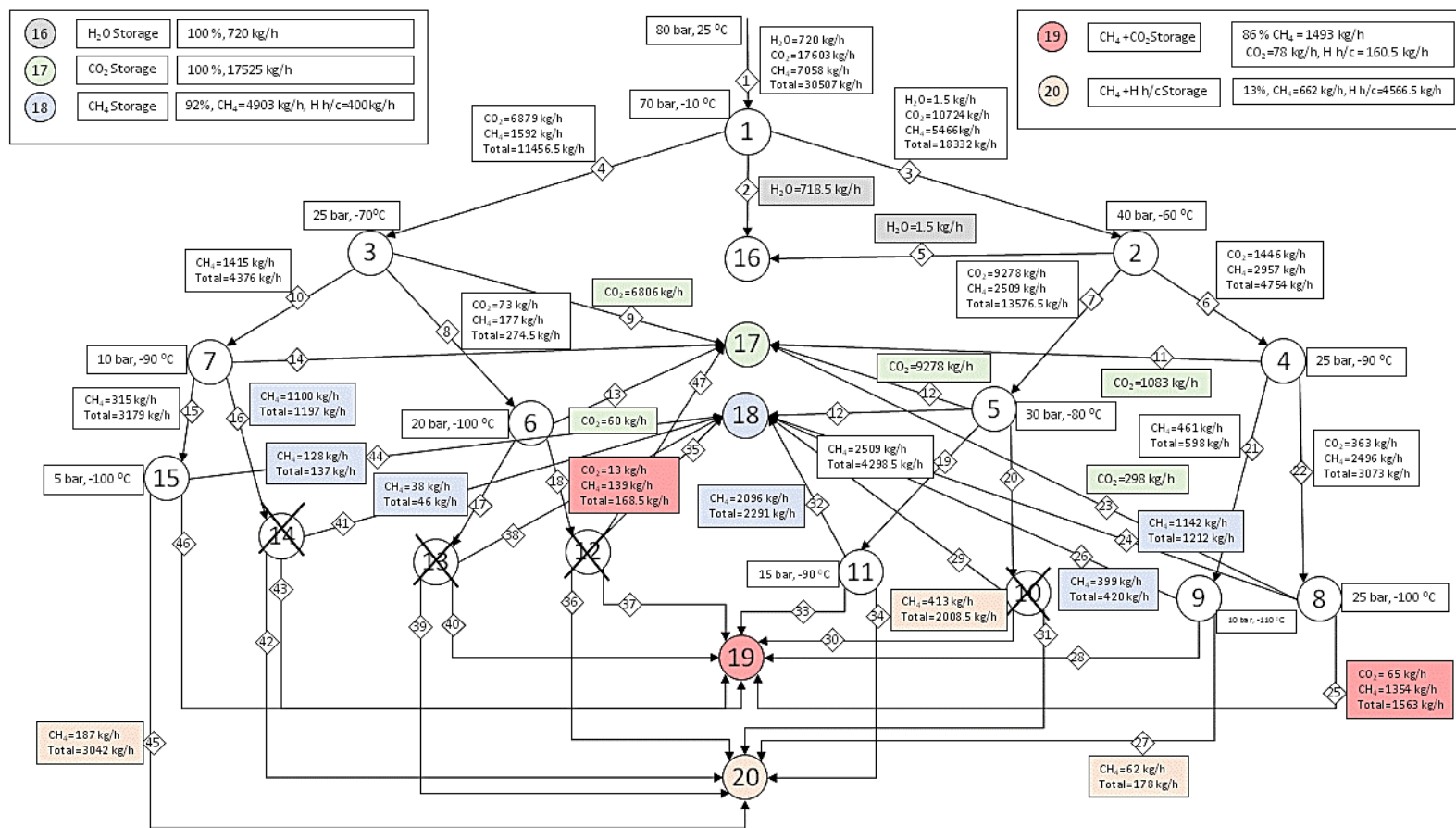
Next, by setting the operation conditions for packed bed 1 to be at 70 bar and -10°C , the sensitivity analysis is conducted on packed bed 2 which further dehydrates the feed gas in order to meet the pipe line specifications. From FIGURE 9 and FIGURE 10 on the next pages, it is observed that by keeping the same pressure and temperature combinations for all packed beds except packed bed 2, the effect on separation is significant. It is also observed that the change in operating temperature affects the purity as well as the amount of the products being produced. For instance, increasing the operating temperature from -60°C to -30°C not only increases purity of high purity methane by 1% (from 94% to 93%), but also increases the amount of high purity methane that is being produced (from 5303 kg/h to 5363 kg/h). This increment in terms of purity and flow rate is desirable as the main objective of this separation process is to have maximum separation with minimum methane loss.

Therefore, it is concluded that maximum separation of water is achieved with minimum energy requirement when packed bed 2 is operated at 40 bar and -30°C . The selected optimal operating temperature also reported for atmospheric feed pressure using experimental measurement for binary mixture of methane and CO_2 by Abulhassan (2014). As shown in the PT diagram, pressure has no effect on the freezing point of the components in the natural gas. However, in this packed bed, substantial reduction of pressure is unavoidable so as to prevent the formation of hydrates in the pipeline.



Analysis	Manipulated Pressure & Temperature combination of Packed Bed	Total product Cost								Φ_1 , Product Cost (\$/cycle)	Energy (MW/hr)	Φ_2 , Energy Cost (\$/cycle)	Φ_4 , equipment cost (\$/cycle)	objective function (\$/cycle)
		kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)					
1	(Packed bed 2) 40 bar, -30 °C	17534	701.36	5361	1160.33	1397.50	246.46	5494.50	593.48	233421199	3.18	30241728	1091452	202088019

FIGURE 9. Node-edge Diagram for PB-2



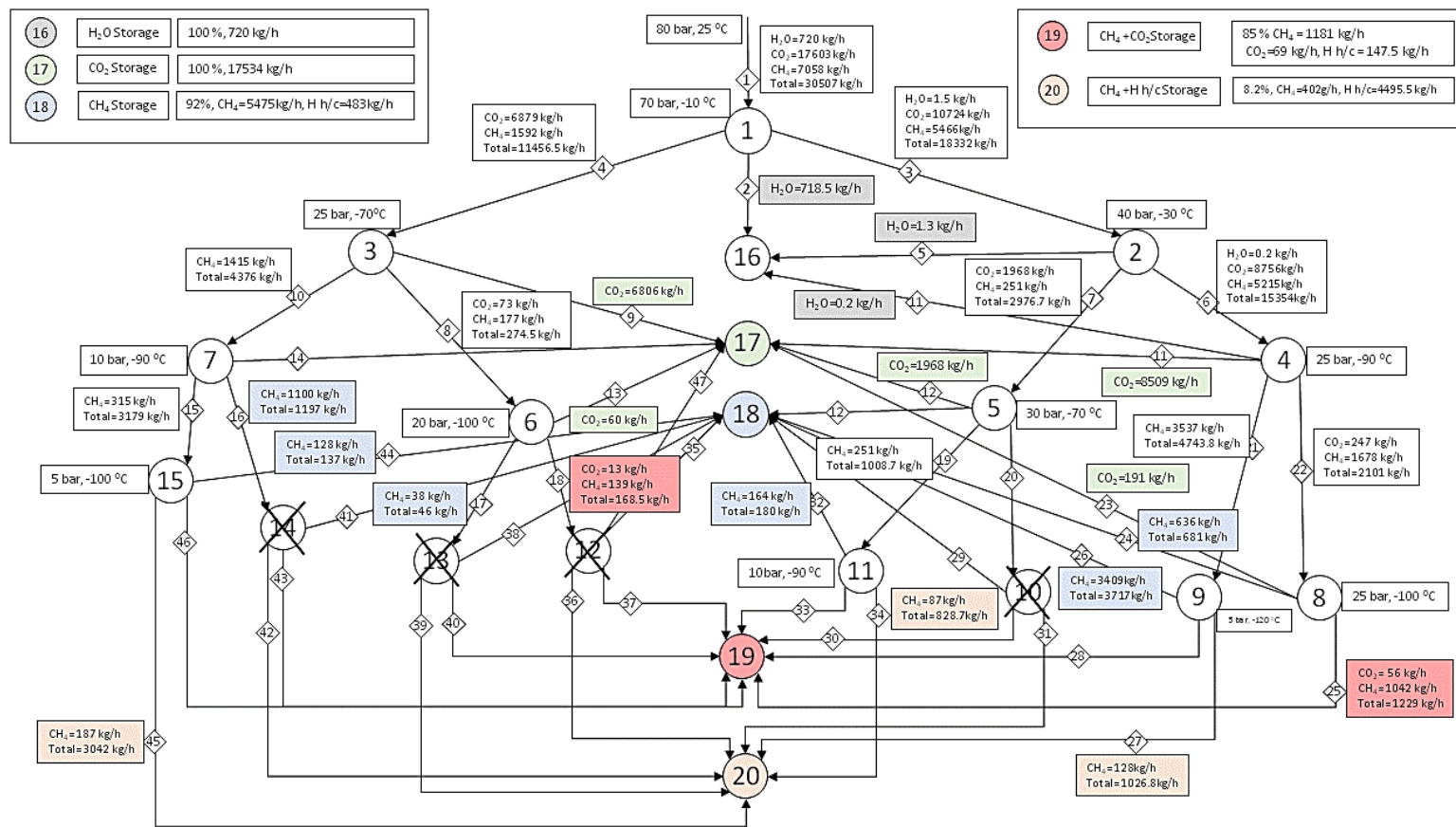
Analysis	Manipulated Pressure & Temperature combination of Packed Bed	Total product Cost								Φ_1 , Product Cost (\$/cycle)	Energy (MW/hr)	Φ_2 , Energy Cost (\$/cycle)	Φ_4 , equipment cost (\$/cycle)	objective function (\$/cycle)
		kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)					
2	(Packed Bed 2) 40 bar, -50 °C	17515	700.60	5020	1074.32	1929.00	355.22	5323.00	570.38	233325523	3.09	29386368	955327	202983828

FIGURE 10. Node-edge Diagram for PB-3

4.2.3 Analysis PB-4 & 5

After the complete removal of water from the natural gas, the subsequent packed beds are used to remove CO₂ from the natural gas. In order to achieve the separation, the bed temperature has to be below the desublimation temperature of CO₂. As shown in the PT diagram earlier, the desublimation temperature of CO₂ is -78.5°C at atmospheric temperature but it greatly affected by changing the pressure.

Similar to the sensitivity analysis that has been done on level 1 and level 2 of the node-edge diagram, by having packed bed 1 and packed bed 2 operated at optimal conditions, 70 bar and -10°C and 40 bar and -10°C respectively, different pressure and temperature combinations of the third level packed beds, namely packed bed 4, 5, 6 and 7 are investigated. As the purity of final methane gas is main objective of this study, packed bed 4 and packed bed 5 where the methane contents are higher are investigated primarily. The simulation results showed that the optimal temperature for packed bed 4 was -90°C for maximum methane recovery (FIGURE 11).



Analysis	Manipulated Pressure & Temperature combination of Packed Bed	Total product Cost								Φ_1 , Product Cost (\$/cycle)	Energy (MW/hr)	Φ_2 , Energy Cost (\$/cycle)	Φ_4 , equipment cost (\$/cycle)	objective function (\$/cycle)
		kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)	kg/h	Cost(\$/h)					
7	(Packed bed 5) 30 bar, -70 OC (Packed Bed 11) 10 bar, -90 OC (Packed bed 9) 50 bar, -110 OC	17534	701.36	5877	1242.11	1397.50	246.46	4978.5	572.50	238674183	3.09	29357856	1385588	207930738

FIGURE 11. Node-edge Diagram for PB-4

The packed bed 5 was investigated for several temperatures in order to find the optimal conditions. It is elucidated from FIGURE 10 and FIGURE 11 that the first bed is operated at -70°C while the other operated at -80°C . It was observed that the small change in temperature does not have the significant effect on CO_2 removal. However, in terms of energy requirement, packed bed 5 which is operated at 30 bar and -80°C requires more energy to cool down the bed. Therefore, in this case, the suitable condition for packed bed 5 is 30 bar and -70°C . A comparative study based on the Node-edge diagram is summarized in

TABLE 11. It is elucidated from TABLE 11 that the optimal scheme for dehydration and CO_2 removal is PB 4&5 where the profit objective function is maximum and the purity of the product is also higher.

TABLE 11. Comparative Study of Different Multiple Packed Bed Schemes

Analysis	Total Mass Flow Rate (kg/h)				Φ_1 , Product Cost (\$/cycle)	Energy (MWh)	Φ_2 , Energy Cost (\$/cycle)	Φ_4 , Equipment cost (\$/cycle)	Profits (\$/cycle)
	17	18	19	20					
PB-1	17534	5877	1397.5	4978	1.80E+08	3.00	2.85E+07	4.98E+05	1.51E+08
PB-2	17534	5361	1397.5	5494.5	2.33E+08	3.18	3.02E+07	1.09E+06	2.02E+08
PB-3	17525	5303	1731.50	5228.50	2.33E+08	3.07	2.91E+07	9.76E+05	2.02E+08
PB-4&5	17534	5877	1397.50	4978.5	2.39E+08	3.09	2.93E+07	1.39E+06	2.07E+08

CHAPTER 5

CONCLUSION AND RECOMMENDATION

1. In this study the optimal operating conditions for cryogenic purification using multiple packed beds are investigated. A detailed simulation study of simultaneous water and CO₂ removal from natural gas was done.
2. A general multiple beds scheme and node edge based strategy was developed for optimal operating conditions. The sensitivity analysis for each node in node-edge diagram was explored and it was observed that even a small change in operating temperature can affect the purity and amount of the products produced.
3. The simulation of 40% CO₂ feed concentration at feed condition 80 bar and 25 °C was done. The optimal conditions for 1st dehydration bed was found to be 70 bar and -10 °C and at these conditions the percentage separation of water was found to be 99.79%.
4. Further simulation results were obtained for the removal of CO₂ with multiple beds. It was observed that in order to achieve the pipeline gas specification, the feed gas pressure need to substantially reduce for minimizing methane losses and maximizing profit objective function.
5. The optimal condition were explored for a node edge diagram, where 92 % methane product with 5475 kg/h and 85% methane product with 1181 kg/h were obtained by introducing 7058 kg/h total flow rate of methane.
6. The present study showed promising results, but more research work need to be done in order to assess the system.

REFERENCES

- Abulhassan. A., Maqsood. K., Syahera, N., Shariff, A., Ganguly S.(2014). Minimization of Energy Consumption In Counter Current Switched Cryogenic Paced Beds during Purification of Natural Gas with High Carbon Dioxide Content, *Chemical Engineering & Technology*, 37(10), 1-12.
- Berstad, D., Anantharaman, R., & Neksa, P. (2013). Low-temperature CO₂ capture technologies – Applications and potential. *International Journal of Refrigeration*, 36(5), 1403-1416.
- Burgers, W. F. J., Northrop, P. S., Kheshgi, H. S., & Valencia, J. A. (2011). Worldwide development potential for sour gas. *Energy Procedia*, 4(0), 2178-2184. doi: <http://dx.doi.org/10.1016/j.egypro.2011.02.104>
- Burt, S. Baxter, A., Baxter, L. (n.d.). *Cryogenic CO₂ Capture to Control Climate Change Emissions*. Brigham Young University, Provo, Utah, USA.
- Darman, N. H., & Harun, A. R. B. (2006). Technical Challenges and Solutions on Natural Gas Development in Malaysia. *Proceedings of The Petroleum Policy and Management (PPM) Project- 4th Workshop of the China-Sichuan Basin Case Study*, 30 May - 3 June, 2006, China.
- Engineer, S. O. (2004). Optimisation CO₂ Removal Design For Block B-17 in MTJDA *SPE Asia Pacific Oil and Gas Conference and Exhibition*, 4.
- Hart, A., & Gnanendran, N. (2009). Cryogenic CO₂ capture in natural gas. *Energy Procedia*, 1(1), 697-706. doi: <http://dx.doi.org/10.1016/j.egypro.2009.01.092>
- Hlanvinka, M. W., Hernandez, V. N., & McCartney, D. (2006). Proper Interpretation of Freezing and Hydrate Prediction Results From Process Simulation. *Proceedings of the Eight-Fifth GPA Annual Convention*, Grapevine, Texas.
- Holmes, A., Price, B., Ryan, J., Styring, R.(1982). in proc. of the 65th Annual Convention of Gas Processor Association, Tulsa.
- Hubbard, B. (2010). New and Emerging Technologies (Petroskills Workshop). Austin, Texas: 2010 Gas Processors Association Convention. John.Campbell Co.
- Karen, H. W. W. (2013). *Purification of Natural Gas Using Cryogenic Multiple Bed Based Dehydration And CO₂ Separation*. (Bachelor of Engineering), University Teknologi PETRONAS, Perak, Malaysia.

- Kelley, B. T., Valencia, J. A., Northrop, P. S., & Mart, C. J. (2011). Controlled Freeze Zone™ for developing sour gas reserves. *Energy Procedia*, 4(0), 824-829. doi: <http://dx.doi.org/10.1016/j.egypro.2011.01.125>
- Li, B., Duan, Y., Luebke, D., & Morreale, B. (2013). Advances in CO₂ capture technology: A patent review. *Applied Energy*, 102(0), 1439-1447. doi: <http://dx.doi.org/10.1016/j.apenergy.2012.09.009>
- Mondal, M. K., Balsora, H. K., & Varshney, P. (2012). Progress and trends in CO₂ capture/separation technologies: A review. *Energy*, 46(1), 431-441. doi: <http://dx.doi.org/10.1016/j.energy.2012.08.006>
- Maqsood, K., AbulHassan, A., Shariff, A., Ganguly, S.(2014).Synthesis of Conventional and Hybrid Cryogenic Distillation Sequence for Purification of Natural Gas, *Journal of Applied Sciences*, 14(21), 2722-2729.
- Maqsood, K., Pal, J., Turunawarasu, D., Pal, A. J. , Ganguly, S. (2014) Performance Enhancement and Energy Reduction Using Hybrid Cryogenic Distillation Networks For Purification of Natural Gas With High CO₂ Content. *Korean J. Chem. Eng*, 31(7), 1120-1135. doi: 10.1007/s11814-014-0038-y
- Northrop, P. S., & Valencia, J. A. (2009). The CFZ™ process: A cryogenic method for handling high- CO₂ and H₂S gas reserves and facilitating geosequestration of CO₂ and acid gases. *Energy Procedia*, 1(1), 171-177. doi: <http://dx.doi.org/10.1016/j.egypro.2009.01.025>
- Olajire, A. A. (2010). CO₂ capture and separation technologies for end-of-pipe applications – A review. *Energy*, 35(6), 2610-2628. doi: <http://dx.doi.org/10.1016/j.energy.2010.02.030>
- Ozturk, M. (2010). *Modelling Vapor-Liquid-Solid Phase Behaviour in Natural Gas Systems*. (Master of Science), Rice University, Houston, Texas.
- Pikaar, M. J. (1959). *A Study of Phase Equilibria in Hydrocarbon-CO₂ Systems*. University of London, London.
- Redza, M. A.(2013) *Multiple Cryogenic Packed Bed Dehydration and Separation of CO₂ and Hydrocarbons from Natural Gas*.(Bachelor of Engineering), University Teknologi PETRONAS, Perak, Malaysia.

- Rufford, T. E., Smart, S., Watson, G. C. Y., Graham, B. F., Boxall, J., Diniz da Costa, J. C., & May, E. F. (2012). The removal of CO₂ and N₂ from natural gas: A review of conventional and emerging process technologies. *Journal of Petroleum Science and Engineering*, 94–95(0), 123-154. doi: <http://dx.doi.org/10.1016/j.petrol.2012.06.016>
- Syahera, M. N. (2012). *Cryogenic Separation of CO₂ From Methane Using Dynamic Packed Bed*. (Bachelor of Engineering), University Teknologi PETRONAS, Perak, Malaysia
- Tuinier, M. J., van Sint Annaland, M., Kramer, G. J., & Kuipers, J. A. M. (2010). Cryogenic capture using dynamically operated packed beds. *Chemical Engineering Science*, 65(1), 114-119. doi: <http://dx.doi.org/10.1016/j.ces.2009.01.055>
- Tuinier, M. J., van Sint Annaland, M., & Kuipers, J. A. M. (2011). A novel process for cryogenic CO₂ capture using dynamically operated packed beds—An experimental and numerical study. *International Journal of Greenhouse Gas Control*, 5(4), 694-701. doi: <http://dx.doi.org/10.1016/j.ijggc.2010.11.011>
- Tuinier, M. J. & van Sint Annaland. (2012). Biogas Purification Using Cryogenic Packed-Bed Technology. *Industrial & Engineering Chemistry Research*, 51(15), 5552-5558.
- Valencia, J. A., & Mentzer, B. K. (2008). *Processing of high CO₂ and H₂S gas with controlled freeze zone™ technology*. Paper presented at the Gas Information Exchange (GASEX) 2008. http://www.kgu.or.kr/download.php?tb=bbs_017&fn=a2b1e8327438313bbb8e869b8914b773.pdf&rn=14b04_paper.pdf
- Xu, G., Li, L., Yang, Y., Tian, L., Liu, T., & Zhang, K. (2012). A novel CO₂ cryogenic liquefaction and separation system. *Energy*, 42(1), 522-529. doi: <http://dx.doi.org/10.1016/j.energy.2012.02.048>

APPENDIX

Wide range of operating conditions was simulated in order to obtain the optimal condition for each node. The simulation results for node 1 (packed bed 1) are given below.

Simulations for Packed Bed 1 (Vapor Stream)

Feed		Vapor												
Pressure (bar)	80.0	80						70						
Temp (°C)	25.0	0	-2	-5	-10	-15	-20	0	-2	-5	-10	-15	-20	-30
CH ₄	440.0	386.06	367.57	332.41	253.11	145.87	1.98	408.52	399.20	381.38	340.82	287.27	221.20	40.44
C ₂ H ₆	50.0	39.37	36.41	31.29	21.53	11.05	0.13	41.82	39.81	36.27	29.33	21.89	14.71	2.00
C ₃ H ₈	27.0	18.79	16.93	13.95	8.93	4.29	0.05	19.47	17.99	15.60	11.51	7.84	4.85	0.58
i-C ₄ H ₁₀	10.0	6.18	5.46	4.36	2.67	1.23	0.01	6.13	5.52	4.59	3.18	2.04	1.21	0.14
n-C ₄ H ₁₀	10.0	5.84	5.11	4.03	2.42	1.11	0.01	5.64	5.02	4.11	2.77	1.75	1.01	0.11
i-C ₅ H ₁₂	10.0	4.98	4.27	3.29	1.90	0.85	0.01	4.43	3.84	3.03	1.94	1.17	0.66	0.07
n- C ₅ H ₁₂	1.0	0.47	0.40	0.30	0.17	0.08	0.00	0.40	0.34	0.27	0.17	0.10	0.06	0.01
C ₆ H ₁₄	1.0	0.36	0.30	0.22	0.12	0.05	0.00	0.26	0.22	0.17	0.10	0.06	0.03	0.00
C ₇ H ₁₆	0.5	0.13	0.11	0.08	0.04	0.02	0.00	0.08	0.07	0.05	0.03	0.02	0.01	0.00
C ₈ H ₁₈	0.5	0.10	0.08	0.06	0.03	0.01	0.00	0.05	0.04	0.03	0.02	0.01	0.00	0.00
H ₂ O	40.0	0.23	0.19	0.14	0.07	0.03	0.00	0.21	0.18	0.14	0.08	0.05	0.02	0.00
CO ₂	400.0	321.63	298.16	256.91	176.99	90.75	1.08	344.28	328.87	300.83	243.79	181.49	121.40	16.42
N ₂	10.0	9.14	8.81	8.15	6.54	4.04	0.06	9.58	9.44	9.17	8.51	7.56	6.23	1.41
Total flow	1000.0	793.29	743.79	655.20	474.53	259.38	3.34	840.87	810.54	755.64	642.26	511.25	371.39	61.19
cool duty (mW)		0.76	0.85	0.99	1.22	1.46	1.70	0.56	0.64	0.77	1.00	1.25	1.50	1.97

Simulations for Packed Bed 1 (Vapor Stream, continue)

Vapor													
60							50						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
423.12	418.66	409.89	388.25	356.90	316.73	211.49	431.32	429.35	425.55	415.86	399.77	375.93	308.87
44.34	43.04	40.65	35.44	29.13	22.61	11.10	46.35	45.58	44.15	40.82	36.05	30.15	18.36
20.78	19.61	17.64	13.94	10.27	7.16	2.92	22.24	21.36	19.85	16.81	13.25	9.76	4.67
6.45	5.91	5.08	3.71	2.53	1.65	0.61	6.95	6.49	5.76	4.47	3.21	2.17	0.91
5.86	5.31	4.47	3.16	2.10	1.34	0.48	6.32	5.81	5.04	3.77	2.61	1.71	0.68
4.35	3.82	3.07	2.03	1.28	0.78	0.27	4.57	4.05	3.33	2.29	1.48	0.92	0.34
0.38	0.33	0.26	0.17	0.11	0.06	0.02	0.40	0.35	0.28	0.19	0.12	0.07	0.03
0.22	0.19	0.14	0.09	0.05	0.03	0.01	0.21	0.18	0.14	0.09	0.05	0.03	0.01
0.06	0.05	0.04	0.02	0.01	0.01	0.00	0.05	0.04	0.03	0.02	0.01	0.01	0.00
0.03	0.02	0.02	0.01	0.01	0.00	0.00	0.02	0.02	0.01	0.01	0.00	0.00	0.00
0.21	0.18	0.15	0.09	0.06	0.03	0.01	0.22	0.19	0.15	0.10	0.06	0.04	0.01
365.55	356.40	338.67	297.16	244.03	187.99	90.61	380.48	375.80	366.66	343.08	305.04	254.11	150.01
9.81	9.76	9.64	9.35	8.91	8.29	6.38	9.92	9.90	9.86	9.75	9.56	9.27	8.36
881.19	863.29	829.71	753.42	655.38	546.69	323.90	909.05	899.11	880.80	837.24	771.24	684.16	492.25
0.36	0.43	0.54	0.76	1.00	1.26	1.76	0.18	0.23	0.32	0.50	0.71	0.97	1.50

Simulations for Packed Bed 1 (Vapor Stream, continue)

Vapor													
40							30						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
435.76	434.88	433.29	429.56	423.51	413.12	373.61	438.21	437.83	437.16	435.71	433.66	430.59	416.36
47.84	47.39	46.59	44.78	42.08	38.04	26.56	48.93	48.69	48.25	47.32	46.01	44.12	36.92
23.69	23.05	21.96	19.72	16.90	13.52	7.04	25.10	24.68	23.94	22.40	20.43	17.97	11.46
7.62	7.22	6.58	5.43	4.22	3.04	1.32	8.45	8.13	7.60	6.60	5.50	4.35	2.19
6.99	6.53	5.82	4.61	3.43	2.37	0.96	7.94	7.54	6.90	5.74	4.57	3.45	1.59
5.11	4.58	3.82	2.74	1.86	1.20	0.44	6.16	5.59	4.77	3.52	2.50	1.70	0.68
0.44	0.39	0.32	0.22	0.15	0.09	0.03	0.54	0.48	0.40	0.28	0.19	0.13	0.05
0.22	0.19	0.14	0.09	0.06	0.03	0.01	0.28	0.23	0.18	0.11	0.07	0.04	0.01
0.05	0.04	0.03	0.02	0.01	0.01	0.00	0.06	0.05	0.03	0.02	0.01	0.01	0.00
0.02	0.02	0.01	0.01	0.00	0.00	0.00	0.02	0.02	0.01	0.01	0.00	0.00	0.00
0.24	0.21	0.17	0.11	0.07	0.05	0.02	0.27	0.24	0.19	0.13	0.09	0.06	0.02
389.72	387.47	383.26	372.90	355.21	324.25	221.37	395.30	394.27	392.39	388.13	381.66	371.14	317.28
9.96	9.96	9.94	9.91	9.85	9.75	9.32	9.99	9.98	9.98	9.97	9.95	9.93	9.81
927.66	921.92	911.93	890.10	857.36	805.46	640.69	941.27	937.74	931.80	919.93	904.64	883.50	796.38
0.01	0.06	0.13	0.26	0.42	0.62	1.16	-0.15	-0.11	-0.04	0.07	0.19	0.32	0.70

Simulations for Packed Bed 1 (Liquid Stream)

Feed		Liquid												
Pressure (bar)	80.0	80						70						
Temp (°C)	25.0	0	-2	-5	-10	-15	-20	0	-2	-5	-10	-15	-20	-30
CH ₄	440.0	53.94	72.43	107.59	186.89	294.13	438.02	31.48	40.80	58.62	99.18	152.73	218.80	399.56
C ₂ H ₆	50.0	10.63	13.59	18.71	28.47	38.95	49.87	8.18	10.19	13.73	20.67	28.11	35.29	48.00
C ₃ H ₈	27.0	8.21	10.07	13.05	18.07	22.71	26.95	7.53	9.01	11.40	15.49	19.16	22.15	26.42
i-C ₄ H ₁₀	10.0	3.82	4.54	5.64	7.33	8.77	9.99	3.87	4.48	5.41	6.82	7.96	8.79	9.86
n-C ₄ H ₁₀	10.0	4.16	4.89	5.97	7.58	8.89	9.99	4.36	4.98	5.89	7.23	8.25	8.99	9.89
i-C ₅ H ₁₂	10.0	5.02	5.73	6.71	8.10	9.15	9.99	5.57	6.16	6.97	8.06	8.83	9.34	9.93
n- C ₅ H ₁₂	1.0	0.53	0.60	0.70	0.83	0.92	1.00	0.60	0.66	0.73	0.83	0.90	0.94	0.99
C ₆ H ₁₄	1.0	0.64	0.70	0.78	0.88	0.95	1.00	0.74	0.78	0.83	0.90	0.94	0.97	1.00
C ₇ H ₁₆	0.5	0.37	0.39	0.42	0.46	0.48	0.50	0.42	0.43	0.45	0.47	0.48	0.49	0.50
C ₈ H ₁₈	0.5	0.40	0.42	0.44	0.47	0.49	0.50	0.45	0.46	0.47	0.48	0.49	0.50	0.50
H ₂ O	40.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
CO ₂	400.0	78.37	101.84	143.09	223.01	309.25	398.92	55.72	71.13	99.17	156.21	218.51	278.60	383.58
N ₂	10.0	0.86	1.19	1.85	3.46	5.96	9.94	0.42	0.56	0.83	1.49	2.44	3.77	8.59
Total flow	1000.0	206.71	256.21	344.80	525.47	740.62	996.66	159.13	189.46	244.36	357.74	488.75	628.61	938.81

Simulations for Packed Bed 1 (Liquid Stream, continue)

Liquid													
60							50						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
16.88	21.34	30.11	51.75	83.10	123.27	228.51	8.68	10.65	14.45	24.14	40.23	64.07	131.13
5.66	6.96	9.35	14.56	20.87	27.39	38.90	3.65	4.42	5.85	9.18	13.95	19.85	31.64
6.22	7.39	9.36	13.06	16.73	19.84	24.08	4.76	5.64	7.15	10.19	13.75	17.24	22.33
3.55	4.09	4.92	6.29	7.47	8.35	9.39	3.05	3.51	4.24	5.53	6.79	7.83	9.09
4.14	4.69	5.53	6.84	7.90	8.66	9.52	3.68	4.19	4.96	6.23	7.39	8.29	9.32
5.65	6.18	6.93	7.97	8.72	9.22	9.73	5.43	5.95	6.67	7.71	8.52	9.08	9.66
0.62	0.67	0.74	0.83	0.89	0.94	0.98	0.60	0.65	0.72	0.81	0.88	0.93	0.97
0.78	0.81	0.86	0.91	0.95	0.97	0.99	0.79	0.82	0.86	0.91	0.95	0.97	0.99
0.44	0.45	0.46	0.48	0.49	0.49	0.50	0.45	0.46	0.47	0.48	0.49	0.49	0.50
0.47	0.48	0.48	0.49	0.49	0.50	0.50	0.48	0.48	0.49	0.49	0.50	0.50	0.50
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
34.45	43.60	61.33	102.84	155.97	212.01	309.39	19.52	24.20	33.34	56.92	94.96	145.89	249.99
0.19	0.24	0.36	0.65	1.09	1.71	3.62	0.08	0.10	0.14	0.25	0.44	0.73	1.64
118.81	136.71	170.29	246.58	344.62	453.31	676.10	90.95	100.89	119.20	162.76	228.76	315.84	507.75

Simulations for Packed Bed 1 (Liquid Stream, continue)

Liquid													
40							30						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
4.24	5.12	6.71	10.44	16.49	26.88	66.39	1.79	2.17	2.84	4.29	6.34	9.41	23.64
2.16	2.61	3.41	5.22	7.92	11.96	23.44	1.07	1.31	1.75	2.68	3.99	5.88	13.08
3.31	3.95	5.04	7.28	10.10	13.48	19.96	1.90	2.32	3.06	4.60	6.57	9.03	15.54
2.38	2.78	3.42	4.57	5.78	6.96	8.68	1.55	1.87	2.40	3.40	4.50	5.65	7.81
3.01	3.47	4.18	5.39	6.57	7.63	9.04	2.06	2.46	3.10	4.26	5.43	6.55	8.41
4.89	5.42	6.18	7.26	8.14	8.80	9.56	3.84	4.41	5.23	6.48	7.50	8.30	9.32
0.56	0.61	0.68	0.78	0.85	0.91	0.97	0.46	0.52	0.60	0.72	0.81	0.87	0.95
0.78	0.81	0.86	0.91	0.94	0.97	0.99	0.72	0.77	0.82	0.89	0.93	0.96	0.99
0.45	0.46	0.47	0.48	0.49	0.49	0.50	0.44	0.45	0.47	0.48	0.49	0.49	0.50
0.48	0.48	0.49	0.49	0.50	0.50	0.50	0.48	0.48	0.49	0.49	0.50	0.50	0.50
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
10.28	12.53	16.74	27.10	44.79	75.75	178.63	4.70	5.73	7.61	11.87	18.34	28.86	82.72
0.04	0.04	0.06	0.09	0.15	0.25	0.68	0.01	0.02	0.02	0.03	0.05	0.07	0.19
72.34	78.08	88.07	109.90	142.64	194.54	359.31	58.73	62.26	68.20	80.07	95.36	116.50	203.62

Simulations for Packed Bed 1 (Solid Stream)

Feed		Solid													
Pressure (bar)	80.0	80						70							
Temp (°C)	25.0	0	-2	-5	-10	-15	-20	0	-2	-5	-10	-15	-20	-30	
CH ₄	440.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
C ₂ H ₆	50.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
C ₃ H ₈	27.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
i-C ₄ H ₁₀	10.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
n-C ₄ H ₁₀	10.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
i-C ₅ H ₁₂	10.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
n- C ₅ H ₁₂	1.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
C ₆ H ₁₄	1.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
C ₇ H ₁₆	0.5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
C ₈ H ₁₈	0.5	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
H ₂ O	40.0	39.77	39.81	39.86	39.93	39.97	40.00	39.79	39.82	39.86	39.92	39.95	39.98	40.00	
CO ₂	400.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	
N ₂	10.0	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	

Simulations for Packed Bed 1 (Solid Stream, continue)

Solid													
60							50						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
39.79	39.82	39.85	39.91	39.94	39.97	39.99	39.78	39.81	39.85	39.90	39.94	39.96	39.99
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Simulations for Packed Bed 1 (Solid Stream, continue)

Solid													
40							30						
0	-2	-5	-10	-15	-20	-30	0	-2	-5	-10	-15	-20	-30
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
39.76	39.79	39.83	39.89	39.93	39.95	39.98	39.73	39.76	39.81	39.87	39.91	39.94	39.98
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

The simulation results for node 3 (packed bed 3) are given below.

Simulations for Packed Bed 3 (Vapor stream)

	Feed	Gas									
Pressure (bar)	80.0	70			60			50			
Temp (°C)	0.0	-20	-25	-30	-20	-25	-30	-20	-25	-30	-40
CH ₄	53.94	0.00	0.00	0.00	6.72	0.00	0.00	25.27	18.19	10.27	0.00
C ₂ H ₆	10.63	0.00	0.00	0.00	0.44	0.00	0.00	1.93	1.17	0.55	0.00
C ₃ H ₈	8.21	0.00	0.00	0.00	0.15	0.00	0.00	0.61	0.34	0.15	0.00
i-C ₄ H ₁₀	3.82	0.00	0.00	0.00	0.04	0.00	0.00	0.14	0.08	0.03	0.00
n-C ₄ H ₁₀	4.16	0.00	0.00	0.00	0.03	0.00	0.00	0.12	0.06	0.03	0.00
i-C ₅ H ₁₂	5.02	0.00	0.00	0.00	0.02	0.00	0.00	0.07	0.04	0.02	0.00
n- C ₅ H ₁₂	0.53	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00
C ₆ H ₁₄	0.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₇ H ₁₆	0.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₈ H ₁₈	0.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
H ₂ O	39.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
CO ₂	78.37	0.00	0.00	0.00	3.69	0.00	0.00	16.33	9.81	4.61	0.00
N ₂	0.86	0.00	0.00	0.00	0.19	0.00	0.00	0.57	0.46	0.30	0.00
Total flow	206.71	-20.00	-25.00	-30.00	-8.72	-25.00	-30.00	25.05	5.16	-14.04	0.00
Cooling Duty(MW)		0.12	0.15	0.17	0.11	0.14	0.17	0.06	0.11	0.15	0.22

Simulations for Packed Bed 3 (Vapor stream, continue)

Gas										
40					30					
-20	-25	-30	-40	-50	-20	-25	-30	-40	-50	-60
38.65	33.84	28.36	15.30	0.00	47.69	45.16	41.80	33.26	22.53	8.71
3.68	2.70	1.88	0.69	0.00	6.02	4.91	3.79	2.02	0.90	0.23
1.18	0.78	0.50	0.16	0.00	2.16	1.54	1.05	0.45	0.17	0.04
0.26	0.17	0.10	0.03	0.00	0.47	0.31	0.20	0.08	0.03	0.01
0.21	0.13	0.08	0.02	0.00	0.37	0.24	0.15	0.06	0.02	0.00
0.11	0.07	0.04	0.01	0.00	0.19	0.12	0.07	0.03	0.01	0.00
0.01	0.01	0.00	0.00	0.00	0.01	0.01	0.01	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
31.70	23.03	15.84	5.70	0.00	52.13	42.74	32.76	16.84	7.25	1.78
0.74	0.70	0.64	0.44	0.00	0.83	0.81	0.78	0.71	0.59	0.33
56.56	36.42	17.44	22.35	0.00	89.88	70.84	50.61	53.45	31.50	11.09
0.003	0.06	0.10	0.19	0.27	-0.08	-0.03	0.03	0.14	0.23	0.31

Simulations for Packed Bed 3 (Liquid stream)

	Feed	Liquid									
Pressure (bar)	80.0	70			60			50			
Temp (°C)	0.0	-20	-25	-30	-20	-25	-30	-20	-25	-30	-40
CH ₄	53.94	53.94	53.94	53.94	47.21	53.94	53.94	28.66	35.74	43.67	53.94
C ₂ H ₆	10.63	10.63	10.63	10.63	10.19	10.63	10.63	8.70	9.46	10.08	10.63
C ₃ H ₈	8.21	8.21	8.21	8.21	8.06	8.21	8.21	7.59	7.86	8.05	8.21
i-C ₄ H ₁₀	3.82	3.82	3.82	3.82	3.78	3.82	3.82	3.67	3.74	3.78	3.82
n-C ₄ H ₁₀	4.16	4.16	4.16	4.16	4.13	4.16	4.16	4.04	4.10	4.13	4.16
i-C ₅ H ₁₂	5.02	5.02	5.02	5.02	5.00	5.02	5.02	4.95	4.98	5.00	5.02
n- C ₅ H ₁₂	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53	0.53
C ₆ H ₁₄	0.64	0.64	0.64	0.64	0.64	0.64	0.64	0.64	0.64	0.64	0.64
C ₇ H ₁₆	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37
C ₈ H ₁₈	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40
H ₂ O	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77
CO ₂	78.37	78.37	78.37	78.37	74.68	78.37	78.37	62.04	68.56	73.76	78.37
N ₂	0.86	0.86	0.86	0.86	0.67	0.86	0.86	0.29	0.40	0.56	0.86
Total flow	206.71	206.71	206.71	206.71	195.43	206.71	206.71	161.66	176.56	190.76	206.71

Simulations for Packed Bed 3 (Liquid stream, continue)

Liquid										
40					30					
-20	-25	-30	-40	-50	-20	-25	-30	-40	-50	-60
-20.00	-25.00	-30.00	-40.00	-50.00	-20.00	-25.00	-30.00	-40.00	-50.00	-60.00
15.28	20.10	25.58	38.64	53.94	6.25	8.78	12.14	20.68	31.41	45.23
6.95	7.93	8.75	9.94	10.63	4.60	5.72	6.83	8.60	9.72	10.40
7.02	7.42	7.71	8.05	8.21	6.04	6.67	7.16	7.76	8.04	8.17
3.55	3.65	3.72	3.79	3.82	3.34	3.50	3.62	3.74	3.79	3.81
3.95	4.03	4.08	4.14	4.16	3.79	3.92	4.01	4.10	4.14	4.16
4.90	4.95	4.98	5.01	5.02	4.83	4.90	4.95	4.99	5.01	5.02
0.52	0.53	0.53	0.53	0.53	0.52	0.52	0.53	0.53	0.53	0.53
0.63	0.64	0.64	0.64	0.64	0.63	0.63	0.64	0.64	0.64	0.64
0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37	0.37
0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40	0.40
39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77	39.77
46.67	55.34	62.53	72.67	78.37	26.24	35.63	45.61	61.53	71.12	76.59
0.12	0.17	0.23	0.42	0.86	0.04	0.05	0.08	0.15	0.27	0.53
130.15	145.29	159.28	184.36	206.71	96.83	110.87	126.10	153.27	175.21	195.62

Simulations for Packed Bed 3 (Solid stream)

	Feed	Solid									
Pressure (bar)	80.0	70			60			50			
Temp (°C)	0.0	-20	-25	-30	-20	-25	-30	-20	-25	-30	-40
CH ₄	53.94	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₂ H ₆	10.63	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₃ H ₈	8.21	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
i-C ₄ H ₁₀	3.82	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
n-C ₄ H ₁₀	4.16	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
i-C ₅ H ₁₂	5.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
n- C ₅ H ₁₂	0.53	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₆ H ₁₄	0.64	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₇ H ₁₆	0.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
C ₈ H ₁₈	0.40	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
H ₂ O	39.77	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
CO ₂	78.37	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
N ₂	0.86	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Total flow	206.71	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Simulations for Packed Bed 3 (Solid stream, continue)

Solid										
40					30					
-20	-25	-30	-40	-50	-20	-25	-30	-40	-50	-60
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00

Process Analysis for Packed Bed 1

The separation efficiencies at each operating condition are investigated at node 1 and the results are shown in the table below. Based on the separation efficiencies and energy requirements for the process, suitable optimal condition for the node was selected and the selected operating condition will be the feed condition for the next node.

Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	80	-5	5331.38	1727.50		0.985	718.10	0	1369.01
		C ₂ H ₆	50.0	1503.50			940.61	562.89					
		C ₃ H ₈	27.0	1190.62			614.86	575.76					
		i-C ₄ H ₁₀	10.0	581.24			253.38	327.86					
		n-C ₄ H ₁₀	10.0	581.24			234.38	346.86					
		i-C ₅ H ₁₂	10.0	721.51			236.99	484.52					
		n- C ₅ H ₁₂	1.0	72.15			21.92	50.23					
		C ₆ H ₁₄	1.0	86.18			19.35	66.83					
		C ₇ H ₁₆	0.5	50.10			8.07	42.03					
		C ₈ H ₁₈	0.5	57.12			6.44	50.68					
		H ₂ O	40.0	720.60			2.50	0.00	718.10				
		CO ₂	400.0	17603.88			11301.87	6302.01					
		N ₂	10.0	280.13			228.32	51.81					
			1000.0			19200.07	10588.97	718.10					
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	80	-10	4058.43	3000.45		1.221	719.29	0	1685.64

		C ₂ H ₆	50.0	1503.50			646.87	856.62					
		C ₃ H ₈	27.0	1190.62			393.51	797.11					
		i-C ₄ H ₁₀	10.0	581.24			154.81	426.43					
		n-C ₄ H ₁₀	10.0	581.24			140.68	440.56					
		i-C ₅ H ₁₂	10.0	721.51			137.24	584.27					
		n- C ₅ H ₁₂	1.0	72.15			12.54	59.61			% separation		% separation
		C ₆ H ₁₄	1.0	86.18			10.75	75.43			99.82		78.42
		C ₇ H ₁₆	0.5	50.10			4.40	45.70					
		C ₈ H ₁₈	0.5	57.12			3.48	53.64					
		H ₂ O	40.0	720.60			1.31		719.29				
		CO ₂	400.0	17603.88			7783.83	9820.05					
		N ₂	10.0	280.13			183.05	97.08					
			1000.0				13530.89	16256.95	719.29				
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	80	-15	2337.28	4721.60		1.461	720.10	0	1939.18
		C ₂ H ₆	50.0	1503.50			331.73	1171.77					
		C ₃ H ₈	27.0	1190.62			188.77	1001.85					
		i-C ₄ H ₁₀	10.0	581.24			71.55	509.69					
		n-C ₄ H ₁₀	10.0	581.24			64.15	517.09					
		i-C ₅ H ₁₂	10.0	721.51			61.07	660.44					
		n- C ₅ H ₁₂	1.0	72.15			5.54	66.61					
		C ₆ H ₁₄	1.0	86.18			4.66	81.52					
										% separation		% separation	
										99.93		90.21	

		C ₇ H ₁₆	0.5	50.10			1.89	48.21					
		C ₈ H ₁₈	0.5	57.12			1.49	55.62					
		H ₂ O	40.0	720.60			0.50		720.10				
		CO ₂	400.0	17603.88			3988.06	13615.82					
		N ₂	10.0	280.13			113.15	166.98					
			1000.0				7169.85	22617.19	720.10				
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	70	-10	5466.74	1592.14		1.005	719.08	0	1638.13
		C ₂ H ₆	50.0	1503.50			881.63	621.86					
		C ₃ H ₈	27.0	1190.62			507.51	683.11					
		i-C ₄ H ₁₀	10.0	581.24			184.64	396.60					
		n-C ₄ H ₁₀	10.0	581.24			161.14	420.10					
		i-C ₅ H ₁₂	10.0	721.51			139.88	581.63					
		n- C ₅ H ₁₂	1.0	72.15			12.21	59.94					
		C ₆ H ₁₄	1.0	86.18			8.72	77.46					
		C ₇ H ₁₆	0.5	50.10			2.93	47.17					
		C ₈ H ₁₈	0.5	57.12			1.88	55.23					
		H ₂ O	40.0	720.60			1.53	0.00	719.08				
		CO ₂	400.0	17603.88			10725.24	6878.64					
N ₂	10.0	280.13	238.38	41.75									
			1000.0			18332.44	11455.63	719.08					
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation

80	25	CH ₄	440.0	7058.88	70	-15	4607.21	2451.67	1.250	719.77	0	1829.67	
		C ₂ H ₆	50.0	1503.50			657.97	845.53					
		C ₃ H ₈	27.0	1190.62			345.61	845.01					
		i-C ₄ H ₁₀	10.0	581.24			118.77	462.47					
		n-C ₄ H ₁₀	10.0	581.24			101.45	479.79					
		i-C ₅ H ₁₂	10.0	721.51			84.61	636.90					
		n- C ₅ H ₁₂	1.0	72.15			7.29	64.86					
		C ₆ H ₁₄	1.0	86.18			5.05	81.13					
		C ₇ H ₁₆	0.5	50.10			1.66	48.44					
		C ₈ H ₁₈	0.5	57.12			1.05	56.06					
		H ₂ O	40.0	720.60			0.84	719.77					
		CO ₂	400.0	17603.88			7983.79	9620.09					
		N ₂	10.0	280.13			211.65	68.48					
		1000.0			14126.93	15660.44	719.77						
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	70	-20	3546.98	3511.90	1.496	720.20	0	1964.74	
		C ₂ H ₆	50.0	1503.50			442.15	1061.34					
		C ₃ H ₈	27.0	1190.62			213.52	977.10					
		i-C ₄ H ₁₀	10.0	581.24			70.06	511.18					
		n-C ₄ H ₁₀	10.0	581.24			58.84	522.40					
		i-C ₅ H ₁₂	10.0	721.51			47.64	673.87					
		n- C ₅ H ₁₂	1.0	72.15			4.06	68.09					
									% separation		% separation		

		C ₆ H ₁₄	1.0	86.18			2.75	83.43			99.94	91.40	
		C ₇ H ₁₆	0.5	50.10			0.89	49.21					
		C ₈ H ₁₈	0.5	57.12			0.56	56.56					
		H ₂ O	40.0	720.60			0.41		720.20				
		CO ₂	400.0	17603.88			5339.17	12264.71					
		N ₂	10.0	280.13			174.57	105.56					
			1000.0				9901.60	19885.34	720.20				
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	60	-20	5080.32	1978.56		1.259	720.05	0	1911.12
		C ₂ H ₆	50.0	1503.50			679.70	823.80					
		C ₃ H ₈	27.0	1190.62			315.47	875.15					
		i-C ₄ H ₁₀	10.0	581.24			96.07	485.17					
		n-C ₄ H ₁₀	10.0	581.24			77.60	503.64					
		i-C ₅ H ₁₂	10.0	721.51			56.37	665.14					
		n- C ₅ H ₁₂	1.0	72.15			4.61	67.55			% separation		
		C ₆ H ₁₄	1.0	86.18			2.66	83.52			99.92	88.91	
		C ₇ H ₁₆	0.5	50.10			0.73	49.37					
		C ₈ H ₁₈	0.5	57.12			0.39	56.73					
		H ₂ O	40.0	720.60			0.56	0.00	720.05				
		CO ₂	400.0	17603.88			8270.21	9333.67					
		N ₂	10.0	280.13			232.24	47.89					
		1000.0				14816.90	14970.19	720.05					

Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition			P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	50	-20	6030.44	1028.43		0.968	719.91	0	1849.36
		C ₂ H ₆	50.0	1503.50			906.42	597.08					
		C ₃ H ₈	27.0	1190.62			430.03	760.59					
		i-C ₄ H ₁₀	10.0	581.24			126.00	455.24					
		n-C ₄ H ₁₀	10.0	581.24			99.22	482.02					
		i-C ₅ H ₁₂	10.0	721.51			66.19	655.32					
		n- C ₅ H ₁₂	1.0	72.15			5.23	66.92					
		C ₆ H ₁₄	1.0	86.18			2.63	83.55					
		C ₇ H ₁₆	0.5	50.10			0.63	49.48					
		C ₈ H ₁₈	0.5	57.12			0.29	56.83					
		H ₂ O	40.0	720.60			0.69	0.00	719.91				
		CO ₂	400.0	17603.88			11180.12	6423.76					
		N ₂	10.0	280.13			259.61	20.52					
			1000.0			19107.49	10679.74	719.91					
Feed					Bed conditions		Composition(kg/h)			Energy	Performance Objective		
P (bar)	T (°C)	Composition	mol/h	kg/h	P (bar)	T (°C)	V	L	S	(MW)	water separated	CO ₂ separation	H-h/c separation
80	25	CH ₄	440.0	7058.88	40	-20	6627.35	431.53		0.624	719.75	0	1738.76
		C ₂ H ₆	50.0	1503.50			1143.70	359.79					
		C ₃ H ₈	27.0	1190.62			595.82	594.79					
		i-C ₄ H ₁₀	10.0	581.24			176.43	404.81					
		n-C ₄ H ₁₀	10.0	581.24			137.70	443.54					

		i-C ₅ H ₁₂	10.0	721.51			86.25	635.26				
		n- C ₅ H ₁₂	1.0	72.15			6.62	65.53			% separation	% separation
		C ₆ H ₁₄	1.0	86.18			2.92	83.26			99.88	80.89
		C ₇ H ₁₆	0.5	50.10			0.61	49.50				
		C ₈ H ₁₈	0.5	57.12			0.24	56.87				
		H ₂ O	40.0	720.60			0.85	0.00	719.75			
		CO ₂	400.0	17603.88			14267.91	3335.97				
		N ₂	10.0	280.13			273.11	7.02				
			1000.0				23319.52	6467.87	719.75			