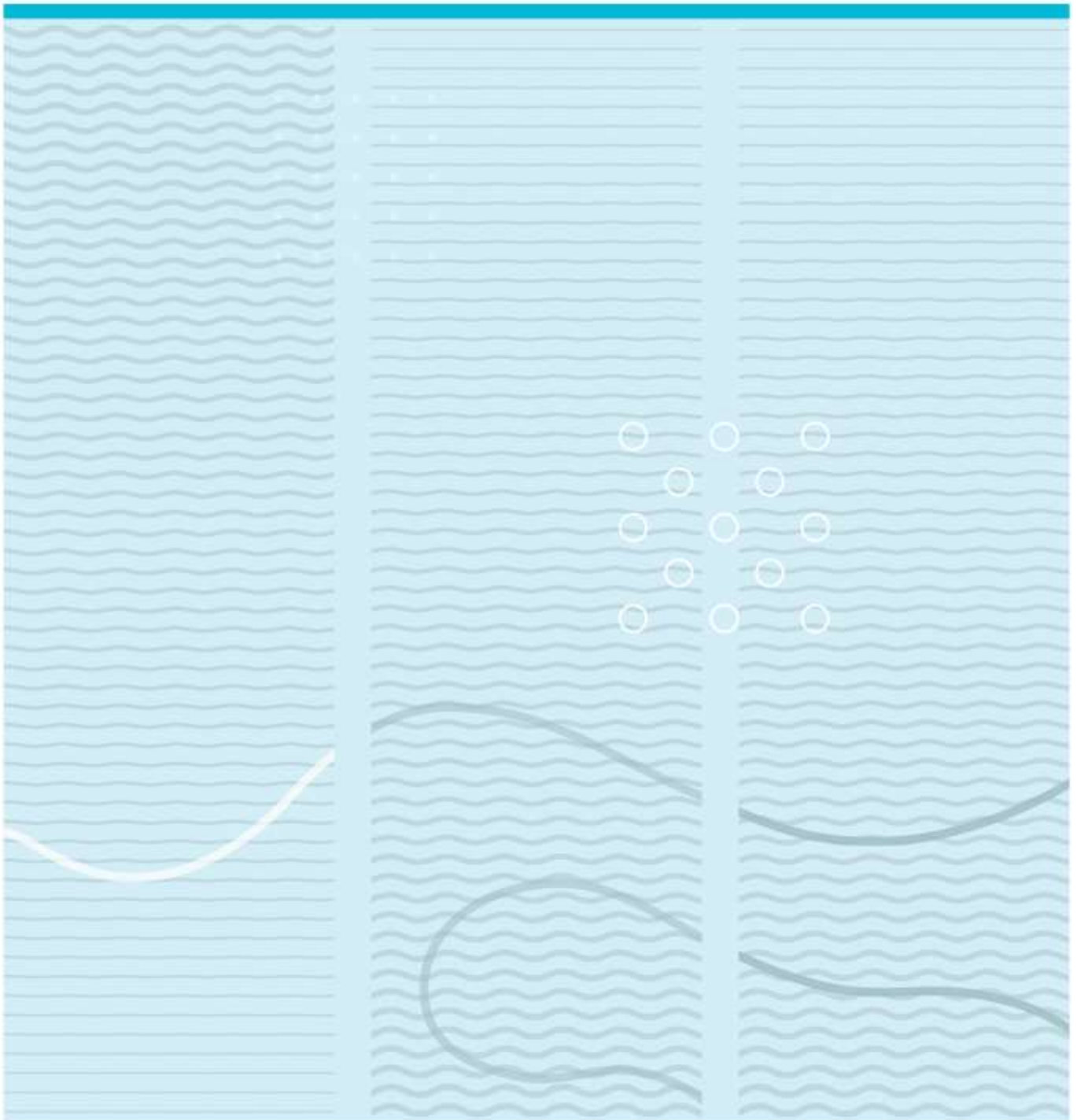Birendra Rai

CO₂ dehydration after CO₂ capture

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Abstract:

The presence of free water is considered as one of the major impurities for the pipeline transportation of gas. Corrosion and hydrate formation in the process plant equipment are common problems associated with the free water. The gas dehydration process has been the most preferred method. Water absorption followed by glycol regeneration are the key stages of a gas dehydration unit.

Although, being in operation for a long period of time there are no rationale standards for water content requirements in the dehydration of gas. Water specifications between 5 and 500 ppm (parts per million) are generally considered. Only a few references have been published on simulation of CO₂ dehydration based on absorption in glycol.

All the simulations have been performed in Aspen HYSYS version 8.0 using Peng-Robinson (PR) and Twu-Sim-Tassone (TST) equilibrium models. The results from the TST model are considered to be the most accurate for glycol dehydration.

Several parameters such as absorption pressure, glycol flow rate, number of absorption stages, and stripping gas flow rate were varied to simulate their effects on water removal efficiency.

The simulation results of a traditional triethylene glycol dehydration process showed optimum dehydration efficiency at a pressure of 5000 kPa with resulting water content of 129 ppm using PR and 105 ppm using TST model.

The traditional dehydration process was enhanced by introducing stripping gas to the reboiler and by adding an extra stripping column. Simulations show that injecting stripping gas to the reboiler can dehydrate gas down to 30 ppm. With an additional extra stripping column, water content was reduced down to about 1 ppm. A Drizo process was also simulated which calculated less than 1 ppm water in dehydrated CO₂.

Contents

Preface

This thesis is written as a part of the Master program in Energy and Environmental Technology at University College of Southeast Norway, during the spring semester 2016. The task description is presented in Appendix 1.

First and foremost, I wish to express my sincere gratitude towards my supervisor, Professor Lars Erik Øi. It would not have been possible to complete my work without his advice, guidance and supervision. I highly appreciate his knowledge, experience and significant contributions to CO_2 Capture research.

Above all, I want to thank my beloved family who motivated and supported me unconditionally throughout my two years stay in Norway. I am thankful to my colleagues and friends at University College of Southeast Norway for always being helpful and cooperative.

I will thank all the staffs from the Library of University College of Southeast Norway for providing books, databases and journal articles relevant to my thesis. I appreciate the IT Service for providing updated version of Aspen HYSYS and also for making the library electronic resources accessible freely on and off campus.

Last but not the least, I gratefully acknowledge all my country fellows for their friendly suggestions and guidance during my difficult times.

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Porsgrunn, 3rd June 2016

Birendra Rai

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Abbreviation

CCS	Carbon Capture and Storage
CO_2	Carbon-dioxide
MEG	Mono-ethylene Glycol
DEG	Di-ethylene Glycol
TEG	Triethylene Glycol
TREG	Tetra-ethylene Glycol
EOS	Equation of state
EOR	Enhanced Oil Recovery
ppm	Parts per million
PR	Peng-Robinson
TST	Twu-Sim-Tassone
SRK	Soave-Redlich-Kwong
CPA	Cubic-Plus-Association
HC	Hydrocarbon
BTEX	Benzene, Toluene, Ethyl benzene and Xylene
T	Temperature
VLE	Vapour-Liquid equilibrium
EIGA	European Industrial Gases Association

1 Introduction

1.1 Background of gas dehydration

The rapid advancement in energy and industrial sectors have increased the atmospheric concentration of carbon dioxide in the environment. Global warming and climate change are the hot issues in the present situation. Various researches and experiments have been carried out for the optimization of Carbon Capture and Storage (CCS). Also, to compensate the increasing demand for crude oil, the potential usage of gas in large scale applications has risen up. They are more focused on capturing harmful greenhouse gases of which CO_2 is the major one and make the best use of them as it will eventually contribute to controlling the global temperature.

Gas dehydration is the process of removing water from the gas stream. Before exporting CO_2 from the production area to the consumption locations certain water specifications of the produced/captured gas have to be fulfilled. Since all gas streams are fully/partially saturated at initial phase, gas streams need to be subjected to the dehydration process before supplying to the commercial market. The main target of this process is to avoid corrosion and hydrates formation in the transportation equipment.
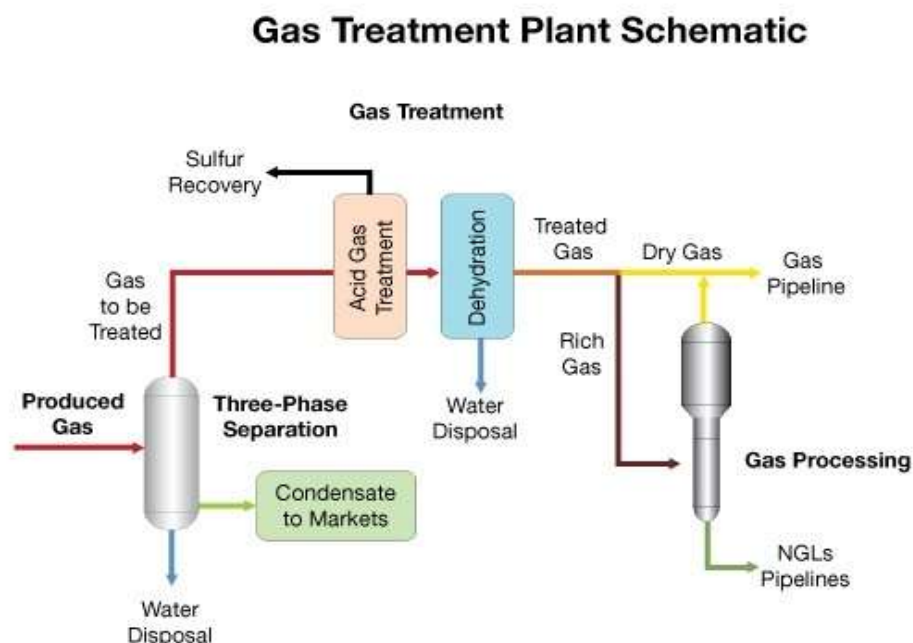
Figure 1.1: *Gas dehydration process as a part of Gas Treatment Plant*

The timeline below shows the historical background of gas dehydration [1]

1929 – Glycerol was the first liquid desiccant used for dehydrating fuel gas.

1930 – Calcium chloride solution was the first liquid desiccant used for natural gas dehydration.

1936 – Diethylene glycol was first used for dehydrating natural gas. Also, triethylene glycol was proved to be the most effective dehydrating desiccant.

1957 – At least 5,000 natural gas dehydration plants based on glycol absorption were estimated to be in operation in U.S. and Canada.

1997 – More than 20,000 plants are estimated to be in operation.

1.2 CO_2 dehydration

CO_2 dehydration process is same as natural gas dehydration process where the dominating gas will be carbon-dioxide. In recent years, CO_2 dehydration has become matter of interest due to the high demand of water free (pure) CO_2 in enhanced oil recovery projects.

The CO_2 gas streams are usually captured at low pressure from the carbon capture system. While transporting CO_2 through the pipelines or during the storage period, it has to meet certain specifications including optimum water dew point, maximum hydrocarbon dew point, allowable concentrations of solids content and contaminants. The presence of water vapour in the gas streams has been a major threat for the storage tanks and transportation pipeline walls.

The CO_2 dehydration processes are generally performed by;
- Absorption by liquid desiccants (Triethylene glycol)
- Adsorption by solid desiccants (Molecular sieve)
- Membrane processes
- Refrigeration

The water specifications, energy consumption, operating conditions, safety and environmental standards are the governing factors for the choice of dehydration. CO_2 dehydration based on glycol absorption is discussed in this work.

1.3 Corrosion and Hydrate formation

When CO_2 comes in contact with water, it is partly hydrated and carbonic acid (carbonates and bicarbonates) is formed. This is a weak acid with moderate pH value. At high pressure, the degree of dissociation increases which makes it stronger. It results in the corrosion of pipelines and compressor materials. Presence of impurities and inert will boost corrosion and also have the adverse effects on vapour/liquid equilibrium. In absence of free water in pure dense phase CO_2, corrosion rate in carbon steel is almost zero. For longer distances, use of highly corrosion resistant materials[1] (stainless steel) for transport pipeline would not be economically feasible. Safe transport and durability of transport infrastructures can be ensured below the water solubility limit (500 ppm) in CO_2 i.e. corrosion, hydrate formation and free water formation will be minimum [2]. Numerous modelling and experimental results based on CO_2 corrosion from various researches are reviewed in [3]. However, the definite mechanisms of corrosion in carbon steel CO_2 pipelines due to water have not been entirely understood. From the Figure 1.2, it can be observed that the corrosion rates are minimum for the sufficiently dry CO_2.
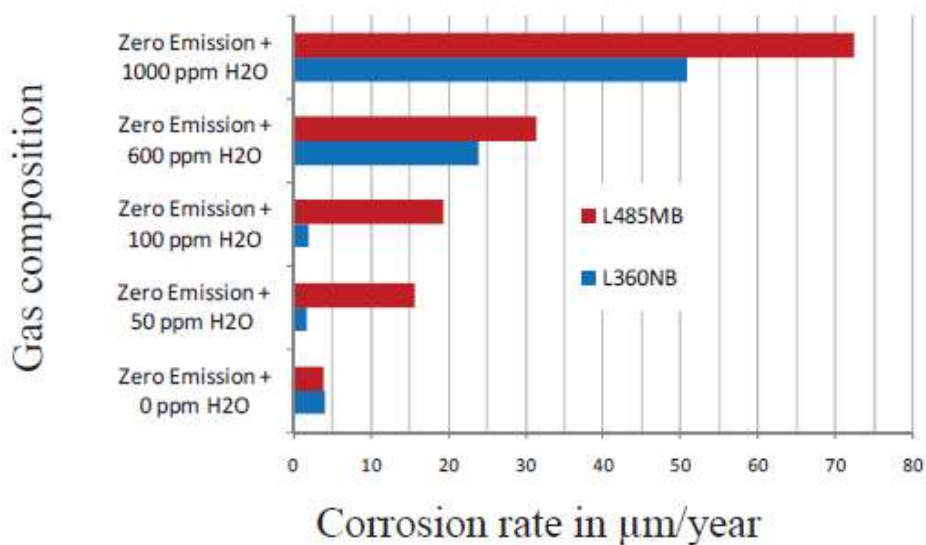
Figure 1.2: *Annual corrosion rates of two different specimens as a function of water content [4]*

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[1] Stainless steels are economically feasible only for wet CO_2 for a relatively small distance. (Sleipner project) [2]   E. Visser de, C. Hendriks, G. Koeijer de, S. Liljemark, M. Barrio, A. Austegard, *et al.*, "DYNAMIS CO2 quality recommendations," 2007.

Hydrates are solid, complex, crystalline compounds formed by the combination of free water with molecules like CO_2, H_2S, and CH_4. High pressure and low-temperature favour hydrate formation. Hydrates are generally formed at temperatures above the freezing point in both liquid and vapour phase. Methanol and other hydrate inhibitors are typically used to avoid hydrate formation in pipelines. However, due to their inability to reduce the water content as prescribed requirements, they are only used for relatively short distance where hydrate avoidance is crucial [5].
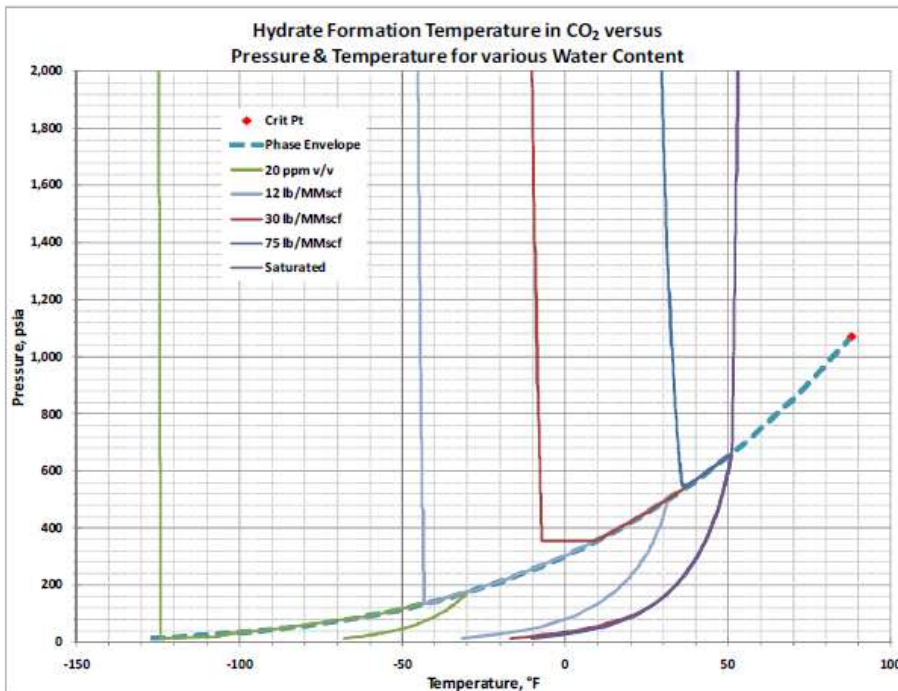
Figure 1.3: *Hydrate formation temperature as a function of water content [5]*

Corrosion of walls of the transmission pipelines as well as storage tanks and plugging of pipelines of the process equipment due to the hydrate formation are the serious operational problems of water vapour present in the gas streams [1]. Furthermore, it creates a dramatic reduction in the efficiency and capacity of the pipelines.

Appropriate selection of non-corrosive materials[2], regular monitoring programs, and injection of hydrate inhibitors could avoid corrosion and hydrate formation. But these

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[2] Use of non-corrosive materials (stainless steel) would be economically unfeasible for pipeline transportation beyond 1 km length.

preventive measures are often expensive and impractical based on process scheme and design considerations.

These problems can also be avoided by keeping the gas stream above the dew point. Therefore, it is essential to know the specific water vapour content of the gas and the conditions under which hydrates are formed [6]. On the other hand, the moisture prediction charts[3] [6] are also considered as a helpful medium to determine the quantity of water vapour at saturation condition with various pressure and temperature [5].Therefore, dehydration of CO_2 before transmission is almost inevitable. Although the dehydration process is relatively small part with respect to full CO_2 capture system, several challenges need to be addressed before full-scale deployment of the gas treatment plants [7].

1.4   Objective of the thesis

The main objective of this thesis is to simulate the CO_2 dehydration process by absorption in triethylene glycol (TEG) using Aspen HYSYS process simulation program. To evaluate the simulation results of numerous process conditions and compositions using Peng-Robinson (PR) and Twu-Sim-Tassone (TST) equilibrium models. Also, to demonstrate the potentialities of TEG dehydration process as an alternative to other expensive dehydration processes.

In the available literature, most of the dehydration processes in various oil and gas processing industries are developed for natural gas. There are no specific companies or vendors that claim supplying of TEG dehydration unit for CO_2 dehydration. Also, most of them have not even disclosed the dehydration capacity of their unit. Moreover, the equilibrium models are developed based on the relevant experimental data and field/operational data obtained from the natural gas. Therefore, several valid issues need to be resolved to achieve cost-efficient water concentration limit.

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[3] The chart is presented in Appendix 3. They are only applicable for acid gas streams and not for sour gas streams.

2 Literature review

2.1 Water specifications

The water content of gas is commonly referred in terms of water dew point. The water dew point is the temperature at which the water just initiates to condense. Determination of water content in the gas is an important aspect of gas dehydration process.

The water solubility in CO_2 is greatly influenced by the variation in pressure and temperature. From Figure 2.1, it can be seen that there will be a significant reduction in the solubility of water in CO_2 when CO_2 changes from liquid to vapour phase. But after certain pressure limit i.e. when there is a transition from vapour to liquid, there will be a considerable increase in the solubility of water in CO_2. When comparing the variation in pressure and temperature parameters, variation in temperature has the most prominent influence on the water solubility. The increment in pressure of magnitude 1 bar raises the water solubility by 3-4 ppm whereas, an increment in temperature of 1℃ raises the water solubility by 50 ppm [8]. Several evaluations for the prediction of solubility of water in CO_2 by the various equation of state models can be found in [9].
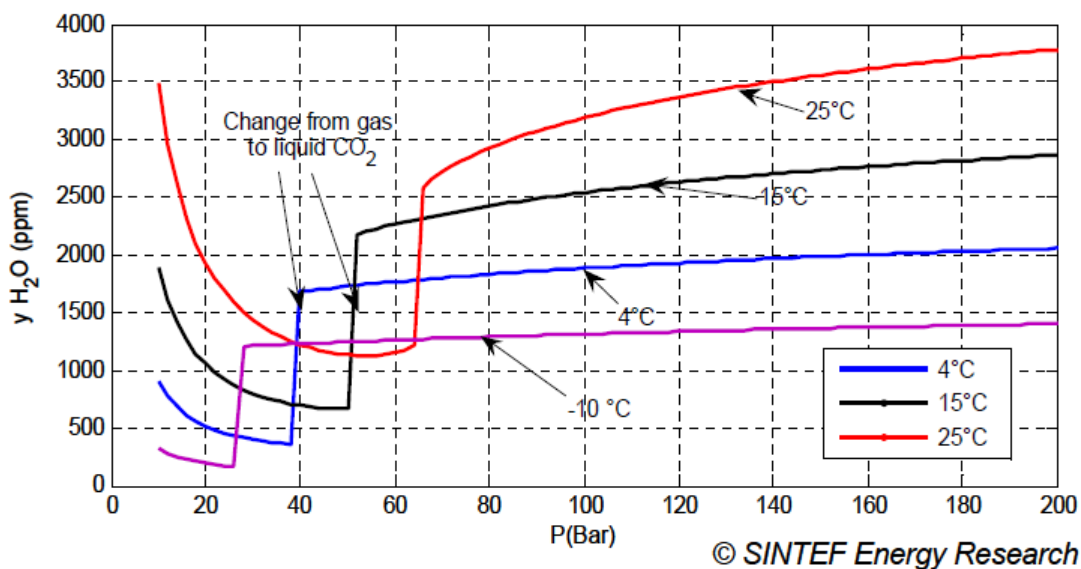
Figure 2.1: *Solubility of water in CO_2 as a function of pressure and temperature [2]*

Sufficient amount of water needs to be purified and dried to prevent formation of free water which facilitates the corrosion and hydrate formation. Since there has been so consensus about the exact limit for the water concentration, different specifications can be found for different locations and climatic conditions.

AMEC has analysed information from different capture processes and concluded the requirement of 550 ppmv typically for pipeline system (high ambient temperature) and 50 ppmv or < 10 ppmv for low temperature or cryogenic conditions [7, 10]. Water concentration of 50 ppm has been specified for the CO_2 pipelines in USA and Snøhvit pipelines in Norway. Also, Institute for Energy Technology has conducted a corrosion study for a planned sub-sea CO_2 transmission pipeline in Norway which specified the requirement less than 50 ppm [11]. At normal operating conditions, 500 ppm water is suitable to avoid the risk of free water formation and has claimed that lowering to 40 ppm will just increase the time and cost [12]. A liquid natural gas plant (Hammerfest, Norway) has a specification of 50 ppm for drying requirement of CO_2. Likewise, for Barendrecht project, the requirement of 40 ppm has been specified. Many CO_2 pipeline operators have accepted 500 ppm water content as a decent safety boundary for avoiding corrosion. However, other experts requested for more low water concentration of 50 ppm to be on a safe side at worst conditions [8]. Kinder Morgan, a large transporter of CO_2 has agreed on the maximum water content of 632 ppmv and 250 ppmv for their EOR systems and lower ambient temperatures through carbon steel pipelines respectively. Furthermore, they have even accepted 1580 ppmv in case of well traced and insulated plant pipelines [5]. However, Arne et al. have argued about the specifications claimed on DYNAMIS project (500 ppmv) and for the Kinder Morgan pipeline (650 ppmv) to be less conservative [13]. ENCAP project has recommended water limit of 5 ppm for severe limit case, 50 ppm for enhanced oil recovery and 500 ppm for pipeline transport. Yara Praxair transports CO_2 by ship at 50 ppm as specified by EIGA [14]. The requirements for removal of water to avoid corrosion and hydrate formation are discussed in [3, 15].

Some natural gas sales specifications are listed below: [6]

Southern U.S.A., Southeast Asia, Southern Europe, West Africa Australia –7 lb/mmscfd

Northern U.S.A., Canada, Northern Europe, Northern and Central Asia – 2-4 lb/mmscfd

2.2 Triethylene Glycol (TEG) as an absorbent

Glycol dehydration is the most widely implemented method in the oil and gas processing industry. The application of glycol in gas dehydration process seems to dominate the gas processing industry for a long period of time from both industrial and environmental aspect. Different companies/vendors have a variety of liquid desiccants/absorbents used for dehydration. Some commercially available absorbents for gas dehydration are given below;

- Mono-ethylene glycol (MEG)
- Di-ethylene glycol (DEG)
- Triethylene glycol (TEG)
- Tetra-ethylene glycol (TREG)

Among them, TEG is the most commonly used in natural gas dehydration and nowadays has been extensively used for CO_2 dehydration because of its characteristics like: [6]

- Low vaporization losses
- Extremely low solubility for salts
- High regeneration efficiency
- High thermal stability
- High affinity to water
- Low affinity to hydrocarbon
- Low viscosity

Table 2.1: *Physical properties of MEG, DEG, TEG, TREG [6]*

Glycol	MEG	DEG	TEG	TREG
Formula	$C_2H_6O_2$	$C_4H_{10}O_4$	$C_6H_{14}O_4$	$C_8H_{18}O_5$
Molecular weight [g/mol]	62.10	106.10	150.20	194.23
Boiling point @ 760 mm Hg, [℉]	387.10	427.60	532.90	597.20
Vapour pressure @ 77℉ [mmHg]	0.12	<0.01	<0.01	<0.01
Viscosity @ 77℉ [cP]	16.5	28.2	37.3	39.9
Decomposition temperature [℉]	329	328	404	460

2.3   Gas dehydration methods

The removal or reduction of water content from the gas is termed as gas dehydration. The main objective of dehydration is to prevent corrosion and hydrate formation. The principally known methods available for the gas dehydration are absorption, adsorption, membrane processes and refrigeration. The selection of dehydration methods is extremely dependent on water absorption efficiency, economical and technical feasibility and operational flexibility [16]. Some of the gas dehydration methods are described below.

Absorption

Dehydration by absorption is achieved by contacting saturated gas with liquid desiccants/absorbents having high chemical affinity to water. This method is accomplished in two different stages, gas dehydration and glycol regeneration. At first, gas is dehydrated in contact with glycol in an absorption column (contactor) and later, the glycol is regenerated in the distillation column (regenerator). Glycol is pumped back, recycled and reused for further dehydration process.

This method is further described in detail later in this work.

Adsorption

In the adsorption process, solid desiccants are used as an adsorbent to remove water from the gas. In general, water molecules are held by desiccants when brought in contact and are removed from the gas. This method can be classified into two categories, physical and chemical. In physical adsorption, dehydration takes place by the attraction of adsorbed liquid (water) into solid desiccants due to van der Waals forces. In chemical adsorption, dehydration takes place by the chemical bonding of adsorbed liquid (water) with solid desiccants [16].

Adsorbents must have following characteristics: [6]

- High adsorptive efficiency
- Easy and economic regeneration
- High rate of adsorption
- Large effective surface area per weight
- Resistance to crushing and dust formation

Some of the commercially used adsorbents are activated alumina, silica gel, alumina gel and molecular sieves [6]. Adsorption process is favoured by low temperatures and high

pressures. It has the capability to achieve water outlet temperature (T_{dew}) < -50°C. This method is generally considered when the dryness requirement of the treated gas is very low[4]. However, it requires high capital cost and space area.
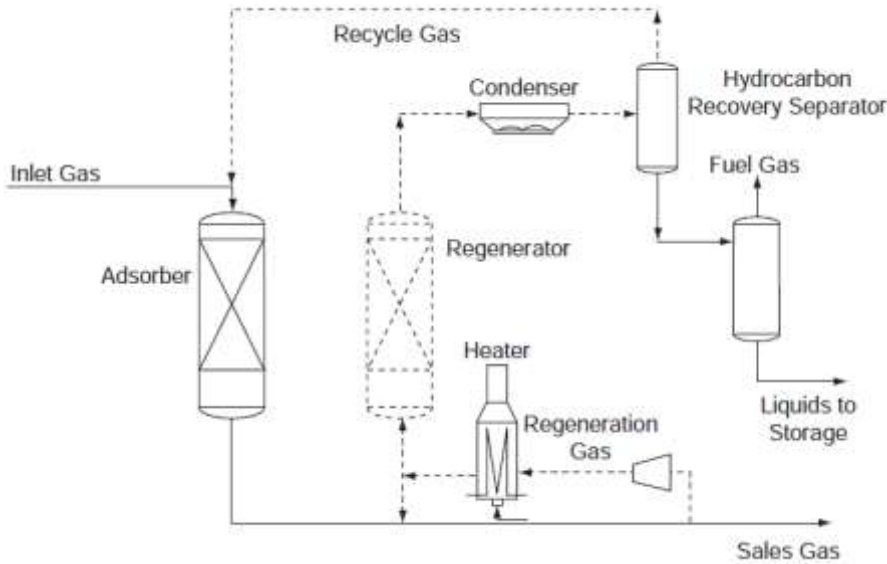
Figure 2.2: *Adsorption process [17]*

Membrane processes

In this process, membranes have an important role for the gas dehydration. Differences in the permeability of the gases enhance this process. Gas permeates through the membrane due to the differences in partial pressure. Also, the permeability varies from one gas to other, gas separation is possible. Water vapour is the fastest permeating component. But the small differences in permeability between these gases can restrict from absolute separation . Thus, this method is economically feasible only for the small plants (low gas flow rates).

Development of more efficient and chemically robust membrane materials has been the main challenge for membrane processes technology [18].

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[4] Adsorption is mainly preferred when the requirement of the water concentration in the gas stream is lower than 1 ppm i.e. when gas is liquefied.
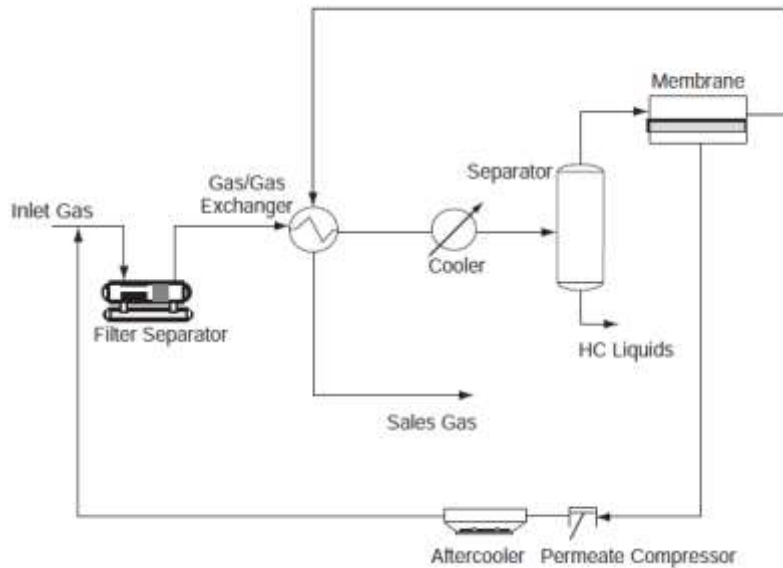
Figure 2.3: *Membrane process [17]*

Condensation (cooling)

In this method, refrigeration system is installed with of purpose of lowering the temperature of the gas for water removal [19]. Refrigeration unit can either consist of mechanical refrigeration or expansion refrigeration depending upon refrigerant flow rate and pressure [20]. This process is mainly practised in places with hot climates where formation of hydrates is unlikely and water specification is not too strict.

Ifpexol^{TM} is a patented technology which operates as simultaneous dehydration/dew pointing process. Methanol is used as hydrate inhibitor generally injected upstream of the heat exchanger [19] and water dew point can be lowered to -80°C [20].
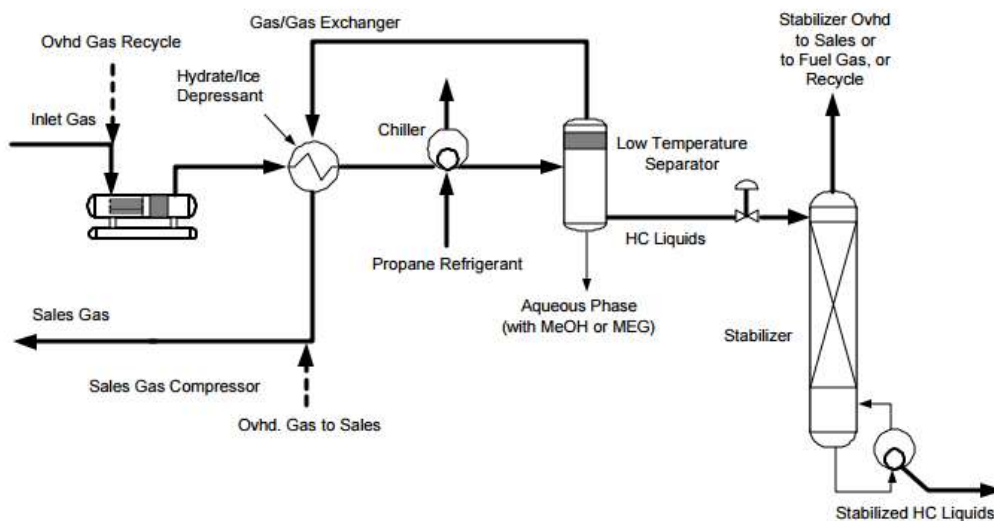
Figure 2.4: *Mechanical Refrigeration (Condensation) process [17]*

In a gas processing plant, these processes can be combined with each other to ensure optimum dehydration of the gas stream.

2.4 TEG dehydration alternatives

Normally 98.6 wt% can be achieved by the standard regeneration system. So, when the glycol concentration requirement is higher than 98.6 wt%, stripping gas injection, Drizo, Coldfinger processes are some the best available alternatives. These methods are the improvisation of standard dehydration process, mainly focussed on increasing the glycol regeneration to improve process performance by reducing the partial pressure of water in gas phase. Some common enhanced dehydration processes are described below.

Stripping gas

Gases like nitrogen, carbon dioxide or flash gas can be used as stripping gas [21]. The gas is either injected into the liquid in the reboiler or in an extra stripping column. Detail description about this topic can be found later in this work.

Vacuum dehydration

The principle of vacuum dehydration is that the partly regenerated glycol from the distillation column is further reheated to 400ºF before feeding into a vacuum drum. The partially condensed vapour from the vacuum drum is pumped into the regenerator. This process can achieve 99.9% by weight TEG concentration [1]. This process is rarely in practice due to its high operating cost and problems encountered with achieving the needed vacuums [6].

DRIZO®

Drizo is licensed by PROSERNAT which has been patented as an alternative to traditional dehydration unit [22]. The working principle of this process is similar to that of conventional TEG dehydration system till the rich glycol enters the regenerator. Generally, superheated HCs are used as stripping gas[5] which are recovered and recycled for reuse. The detail process is described later in this work.

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[5] In this work, benzene, toluene, and n-heptane are used as stripping gas.

This method is usually in practice when the glycol purity requirement is 99.998% without the use of external stripping gas [20]. There is a possibility of recovering surplus liquid aromatics by this process. This process is environmental friendly as it can reduce the BTEX (Benzene, Toluene, Ethyl benzene and Xylene) and CO_2 emissions into the atmosphere. It allows dew point depressions up to 110ºC (180ºF). This is the cheapest technology to achieve very low water dew point. More than 60 gas dehydration units have been installed worldwide till date. Being similar to the traditional process it can be incorporated into the existing glycol units [20]. The disadvantage of this process is that additional equipment is required for the regeneration of the liquid solvent and a pump to recirculate the stripping gas. The Drizo process is approximately 20% cheaper than glycol stripping unit and 50% cheaper than molecular sieve unit [23].

COLDFINGER®

The working principle of the Coldfinger process is to condensate and remove water from the partly regenerated glycol [24]. The lean glycol is sent into a two-phase tank occupying half volume. A bundle of tubes is inserted into the other half part, occupied by the vapour of water and glycol. This initiates condensation process of equilibrium vapour and begins discharging the condensate. The water in lean TEG starts to evaporate in order to restore the vapour equilibrium condition resulting to high concentration of glycol. This process can retain 99.7% by weight TEG concentration [23]. If triethylene glycol is to be used then this process is most efficient at working condition of 400ºF and 40 to 45% by weight TEG and 55 to 60% water [23].

This is the one of the most reliable oil and gas dehydration and glycol regeneration processes. High concentrated glycol can be achieved without the use of stripping column. COMART is the licensor of this process [25].
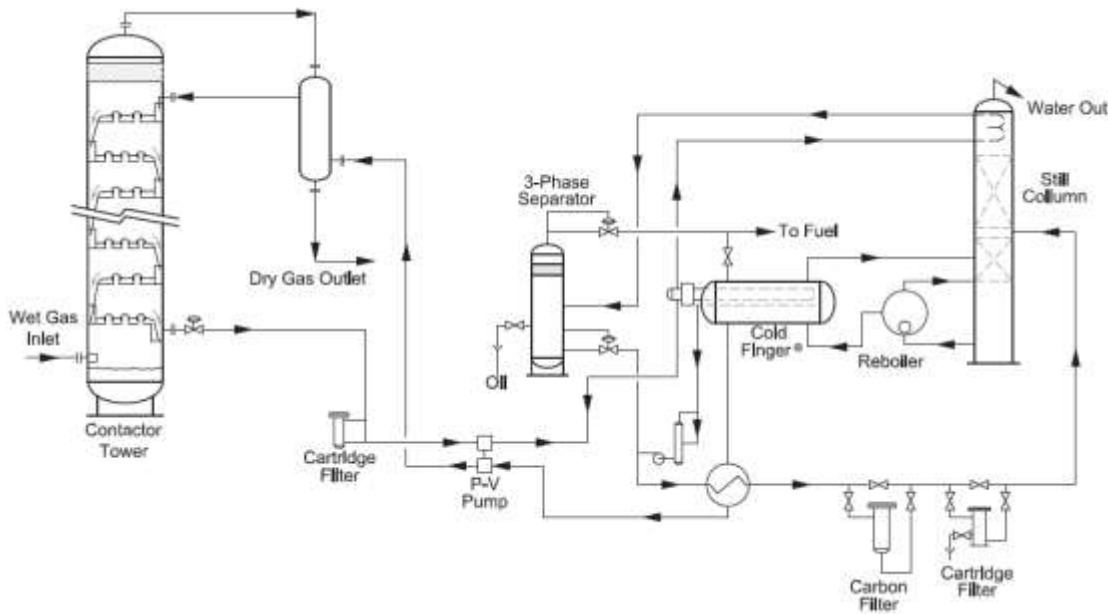
Figure 2.5: *Coldfinger condenser process [6]*

2.5 Vapour/Liquid equilibrium models

The equation of state can be termed as the general description of interconnection between thermodynamic variables like pressure, temperature, and volume for fluids [24]. Improvisation of equation of states has helped to develop the models for mixture components than being limited to pure components. Equilibrium models have been most reliable for the prediction of fluid properties at different operating conditions. There have been several efforts in developing a precise and most reliable equation of state models and mixing rules for associating fluids [26]. It is essential to calculate the properties of pure CO_2 and CO_2 in mixtures precisely.

At the preliminary time, concentration difference exists when liquid and vapour comes in contact with each other, and after certain interval, an equilibrium stage is achieved. This concentration difference assists separation of fluid mixtures and therefore, quantitative prediction of the equilibrium properties of the mixtures has to be made. These predictions are very crucial to represent the TEG-water phase equilibrium for the successful design of the typical TEG dehydration process found in chemical and refining industries [27]. Cubic equations of state have ease these complex phase equilibrium calculations. The non-ideal TEG-water systems can be correlated accurately by describing the liquid phase by an activity model, the gas phase by an equation of state and the total pressure dependence by Poynting correction [28].

There have always been uncertainties in proposed TEG/water equilibrium data. Furthermore, use of accurate equilibrium data has increased the overall bubble cap tray and Murphree efficiencies in absorbers [28]. The accurate calculation of solubility of CO_2 in a TEG/water solutions is essential.

Peng-Robinson, Glycol Package (TST) and Soave-Redlich-Kwong (SRK) are some recommended equation of state models for gas dehydration using glycol [29].

Peng Robinson

Peng-Robinson model is a standard cubic equation of state which is ideal for calculations of vapour/liquid equilibrium as well as liquid densities for hydrocarbon systems [29]. This model was developed by D. Peng and D.B. Robinson specifically focused for modelling of natural gas systems. Later several improvisations were made to outspread the application area and for predictions of some non-ideal systems [30]. It is relatively simple for VLE calculations. The PR property package is more reliable and efficient within a temperature range of $> -271°F$ or $-456°F$ and pressure range of < 100 MPa or 15 kpsia [30].

The Peng-Robinson equation of state is given by [31, 32]

$$Z = \frac{V}{V-b} - \frac{aV}{RT(V^2 + 2bV - b^2)} \tag{1}$$

Which can also be written as,

$$Z^3 + (B-1)Z^2 + (A - 3B^2 - 2B) + (B^3 + B^2 - AB) = 0 \tag{2}$$

where,

$$Z = \frac{PV}{RT}$$

$$A = \frac{aP}{R^2 T^2} \tag{3}$$

$$B = \frac{bP}{RT} \tag{4}$$

$$V = \text{molar volume}$$

For the standard mixing rules of multicomponent systems:

$$x = \sum_i x_i \tag{5}$$

$$a = \sum_{ij} \sqrt{a_i a_j} \left(i - k_{ij}\right) x_i x_j \tag{6}$$

$$b = \sum_i x_i b_i \tag{7}$$

$$a = 0.45724 \frac{R^2 T_c^2}{P_c} \alpha \tag{8}$$

$$b = 0.0778 \frac{RT_C}{P_C} \tag{9}$$

$$Z_c = 0.307$$

$$\alpha = \left(1 + k\left(1 - \sqrt{T_R}\right)\right)^2 \tag{10}$$

$$k = 0.37464 + 1.54226\omega - 0.26992\omega^2 \tag{11}$$

T_C = critical temperature, P_C = critical pressure

$$\omega = \text{acentric factor} = \left[-\log\left(\frac{P^{SAT}}{P_C}\right)_{T_{R=0.7}}\right] - 1$$

R = gas constant, $T_R = T/T_C$, T = absolute temperature

Although being simple, reliable and efficient model for gas oil and petrochemical applications [29], it has some limitations and is not sufficiently accurate. Because of its only one adjustable parameter for each binary component pair in the system, it is very challenging to predict vapour/liquid equilibrium data precisely [21]. However, due to the better performance at the critical conditions, PR EOS is preferred over SRK EOS.

TEG Dehydration, Cryogenic Gas Processing, Vacuum Towers, Hydrate Inhibition are some of the simulation processes that use Peng-Robinson property package [29].

The binary interaction parameter for Peng-Robinson EOS in Aspen HYSYS program is 0.04450 (for $K_{H2O/CO2}$), whereas, Austegard et al. [9] have suggested 0.193. There is a deviation in the parameter values.

Table 2.2: Peng-Robinson parameters [29]

Setting	Options
Enthalpy	Property Package EOS or Lee-Kessler
Density	COSTALD, Rackett (recommended for petroleum and hydrocarbon liquid mixtures at low and moderate pressure) or use EOS Density
Modify Tc, Pc, for H2, He	Modify or un-modified
Indexed Viscosity	HYSYS or Indexed
Peng-Robinson Options	HYSYS or Standard
EOS Solution Methods	Cubic EOS Analytical or Numerical Method
Phase Identification	Default - If Default you can modify the tuning factor Venkatarathnam-Oellrich - Uses a thermodynamic method for determining the phase of a fluid from the partial derivatives of pressure, volume and temperature without reference to saturated properties. Particularly useful for liquid-liquid or vapor-liquid-liquid equilibria calculations.
Surface Tension Method	HYSYS or API 10A3.2 Method
Thermal Conductivity	API 12A3.2-1 or API 12A1.2-1 Method

Glycol Package (Twu-Sim-Tassone)

The glycol package contains TST equation of state for determining the phase behaviour of TEG-water binary system. The TST model was developed by Twu, Sim and Tassone considered as an advanced cubic equation of state [33]. This EOS was developed for the better predictions of properties of polar and heavy components and G^E model. It allows description of highly non-ideal mixtures in combination with the mixing rules over a broad range of pressures and temperatures. The TST equation of state is considered more consistent and accurate for the prediction of activity coefficients of the TEG-water solutions (2% deviation), dew point temperatures (±1ºC error) and water content of gas (1% deviation) in natural gas systems [30].

The property package has a large range of application in terms of temperatures (15-50ºC for dehydrator and 202-206ºC for regenerator), pressures (10-100 atm for dehydrator and 1.2 atm for regenerator) and component concentration encountered in glycol gas dehydration system. The TST model possesses more adjustable parameters like vapour liquid equilibrium can be correlated by using three adjustable parameters [30].

It uses Cavett model for entropy and enthalpy calculations [29].

The TST cubic equation of state is given by: [34]

$$P = \frac{RT}{v-b} - \frac{a}{v^2 + 2.5bv - 1.5b^2} \tag{12}$$

where,

$$a_c = 0.470507 \, \frac{R^2 T_C^2}{P_C} \tag{13}$$

$$b_c = 0.0740740 \, \frac{RT_C}{P_C} \tag{14}$$

$$Z_c = 0.296296$$

$$c = \text{critical point}$$

For the mixing rules of multi component system;

$$\frac{G^E}{RT} = \sum_i^n x_i \, \frac{\sum_j^n x_j \tau_{ji} G_{ji}}{\sum_k^n x_k G_{ki}} \tag{15}$$

$$\tau_{ji} = \frac{A_{ji}}{T} \tag{16}$$

$$G_{ji} = \exp(-\alpha_{ji} \tau_{ji}) \tag{17}$$

where,

τ_{ji} and G_{ji} are binary interaction parameters

Table 2.3: *Glycol package parameters [29]*

Calculation	Option
Vapor Enthalpy	Property Package EOS - TST EOS with infinite-press mix rule. (Default) Lee Kessler TST EOS - TST EOS with van der Waals mix rule.
Liquid Enthalpy	Cavett Proprietary Package EOS (Default) Lee- Kessler
Density	Use EOS Density COSTALD (Default)
EOS Solution methods	Cubic EOS Analytical (Default) Newton-Raphson
Phase Identification	Default - If Default you can modify the tuning factor Venkatarathnam-Oellrich - Uses a thermodynamic method for determining the phase of a fluid from the partial derivatives of pressure, volume and temperature without reference to saturated properties. Particularly useful for liquid-liquid or vapor-liquid-liquid equilibria calculations.
Surface Tension Method	HYSYS or API 10A3.2 Method
Thermal Conductivity	API 12A3.2-1 or API 12A1.2-1 Method

Table 2.4: *Binary interaction parameters for use in TST model (TEG/H$_2$O) [29, 34]*

Binary	Aspen HYSYS (default)	Twu et al.
A_{12}	-281.200	-141.49000
A_{21}	314.300	158.16600
B_{12}	0.50570	0.2554489
B_{21}	11.5900	5.8338000
α_{12}	0.27890	0.2788790

From Table 2.4, it can be seen that there are deviations in the values obtained from Aspen HYSYS (default) and Twu et al.[34]. The values for A and B from Aspen HYSYS are very close to twice the values suggested by Twu et al. However, the default alpha (α) value of Aspen HYSYS is almost same to that of Twu et al.

Determination of these binary interaction parameters is significant to improve the accuracy of cubic equations. Generally, they are determined by comparing the predicted values with experimental data.

2.6 Previous work of TEG dehydration of natural gas

The use of glycol for natural gas dehydration has been the most demanding process over many years. This method is fundamentally recognised for its optimum functionality and versatility. Mentioned below are some previous work of TEG dehydration of natural gas.

Bråthen [35] compared the simulation results[6] of TEG injection process and conventional absorber dehydration and concluded that to dehydrate water content to the same amount, TEG injection requires approximately 50% more TEG circulation, more energy and releases hydrocarbon higher than conventional dehydration. He stated TEG is more appropriate than MEG and DEG for gas dehydration. The dehydrated gas including flash gas was used as stripping gas and its few drawbacks including their probable solutions were highlighted. Aspen HYSYS with CPA equation of state as fluid package was used to develop the model.

Christensen [36] conducted the thermodynamic simulation of water/glycol mixture using Peng-Robinson and Glycol Package equation of state. He stated inserting a component splitter to be the easiest way for simulation of a dehydration plant. Glycol was used as absorbent and pure nitrogen as stripping gas. He claimed the requirement of stripping gas to achieve 99.6 wt % TEG purity.

Ryba [37] performed simulation of TEG dehydration of natural gas using Peng-Robinson EOS in Aspen HYSYS. He also claimed the requirement of stripping gas to achieve 99.5 wt % TEG purity. Approaches to limit the TEG losses and energy consumption during the dehydration process were presented. He claimed that for the dew point temperature of 5ºC and pressure 4000 kPa water content of 220 ppm in natural gas will be adequate to avoid corrosion during summer season and likewise, for −10ºC and pressure 4000 kPa, water content of 75 ppm will be sufficient.

Emah [27] conducted process simulation of natural gas dehydration in both Aspen HYSYS and Aspen Plus. In Aspen HYSYS; Peng-Robinson, Glycol Package and Soave-Redlich-Kwong models were compared whereas, in Aspen Plus; SR-POLAR and PSRK models were compared. The minimum water content achieved was 21 ppm by using

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[6] Results obtained from Dehydration performance, Energy consumption and HC emission. The input data were from Snohvit LNG.

Glycol Property package at TEG purity of 99.99 wt%. Also, above 10 absorption stages, only slight removal of water in the dehydrated gas was observed.

Ghati [38] simulated TEG dehydration process[7] using Peng Robinson. TEG dehydration method was able to achieve the targeted water content of $7.9*10^{-5}$ mole fractions of water corresponding to 98.2% absorption capacity. She recommended addition of stripping gas to increase the TEG purity from 99% to 99.95%. The influence of a number of equilibrium stages in the absorption capacity was emphasized.

Hansen et al. [24] performed simulation using glycol property package in Aspen HYSYS. They concluded that addition of stripping gas (Nitrogen) resulted in 99.6% TEG purity compared to 98.85% without stripping gas and possibility of removing 99% of water from the gas. They recommended glycol package to be best suitable for TEG dehydration.

Husby [39] simulated natural gas dehydration process using software like Pro/II (V9.1), Aspen HYSYS (V8.3) and ProMax (V3.2). The Glycol Package was used in Pro/II, Glycol Package, Peng-Robinson and two versions of the Cubic-Plus-Association-model (CPA) developed at Technical University of Denmark (DTU, V3.8) and Statoil (NeqSim) were used in Aspen HYSYS and Soave-Redlich-Kwong (SRK) and PR were used in ProMax. He stated CPA NeqSim to be the appropriate fluid package for dehydration of natural gas in Aspen HYSYS using TEG as an absorbent. Field data were provided by Statoil from Gullfaks.

Kinigome et al. [40] mentioned the easiness in natural gas transport and storage when converted into liquid. Operation of TEG dehydration plant would be more economical and efficient than conventional process using molecular sieve. Recommended Aspen software to run a simulation the TEG dehydration. They concluded operation of dehydration unit with TEG to be cheapest and efficient in comparison to molecular sieves.

Aboudheir et al. [41] conducted optimization study of natural gas dehydration with a target of <10 ppm water content and −50ºF dew point for the pipeline transportation. They stated that it can be achieved by using just molecular sieve system or with TEG/molecular sieve system combined.

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[7] Input data were from Songo Songo gas field in Tanzania.

2.7   Previous work of TEG dehydration of carbon dioxide

Only a few references were found on simulation of dehydration of CO_2 based on absorption in glycol. Because of very little information provided in open literature, the accurateness and effectiveness of the calculated results are uncertain. Some of the previous works are presented below.

Farhat [42] performed simulation of TEG dehydration of CO_2. Portion of CO_2 recovered during the absorption was used as stripping gas. The TEG regeneration was achieved by binary liquid distillation followed by stripping. The targeted water content of the dehydrated gas was 20 ppm.

Mirela [43] simulated both traditional and enhanced TEG dehydration of CO_2 with/without stripping gas and with/without extra stripping column. She used both Peng Robinson and Glycol Package EOS. Less than 5 ppm and 50 ppm water content in the dehydrated gas was achieved by introducing stripping gas to the extra stripping column and to the reboiler respectively. High concentration[8] of TEG was achieved with stripping gas to the extra stripping column compared to stripping gas to the reboiler.

Shrestha's [44] work was similar to Mirela. The simulation results were verified with Mirela and found to be acceptable. In addition, number of stages were varied above the rich TEG feed, below the reboiler and between feed and reboiler in the regenerator column. No significant increase in dehydration efficiency was observed when the stages between stripping gas feed and reboiler heat addition was increased above 4. The non-smooth curve was plotted while varying number of stages above the feed whereas, the smooth curve was obatined when the number of stages was varied below the reboiler.

2.8 Industries and TEG dehydration unit vendors

The reduction in water content of natural gas by dehydration method has been significant manoeuvre in the gas processing industry. There are many companies worldwide that design, build and commission glycol-based gas dehydration unit mainly devoted to

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[8] 99.84% TEG purity – stripping gas to the reboiler, 99.94% TEG purity – stripping gas to the extra stripping column.

dehydration of natural gas over CO_2 gas. Listed below are some companies that supply TEG dehydration units for natural gas.

- **FRAMES** (Netherlands)
- CAMERON (USA)
- BS & B Process Systems (USA)
- QB Johnson Manufacturing Inc. (USA)
- Aker Solutions (Norway)
- PROSERNAT (France)
- Process Group Pvt. Ltd. (USA)

No distinct evidence has been found on supply for CO_2 dehydration unit except FRAMES (Netherlands). Frames claim to remove water from both natural gas and CO_2. Moreover, very little information is provided on glycol based CO_2 dehydration process.

Table 2.5: *Specifications and Technical details of Stublach Gas Storage Project UK carried out by* **Frames** *[45]*

2 gas dehydration trains comprising the following:	Gas dehydration trains:
Slug catcher with integrated inlet separator	Gas flow rate: 8.25 MMNm^3/day/train
Dehydration tower	Glycol type: TEG
Filter skid with high-pressure TEG filters	Design pressure: 98 barg
Heat exchanger skid with shell and tube heat exchanger	Regenerator type: OVC
20 interconnecting piping	Operating temperature: 16-40℃
3 glycol regeneration units, with TEG reboiler and economizer IC	Inlet water content: 700 mg/Nm^3
Pump skid with 3 high-pressure TEG pumps	Outlet water content: <30 my/Nm^3
Hot water heating package with two gas-fired 2.8 MW boilers	

Table 2.6 represents the lists of some the case studies of the projects carried out by **Process Group Pvt. Ltd.**

Table 2.6: *List of Projects [46]*

Projects	TEG purity [wt%]	Outlet water content [lb/mmscf]
Jambi Merang Gas Production Facility (Indonesia)	99.60	< 10
ICP-R Process Platform Project (India)	99.70	< 5
PNG LNG Associated Gas Related Projects	99.89	5
Platong Gas II Development Project (Thailand)	99.80	2.5
Fairview CS3 Project (Australia)	99.60	< 4
GLNG Upstream Project (Australia)	99.65	4

Some performance details of several dehydration processes from **Prosernat**:
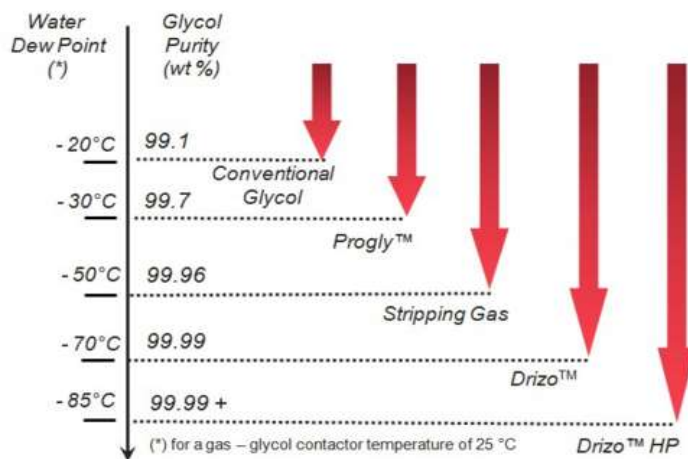
Figure 2.6: *Water dew point depression in accordance to TEG purity [20]*

3 Process description

3.1 Selection of CO₂ dehydration process

The appropriate dehydration processes are often selected based on dew points requirements, investment costs, safety standards, dehydration capacity, and energy consumptions. In this work, among several dehydration methods as presented in section 2.3, only absorption process using triethylene glycol and the procedures to enhance this process are discussed.

Traditional dehydration process is the most common process and proven technology for dehydration process. This process is simulated in Aspen HYSYS software. This process has been very well known for water removal in natural gas.

Since the traditional TEG dehydration process cannot achieve high dew point depressions, the process is improved by feeding stripping gas to the reboiler and by adding extra stripping column.

3.2 Traditional TEG dehydration process

The TEG dehydration unit as shown in Figure 3.1, consists of a contactor, flash tank, heat exchangers and a regenerator.
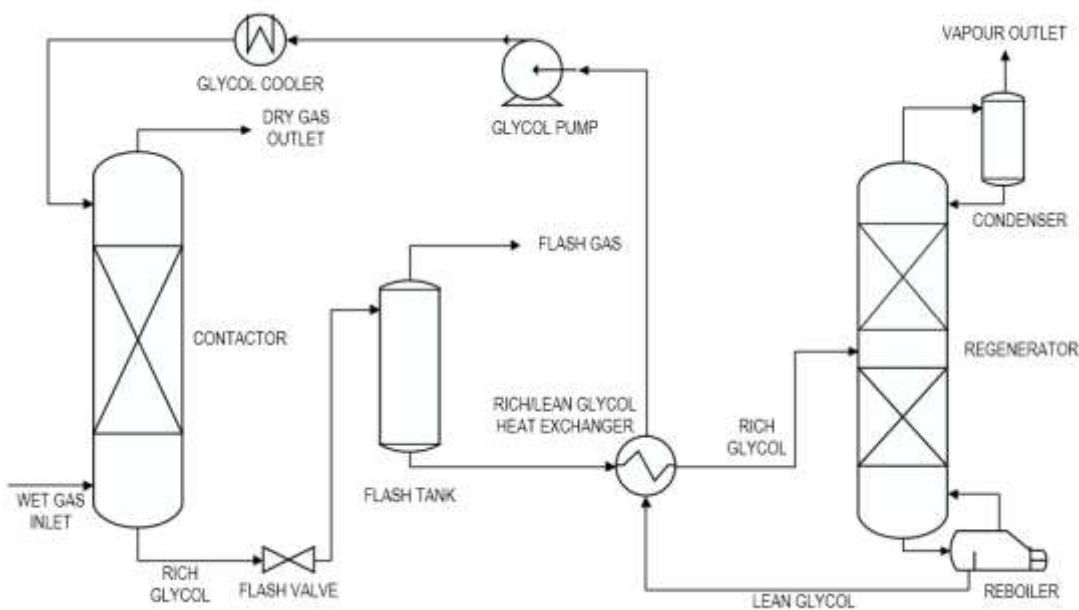
Figure 3.1: *Traditional Triethylene glycol dehydration process*

This absorption process proceeds with the low-temperature wet gas (saturated CO_2) being fed to the glycol contactor (absorption column) from the side bottom and lean TEG from the top. At contactor, high temperature can result in high vapour loss of TEG [47]. In a contactor, there will be the counter-current flow of TEG and the wet gas. In this column, due to the absorption of water present in the CO_2 by TEG, glycol is enriched by water which is often referred to as "rich glycol" and exits out of the bottom part of the contactor. Dehydrated CO_2 escapes out from the top as dry gas. The glycol absorber may contain either structural packing or bubble cup or valve trays. The volatile organic compounds are also absorbed by TEG, which are vaporized including water in the reboiler [47].

The glycol circulation rate plays a vital role in dehydration process as over circulation can result in overloading of reboiler preventing good glycol regeneration while under circulation rate can generate problems with tray hydraulics and contactor performance [47].

The stream is then depressurized by the valve and heated before passing it to the regenerator. The saturated CO_2 with water can also be removed in the flash tank. Heat exchange takes place between cooler rich glycol and hotter lean glycol in heat exchanger before passing into the regenerator. Maximum reboiler temperature is normally limited to 204ºC in order to prevent glycol thermal decomposition. Regenerator operates almost at atmospheric pressure.

In the combination of desorption column and reboiler, the mixture separation takes place because of the high temperature as boiling point of TEG much higher (285ºC) [47] to that of water and TEG is regenerated. The regenerated glycol flows down to the packed bed section into the reboiler. As the stream enters, absorbed water is stripped out of the glycol as it rises up the packed bed. Therefore, water vapour escapes out from the top and the regenerated TEG will flow out from the bottom. Some amount of glycol can be vaporized due to the high temperature which can be prevented by installing reflux condenser at the top of the column.

The hot regenerated lean glycol again passes through the heat exchanger where it loses its temperature to the rich glycol before feeding it into a glycol pump. The main function of glycol pump is to raise the pressure similar to that of absorber column and also to drive the stream throughout the circulation path. In glycol cooler, the lean TEG is cooled to maintain the efficiency in the contactor. The lean glycol enters the contactor and the absorption process continues again.

3.3 Enhanced TEG dehydration process

Higher the purity of TEG, greater will be the water absorption capacity. Glycol can be regenerated to higher purity either by increasing the reboiler temperature or by introducing stripping gas to the reboiler. In this work, enhanced TEG dehydration process is simulated by injecting stripping gas into the reboiler and by adding extra stripping column.

Maximum concentrations achievable in an atmospheric regenerator operating at a decomposition temperature of 204°C for TEG will be 98.8 mass percent. Due to the risk of thermal decomposition of TEG, increasing the reboiler temperature does not seem to be feasible. Also, regeneration column cannot be operated below atmospheric pressure because of the risk of explosion due to the oxygen leakage into the system [21]. Introducing stripping gas will have a much greater outcome than increasing reboiler temperature. Moreover, for the efficient use of stripping gas, it should be introduced in an extra stripping column after the hot glycol is removed from the reboiler [48].

All these methods are related to the principle of effective reduction of partial pressure of water in the vapour space of the reboiler, and hence obtaining a higher regeneration concentration at the specified temperature.

3.3.1 Stripping gas to the reboiler

Among various glycol regeneration processes, injecting stripping gas into the regeneration column as shown in Figure 3.2, is the simplest and most common employed process for the enhancement of glycol concentration. The enhancement of the glycol is basically performed before being pumped to the absorption column.
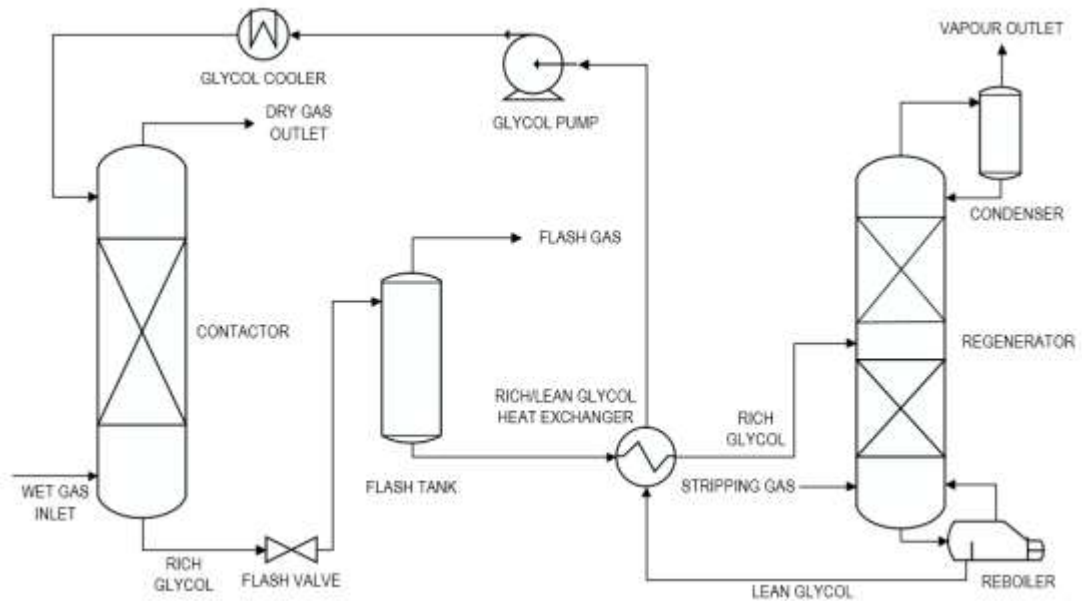
Figure 3.2: *TEG dehydration process with stripping gas to reboiler*

Even a small portion of stripping gas can take up large amount of water at the maximum regeneration temperature (204ºC) and near the ambient pressure. Adding stripping gas directly into the reboiler lowers the partial pressure of the water in the vapour phase. Furthermore, the concentration of water in the liquid phase decreases and the TEG concentration increases [49].

This process is mostly considered for the requirements of the TEG purity between 99.1 and 99.6 mass percent [1]. 99 % TEG purity will give the order of magnitude approximately 150 ppm in the treated gas while, 99.6% will give 50 ppm in the treated gas [33].

3.3.2   Stripping gas to the extra stripping column

This process is considered when the glycol purity requirement is above 99.6 mass percent. This process is also similar to the traditional process with an extra stripping column incorporated below the reboiler (Stahl method). In doing this, more effective use of stripping gas can be done to achieve the specified water concentration in the treated gas.

A schematic drawing of this process is illustrated in Figure 3.3,
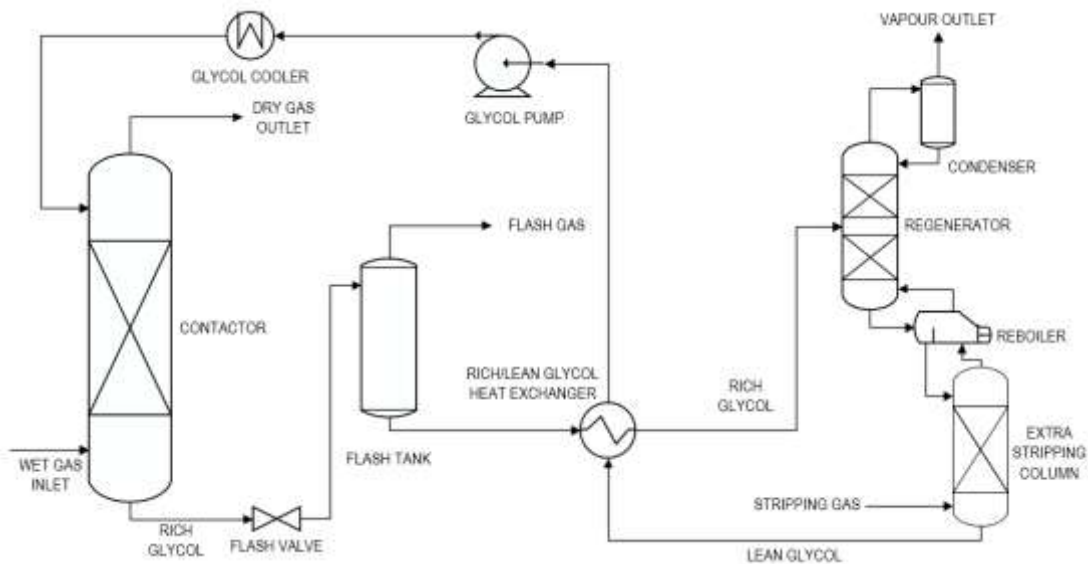
Figure 3.3: *TEG dehydration process with an extra stripping column in the TEG regeneration system*

The stripping gas is introduced into the bottom of the extra stripping column. Being the hot glycol flowing down the column from the reboiler and stripping gas moving upwards, there will be counter-current contact between them. This causes the water present in TEG to be gradually transported to the gas phase.

This process can obtain the TEG purity of 99.99 mass percent [1].

3.3.3 Drizo

The stripping medium required by this process is usually achieved from the BTEX (Benzene, Toluene, Ethylbenzene and Xylene) compounds present in the natural gas which are recovered from the feed gas during the dehydration process or can be n-heptane which is generally introduced externally [21]. These compounds are used as stripping solvent which can be recovered, recycled and reused again.

A schematic drawing of this process is illustrated in Figure 3.4,
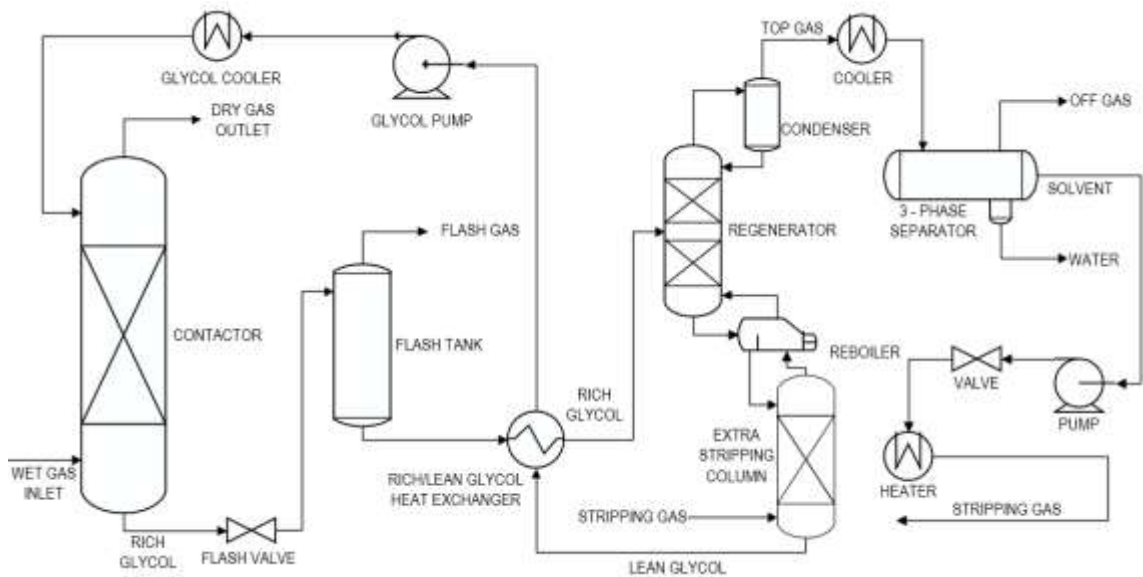
Figure 3.4: *TEG dehydration process by Drizo process*

The working principle of this process is similar to that of traditional TEG dehydration process till the rich glycol enters to the regenerator. This process uses recoverable and vaporized liquid hydrocarbon solvent to contact the glycol. The water vapour and solvent from the regeneration column are taken overhead and recovered as a liquid by the condensing process in a three phase separator. Here the water is discharged, the off gas is vented into the atmosphere and the solvents are recycled and pumped back into the bottom of the regenerator for reuse as stripping medium via a valve and heater.

In this work, TEG is used as a dehydrating agent and benzene/toluene/n-heptane are used as a regeneration solvent which is added externally. Also, loop for recirculation of n-heptane is not closed because some portion of n-heptane is dissolved in the solvent. However, this argument does not influence much for the evaluation of the process [50].

This process can achieve TEG purity of 99.998 mass percent [20].

4 Process simulation

Aspen HYSYS is a comprehensive process simulation tool used by the oil and gas producing companies for process modelling and optimization in design and operations [51]. It was developed the company named Aspen Technology.

Aspen HYSYS, Aspen Plus, PRO/II and ProMax are some commercial thermodynamic process simulation programs recommended for absorption processes. The non-ideal liquid phase behaviour are incorporated in the calculations. This software has been extensively used for oil and gas process simulation in the energy industry. It is user-friendly and enables enhancement of conceptual design, performances, and operations. The complex VLE models are incorporated in this program. In addition, it provides access to the world's most widespread property database [51]. Therefore, all the simulations are performed in this simulation program.

Table 4.1 represents the pure component parameters for Aspen HYSYS. The critical properties are for the parameters like temperature (T_c), pressure (P_c) and acentricity (ω) for CO_2, H_2O and TEG. These parameters do not vary much compared to the previous version of Aspen HYSYS.

Table 4.1: *Critical parameters [29]*

Parameters	H_2O	CO_2	TEG
T_c [°C]	374.1	30.95	453.9
P_c [kPa]	22120	7370	1416
ω	0.344	0.2389	0.69

In this report, all the simulations were performed in Aspen HYSYS V8.0 using default Peng-Robinson and TST models (Glycol property package). The glycol property package being principally developed for the simulation of TEG dehydration, it is considered to be most accurate. Therefore, simulation results obtained from TST model are considered in this work. However, for liquid enthalpy calculation, Cavett model was used instead of default property package EOS to avoid unrealistic negative temperatures.

4.1 Base case

A traditional TEG dehydration process with specifications for the base case given in Table 4.2 was simulated in Aspen HYSYS. The flow-sheet model of traditional TEG dehydration process as shown in Figure 4.1 was developed by Mirela [43] in Aspen HYSYS V7.2 using Peng-Robinson and Glycol Package equation of state.

Table 4.2: *Base case specifications for TEG dehydration of CO_2 [43]*

Inlet gas temperature before saturation [ºC]	31.91
Inlet gas temperature after saturation (to absorber) [ºC]	30
Inlet gas pressure [kPa]	3000
Inlet gas flow (to absorber) [kmol/h]	501.1
Water in inlet gas (to absorber) [mol-%]	0.23
Lean TEG temperature [ºC]	35
Lean TEG pressure [kPa]	3000 kPa
Lean TEG rate (after convergence) [kmol/h]	3.583 kmol/h
Water in lean TEG	1.04 mass-%
Number of stages in absorber	10
Murphree efficiency in absorber	0.5
Pressure after the depressurization valve [kPa]	110
Heated rich TEG temperature (for rich/lean heat exchanger minimum temperature difference 10 ^{0}C) [ºC]	153
Number of stages in stripper	4 + condenser + reboiler
Murphree efficiency in stripper	1.0
Reflux ratio in stripper	0.5
Reboiler temperature [ºC]	200
Pressure in stripper [kPa]	101
Lean TEG pump pressure [kPa]	3000
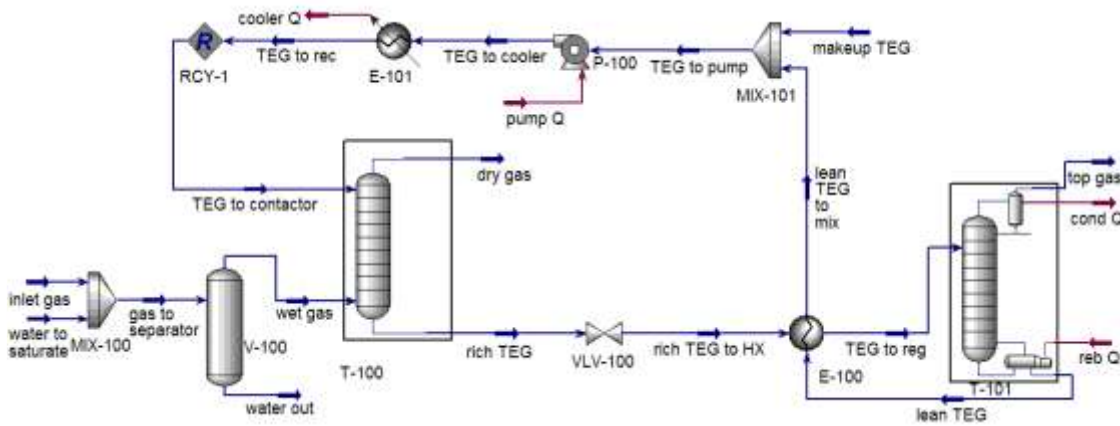
Figure 4.1: *Aspen HYSYS flow-sheet model for traditional TEG dehydration process*

The minimum temperature difference of the rich/lean heat exchanger is adjusted to about 10℃. The feed stream was introduced to stage 2 in the regenerator.

The resulting water content of the dehydrated gas was 153 ppm with Peng-Robinson and 132 ppm with Glycol property package.

4.1.1 Variation of inlet gas pressure

The simulation was performed for the estimation of water removal efficiency as a function of inlet gas pressure. The inlet gas pressure was varied from 3000 kPa to 7000 kPa. The water content in dehydrated gas as a function of absorption pressure is presented in Figure 4.2,
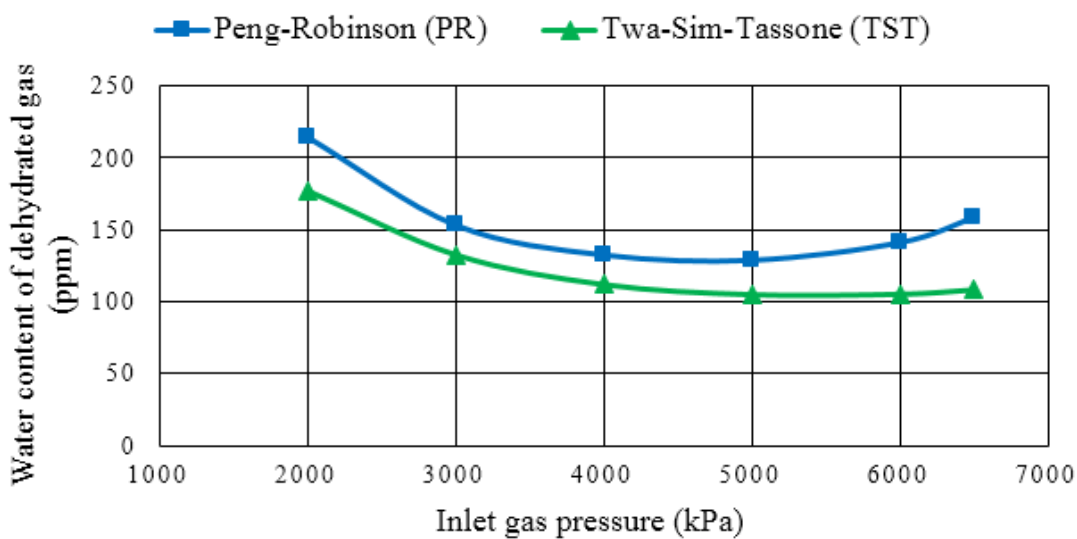
Figure 4.2: *Water content in dehydrated gas as a function of absorption pressure*

From Figure 4.2, it can be observed that higher the inlet gas pressure, more is the increment rate in water removal efficiency. But beyond 5000 kPa, water removal efficiency started to decrease. Above 6500 kPa, there was a convergence problem. Furthermore, the base case was also simulated with 5 and 15 absorption stages which showed the minimum water content of the dehydrated gas at 5000 kPa.

Therefore, from this results, we can conclude that maximum water removal efficiency in both models (PR and TST) was at 5000 kPa.

4.1.2   Variation of lean TEG flow rate

For the first iteration of the model, industrial rule of thumb procedures were applied for the estimation of lean TEG flow rate. The lean TEG flow rate was obtained approximately 500 kg/h [43]. Therefore, the lean TEG flow rate was varied from 300 kg/h to 750 kg/h. The water content in dehydrated gas as a function of lean TEG mass flow rate is presented in Figure 4.3,
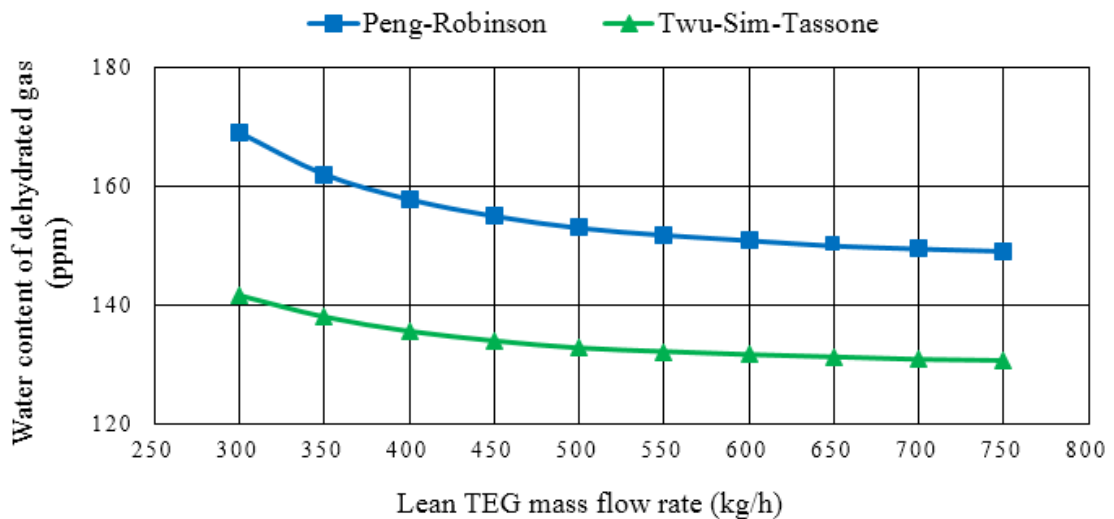

Figure 4.3: *Water content in dehydrated gas as a function of lean TEG flow rate*

From Figure 4.3, it can be seen that water is removed from the gas considerably up to flow rate of 500 kg/h. Beyond this flow rate, there was only slight removal of water from the gas.

4.1.3 Base case simulation including flash tank

In the previous base case model, to flash off some of the CO_2, flash tank[9] was included after the flash valve. The rich TEG was depressurized prior to flash tank equivalent to operating pressure[10] of the regenerator. Flash tank assists in evaporation of absorbed CO_2 in glycol. The simulation was performed using both PR and TST models as shown in Figure 4.4.
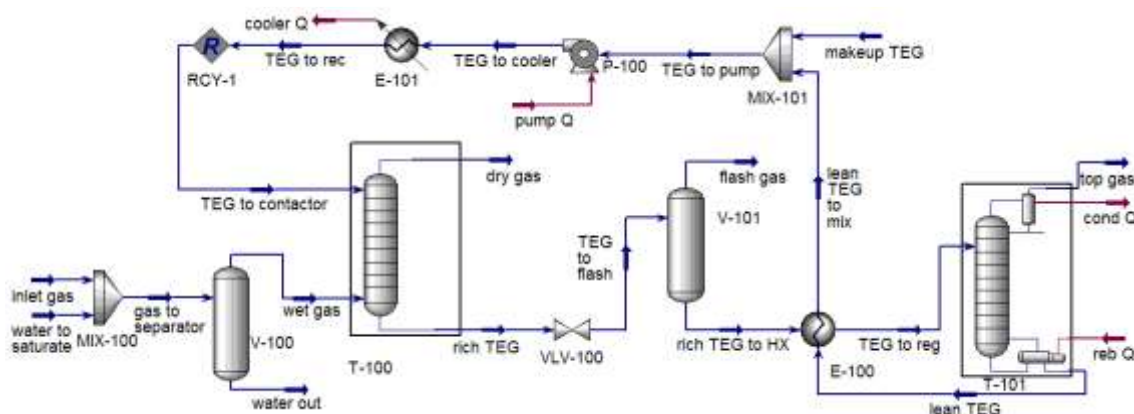
Figure 4.4: *Aspen HYSYS flow-sheet model for traditional TEG dehydration process including flash tank.*

The simulation results obtained using PR and TST models are presented in Table 4.3.

Table 4.3: *Results of TEG dehydration of CO_2 including flash tank.*

	Peng–Robinson	**Glycol package**
Water content in dry gas (ppm)	153.1	131.8
Flash gas molar flow (kmol/h)	0.8868	0.6251
Flash gas composition	99.14 mol-% CO_2 0.86 mol-% H_2O	99.37 mol-% CO_2 0.63 mol-% H_2O

There was not much deviation in the results including and excluding flash tank. The discrepancy in the water content in comparison to the previous results from both models was less than 1 ppm.

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[9] The flash tanks are generally introduced in the gas dehydration unit to prevent hydrocarbons from entering the regenerator.

[10] Typical operating pressure should not be lowered too much to avoid evaporation of glycol.

4.2 TEG regeneration enhancement

4.2.1 Stripping gas to the reboiler

To increase the regeneration of TEG purity, stripping gas was injected into the regeneration column at the reboiler. The simulation model of dehydration process in presented in Figure 4.5. The specifications of the stripping gas are specified below [43]:

Temperature: 190ºC

Pressure: 101kPa

Flow rate: 0.2 kmol/h

Composition: CO_2 = 99.17 mol-%

H_2O = 0.83 mol-%

Figure 4.5: *Aspen HYSYS flow-sheet model with stripping gas introduced to the reboiler*

Simulations were performed with 5, 10, 15 and 20 stages in the absorption column. Stripping gas flow rate was varied from 0.2 kmol/h to 2 kmol/h.
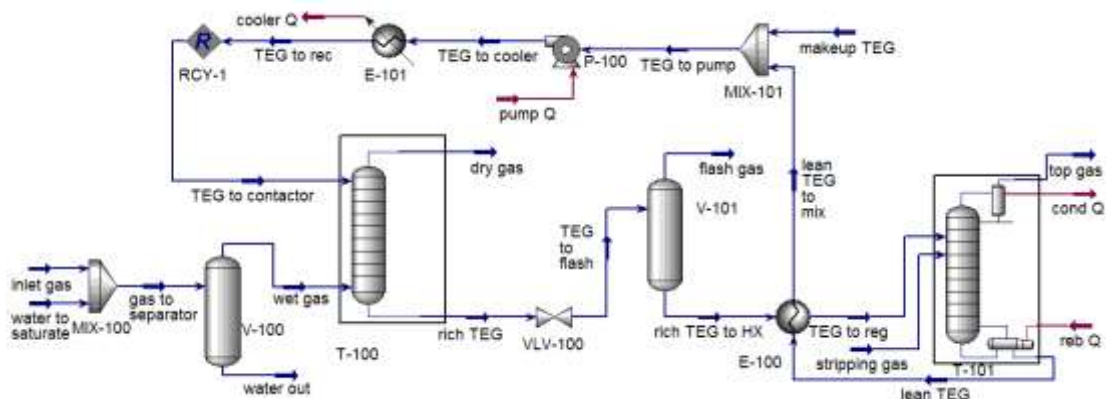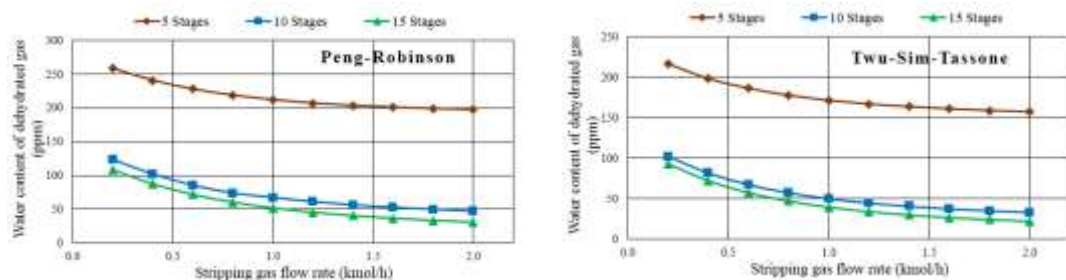

Figure 4.6: *Water content as a function of stripping gas and number of absorber stages with stripping gas to the reboiler using PR and TST models*

From Figure 4.6, it can be observed that water removal efficiency increases with the increase in the stripping gas flow rate. Even a small stripping gas flow rates have pronounced difference. There was a considerable reduction in the water content at small flow rate than high flow rate. It makes clear that low water content cannot be obtained just by increasing the stripping gas flow rate.
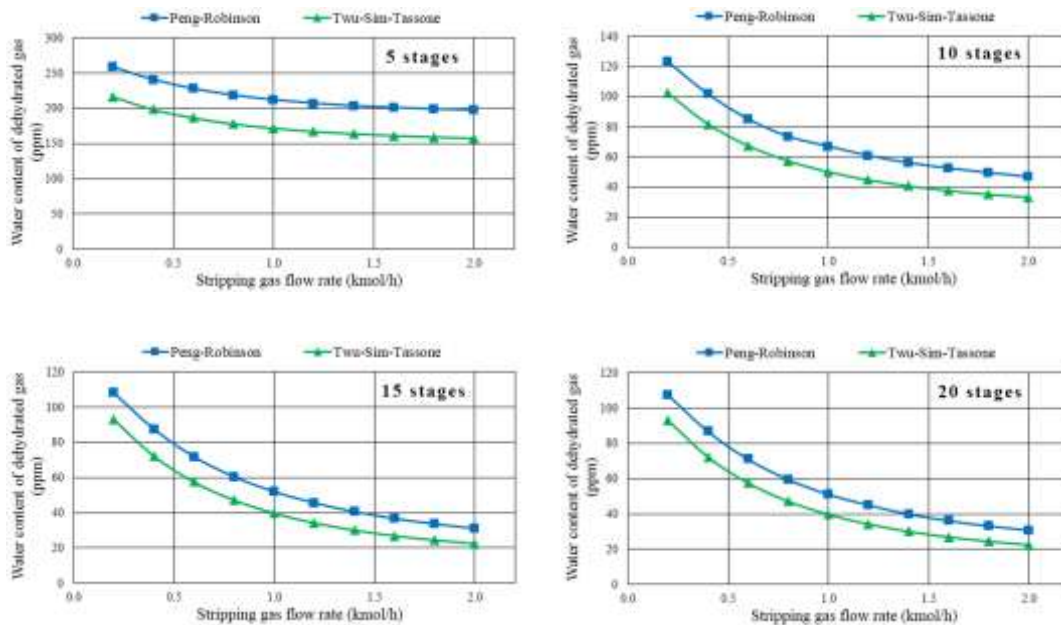
Figure 4.7: *Water content as a function of stripping gas and number of absorber stages(5,10,15 and 20) with stripping gas to the reboiler using PR and TST models*

From Figure 4.7, it can be illustrated that as the number of stages was increased, better water removal results were obtained. There was significant removal of water when number of stages was increased from 5 to 10. Furthermore, results at 15 stages were better than 10 stages while results from 20 stages were almost the same. This clears that use of 15 stages in the absorber will be sufficient to achieve low water content.

Also, for all stages, the stripping gas flow rate above 1.2 kmol/h did not show any significant removal of water in the dehydrated gas.

Therefore, it can be concluded that no significant changes can be achieved with continuous increase of stripping gas flow rate or absorber stages.

4.2.2   Flash gas as stripping gas to the reboiler

The simulation was performed by injecting flash gas to the regeneration column at the reboiler. The gas stream was depressurized to the operating pressure of regenerator by the flash valve.
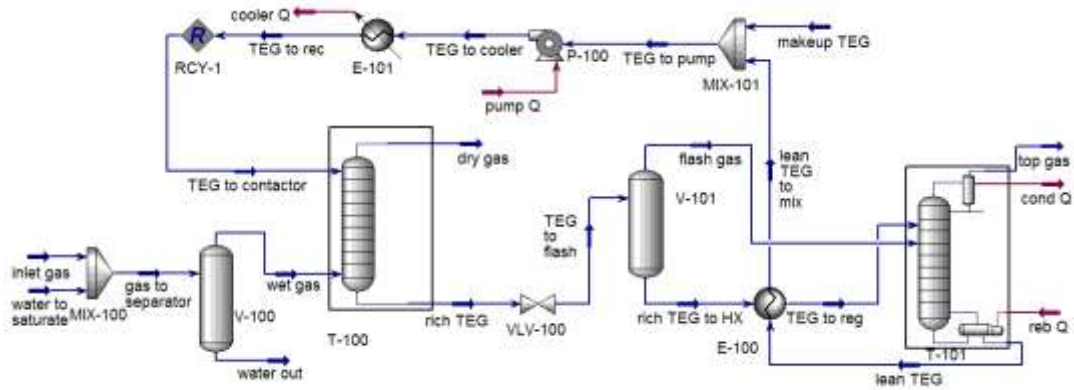
Figure 4.8: *Aspen HYSYS flow-sheet model with flash gas as stripping gas to the reboiler*

The simulation results obtained using PR and TST models are presented in Table 4.4.

Table 4.4: *Simulation results of flash gas as a stripping gas to the reboiler*

Number of stages in absorber	Water content (ppm)	
	Peng-Robinson	**Glycol Package**
5	211.9	186.0
10	74.12	67.50
15	60.30	57.47
20	60.30	57.47

From Table 4.4, it can be seen that there is less difference in the water content result compared to the previous result using CO_2 as stripping gas. This is due to the high amount of flash gas produced from PR model (0.89 kmol/h) than TST model (0.63 kmol/h).

Since the flash gas recovered from the flash tank is pure CO_2 (almost the same specifications as stripping gas introduced externally), the water content in the dehydrated gas by using flash gas and CO_2 separately as stripping gas were similar.

4.2.3   Regeneration and extra stripping column simulated as one column

This simulation process was performed in with two different techniques. One is to simulate the process with two different columns (desorption and extra stripping column) and the other is with one column (combining desorption and extra stripping column). In this work, second technique i.e. desorption column and extra stripping column calculated as one column was chosen. The second technique was preferred for the simplification of the flow-sheet model and also for the elimination of simulation convergence problems. Moreover, similar numerical results were obtained from these both methods.

This column was simulated with 7 (1+3+3) stages. The feed stream was injected to stage 2 and the reboiler heat addition to stage 4. There were 3 stages between the reboiler heat addition and the stripping gas feed at the bottom stage of the column. The efficiency of each stage was 1.

The given model was simulated in three different ways:

1. Stripping gas to the regenerator

The Aspen HYSYS model for this process has been presented in Figure 4.9. The specified stripping gas as mentioned in section 4.2.1 was introduced to the regenerator.
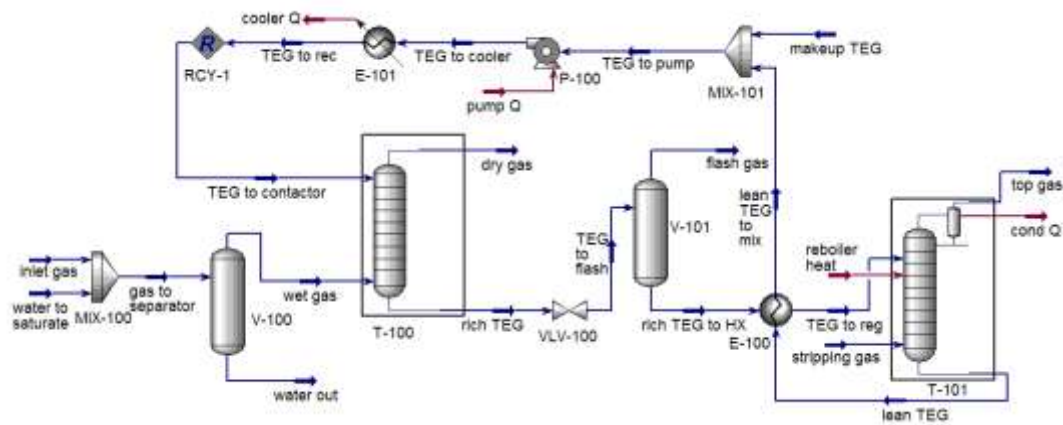
Figure 4.9: *Aspen HYSYS flow-sheet model of desorption column and extra stripping column simulated as one column*

For the simulation process, stripping gas flow rate was varied from 0.2 kmol/h to 1.4 kmol/h. Also, number of stages were varied for both models. The results from these calculations as a function of stripping gas and number of absorber stages are presented in Figure 4.10.
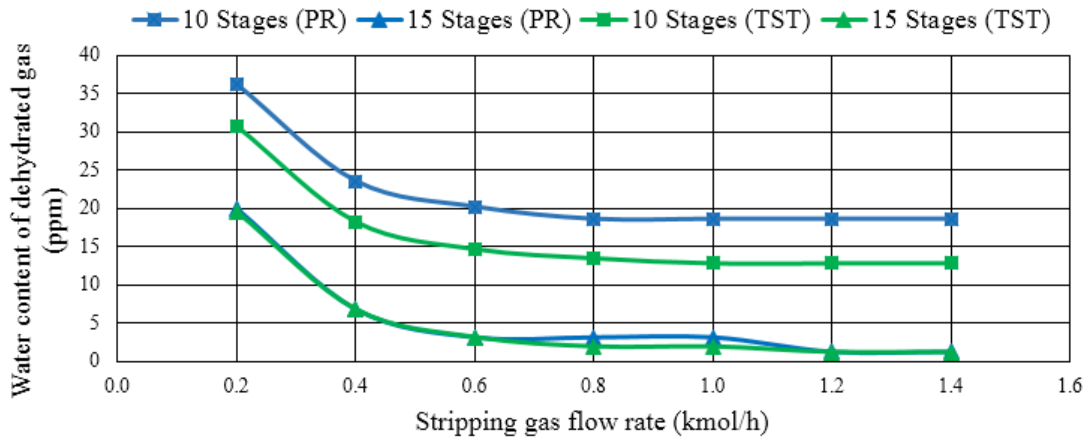
Figure 4.10: *Water content as a function of stripping gas and number of stages with desorption column and stripping column simulated as one using Peng-Robinson and Twu-Sim-Tassone*

Figure 4.10 illustrates that there is considerable removal of water vapour up to 0.6 kmol/h stripping gas flow rate until it reached a constant value. It can be attributed to the fact that equal amount of available flash gas can be enough to achieve lower than 5 ppm.

Figure 4.11: *Water content as a function of stripping gas and number of absorber stages (5,10,15 and 20) with extra stripping column using PR and TST models*

From Figure 4.11, it can also be noted that increasing flow rate beyond 0.6 kmol/h does not improve the water removal efficiency significantly. Moreover, increasing the number of absorption stages more than 15 does not influence the dehydration process.

2. Portion of dehydrated pure CO_2 from the process as a stripping gas

The small portion of CO_2 dry gas obtained from the absorption column was introduced to the regenerator. Stream splitter was used to separate the required amount followed by the depressurization valve to maintain the operating pressure. Heater was used to heat the dry gas before being injected to the regenerator.

The Aspen HYSYS model for this process is shown in Figure 4.12.

Figure 4.12: *Aspen HYSYS flow-sheet model of regeneration column and extra stripping column simulated as one with portion of dehydrated CO_2 as stripping gas*

The simulation results obtained using PR and TST models are presented in Figure 4.13.
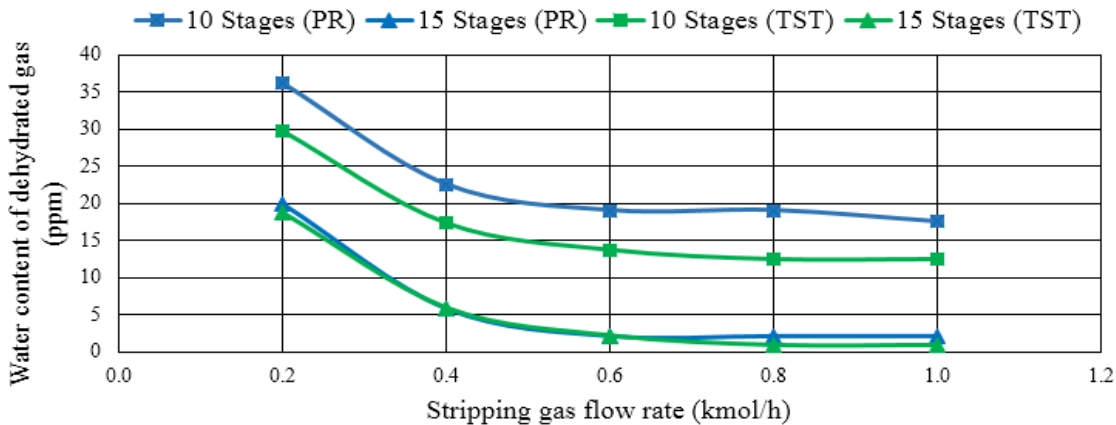
Figure 4.13: *Water content as a function dehydrated CO_2 as stripping gas with regeneration column and stripping column simulated as one using PR and TST*
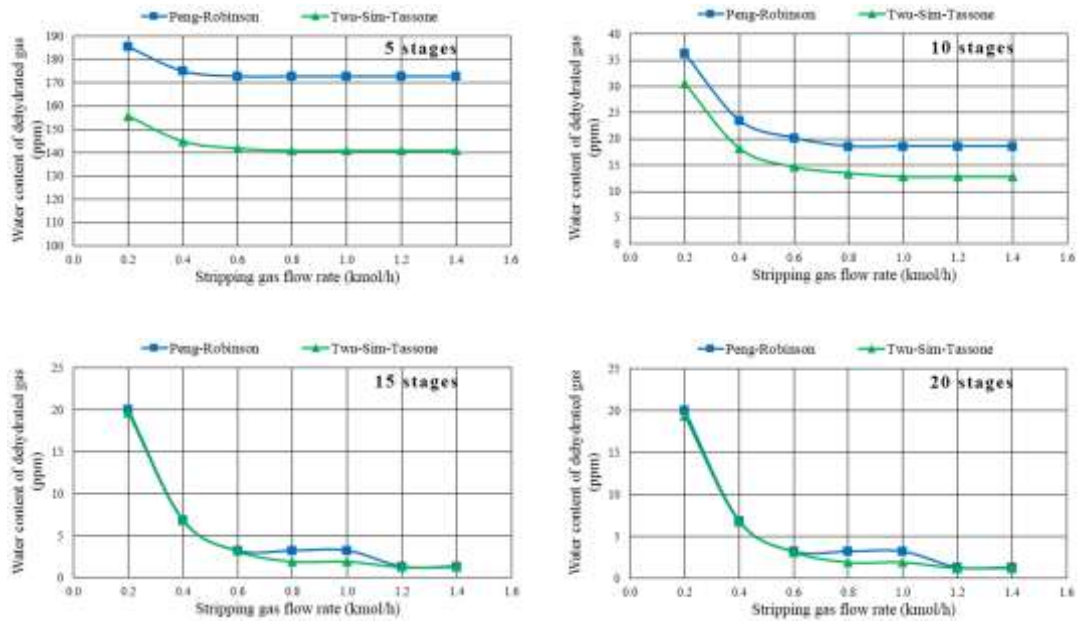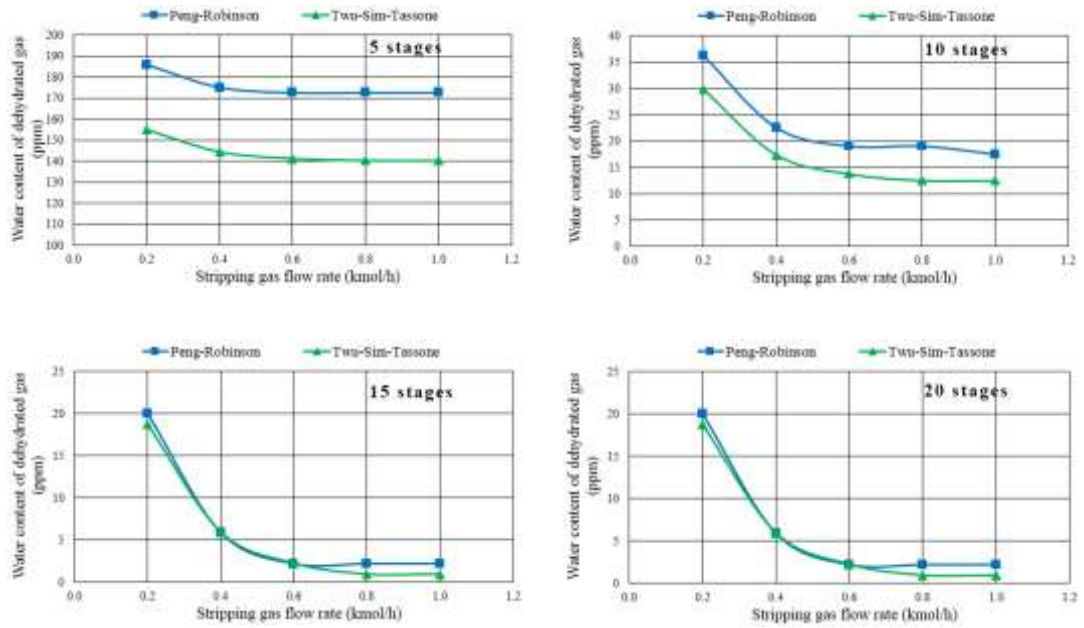
Figure 4.14: *Water content as a function of stripping gas (dehydrated CO_2) and number of absorber stages (5,10,15 and 20) with extra stripping column using PR and TST*

The results obtained from this simulation is similar to the previous simulation. However, in this case, the water content in the dehydrated gas is slightly lower because the stripping gas composition is 100 % carbon dioxide whereas in previous case specifications, small amount of water was also included.

3. Flash gas used as stripping gas to an extra stripping column

The flash gas from the flash tank was introduced to the regenerator as a stripping gas. This process was carried out for 7 stages in the regenerator. The feed stream was introduced to stage 2 and the additional reboiler heat to stage 4. Four different number of stages was simulated in the given Aspen HYSYS model shown in Figure 4.15.
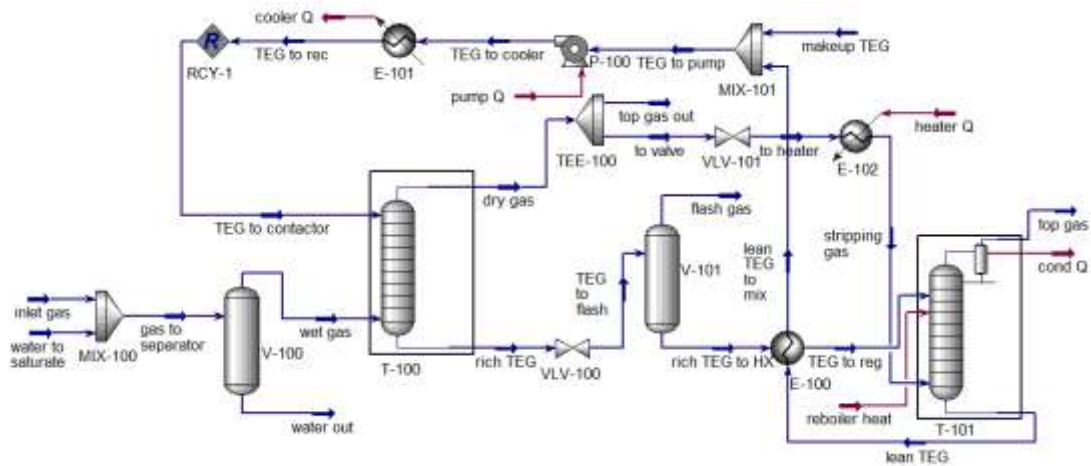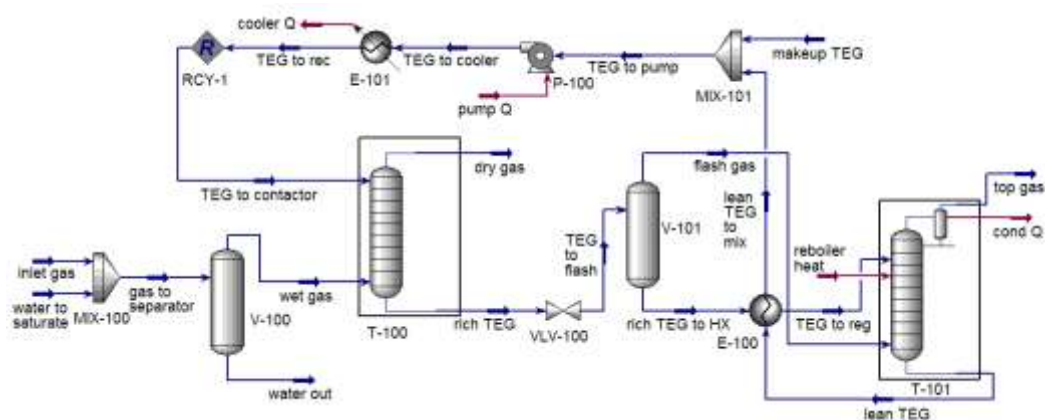

Figure 4.15: *Aspen HYSYS flow-sheet model of regeneration column and extra stripping column simulated as one with flash gas as stripping gas*

The simulation results obtained using PR and TST models are presented in Table 4.5.

Table 4.5: *Simulation results of flash gas as a stripping gas to extra stripping column simulated as one with regeneration column simulated as one*

Number of stages in absorber	Water content of dehydrated gas (ppm)	
	Peng-Robinson	**Glycol Package**
5	170.6	141.2
10	18.6	13.7
15	1.8	2.2
20	1.8	2.1

From Table 4.5, it is clear that it is possible to improve the water removal percentage just by increasing the number of stages in the absorption column.

4.3 Simulation of Drizo process

The Drizo process was simulated in Aspen HYSYS with n-heptane/benzene/toluene as a stripping gas. The flow-sheet model of Drizo process is presented in Figure 4.16.
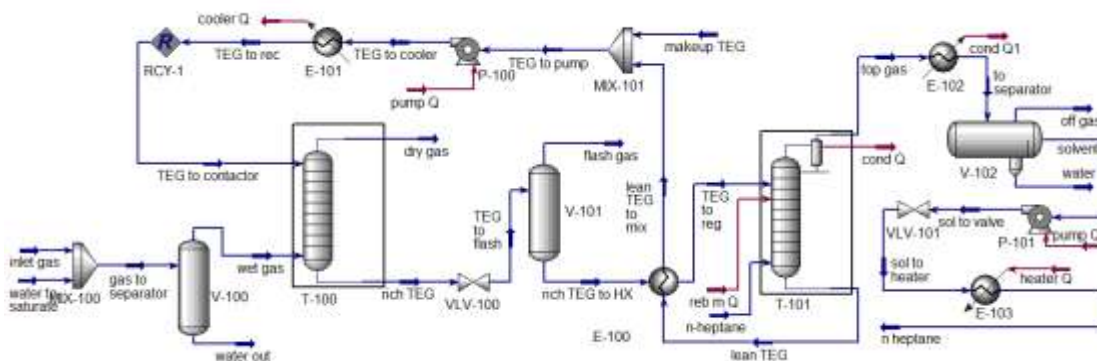
Figure 4.16: Aspen HYSYS flow-sheet model of Drizo process using n-heptane as stripping gas

All the solvents showed almost the same amount of dehydration, however, there was differences in added stripping gas amount and recirculated stripping gas amount. The deviation was maximum with toluene of about 0.15, with benzene of about 0.11 and least with n-heptane of about 0.01.
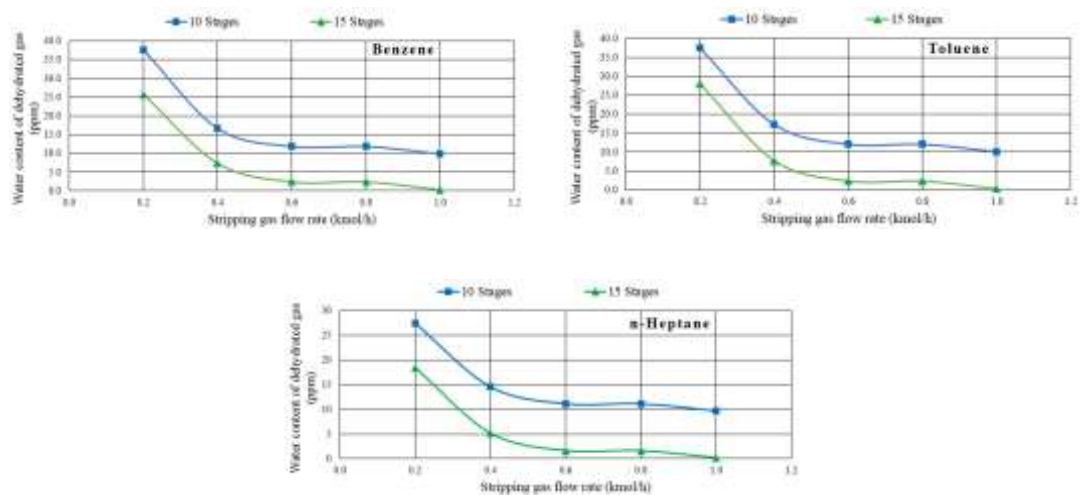
Figure 4.17: *Water content in Drizo process with benzene, toluene and n-heptane as a function of stripping gas and number of absorber stages using TST model*

From Figure 4.17, it can be observed that with all the solvents it can be possible to reduce the water content of the dehydrated gas down to 10 ppm using 10 absorption stages. Moreover, with the available amount of flash gas, it is possible to achieve less than 15 ppm and 5 ppm with 10 and 15 stages respectively.

5 Discussion

5.1 Verification of previous simulation results and deviations

In this chapter, simulation results are compared with Mirela [43] and Sujan [44].

In base case simulation, the water content of the dehydrated gas was same using PR equilibrium models while, with TST model the result is different with Mirela but similar with Sujan's result. The water content results from TST model are lower as well as less deviated from PR result compared to Mirela.

When the stripping gas was introduced to the reboiler, Mirela achieved less than 50 ppm water content with 20 absorption stages and 1.2 kmol/h stripping gas flow rate whereas, Sujan required 20 absorption stages[11] and 0.8 kmol/h stripping gas which is similar to this work. In this work the lowest water content obtained by this process was 30 ppm which is same as Mirela and much lesser value of 20 ppm was achieved by Sujan.

In this work when the stripping gas was introduced to the extra stripping column, 15 absorption stages and 0.6 kmol/h stripping gas was required to achieve less than 5 ppm, Sujan needed 15 absorption stages and 1 kmol/h stripping gas and Mirela needed 20 stages and 1.2 kmol/h stripping gas. In all work, it was possible to obtain water content down to 2 ppm.

When using dehydrated CO_2 as stripping gas obtained from the absorption column to the extra stripping column, the water content was lowered below 2 ppm with 20 absorption stages. However, in Mirela and Sujan's work more stripping gas (1.8 kmol/h) was required compared to this work (0.8 kmol/h).

The amount of flash gas obtained from the flash tank was same 0.89 using PR model whereas, with TST model the amount was 0.63 which is slightly higher compared to Mirela's 0.4 respectively.

Although there were deviations in the results, at the final stage, the amount of water content in the dehydrated gas was almost similar with each other's work. Thus, it can be concluded that all the results obtained from this work are acceptable.

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[11] In this work, all simulations with 15 and 20 absorption stages gave the same results.

5.2 Comparison of Peng-Robinson and Twu-Sim-Tassone

TEG dehydration of carbon dioxide gas was performed in Aspen HYSYS V8.0 using Peng-Robinson and Twu-Sim-Tassone equilibrium models.

For the base case specifications, 153 ppm and 133 ppm water content were achieved with PR and TST models respectively. The results for TST model were different from Mirela which might be due to the updated version of simulation program[12]. However, in this work, the difference in water content results between the two models was lesser compared to the work of Mirela. Since the trend of the curve was similar, it can be concluded that the obtained results are correct.

5.3 Optimum dehydration pressure

Both Peng-Robinson and Twu-Sim-Tassone models gave the minimum water content at 5000 kPa. The minimum water content in dehydrated gas was 129 ppm using PR model and 105 ppm using TST model at optimum dehydration pressure.

But there was a deviation in absorption pressure, as in case of Mirela and Sujan, maximum water dehydration was at 3000 kPa with TST model.

5.4 Evaluation of simulations

There was a significant reduction in water content until the lean TEG mass flow rate of 500 kg/h but increasing beyond that value had little impact on dehydration efficiency.

When the stripping gas was introduced to the reboiler, less than 50 ppm was achieved with 10 absorption stages and 1.8 kmol/h stripping gas flow rate using PR equilibrium model. Likewise, using TST equilibrium model, 10 absorption stages and 1 kmol/h stripping gas flow rate was required. Similarly, using 15 stages in both models, 1 kmol/h for PR model and 0.8 kmol/h for TST model was necessary to achieve less than 50 ppm in the dehydrated gas stream. Furthermore, it was possible to dehydrate gas down to 30 ppm by increasing the amount of stripping gas using both models. However, there was

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[12] Mirela performed the simulation in Aspen HYSYS V7.2 while, in this work Aspen HYSYS V8.0 was used.

not much effect on the results above 15 absorption stages and 1.6 kmol/h stripping gas flow rate.

With available flash gas recovered from the flash tank as stripping gas, less than 100 ppm can be achieved by the both equilibrium models.

When the stripping gas was introduced to the extra stripping column, significant reduction in the water content of dehydrated gas was achieved. With 10 absorption stages, water content was reduced down to 20 ppm using PR model and down to 15 ppm using TST model. Addition of stripping gas above 1.2 kmol/h did not affect the dehydration efficiency. With the available flash gas amount, less than 5 ppm was possible to achieve using 15 absorption stages for both models. Moreover, water content was reduced down to about 2 ppm by increasing the stripping gas flow rate. In this case also increasing absorption stages more than 15 did not have much impact on water removal process.

Flash gas was also introduced as stripping gas to extra stripping column, which gave similar results. Less than 20 ppm with 10 absorption stages and less than 5 ppm with 15 absorption stages were achieved.

When dehydrated pure CO_2 from the absorption column was used as stripping gas, water content was reduced a little bit more compared to introducing CO_2 externally. As in this case the CO_2 was pure but in previous case, some amount of water existed. Water content below 1 ppm was achieved by using 15 absorption stages and 0.8 kmol/h stripping gas flow rate.

In Drizo process, by using toluene, benzene and n-heptane as stripping gas equivalent to flash gas amount, water content was reduced down to 15 ppm with 10 stages and down to 5 ppm with 15 stages in the absorption column. On further increasing the stripping gas to 1 kmol/h, less than 1 ppm was obtained.

Aspen HYSYS showed the deviation of 0.04% when the TEG flow rate or stripping gas flow rate was increased gradually (in small amount) and rapidly (in large amount). Results obtained from increment by 0.2 kmol/h stripping gas slightly varies with an increment by 1 kmol/h.

The conference paper, "Simulation of glycol processes for CO_2 dehydration" by Lars Erik Øi and Birendra Rai has been submitted to EUROSIM 2016 – The 9th Eurosim Congress on Modelling and Simulation in Oulu, Finland. It was accepted in 24.04.2016. The paper is presented in Appendix 2.

5.5 Choice of process dependent on dehydration requirement

The water specification of the dehydrated gas is the one of the key governing factors for the selection of dehydration processes.

It would be rationale to recommend a traditional TEG dehydration process for the requirement of water content in the dehydrated gas is near about 150 ppm.

The traditional TEG dehydration process can be acceptable by enhancing the process by injecting stripping gas to the reboiler to achieve 50 ppm water content in the dehydrated gas. Flash tank can be introduced in the process so that flash gas can be used as stripping gas which will reduce the cost. Increasing the number of stages in the absorption column will raise the cost significantly.

Drizo process is generally preferred when the water content requirement is less than 1 ppm, addition of an extra stripping column can also be acceptable to achieve the same water content limitation. For Drizo process more additional equipment will be required which makes the process costly. Also, the cost for extra height of an extra stripping column increases but it will be relatively less due to its small diameter.

6 Conclusion

The traditional and enhanced glycol dehydration processes can be simulated in Aspen HYSYS simulation program using both Peng Robinson (PR) and Twu-Sim-Tassone (TST) equilibrium models with the convincible outputs.

Both PR and TST models did not show any convergence problems. The simulation results and the observed similar trends from both models conclude these models to be well predicting models.

The optimum dehydration pressure is 5000 kPa for both PR and TST equilibrium models.

The deviations in the flash gas amount from PR and TST models is due to the calculation differences of CO_2 solubility in TEG/water solutions with those two models.

In traditional dehydration process, the dehydration efficiency can be enhanced either by increasing the number of stages in the absorption column or by increasing the TEG circulation rate. Using this process water concentration can be reduced below 150 ppm in the dehydrated CO_2.

In enhanced dehydration process, in addition to above strategies, stripping gas can be fed to the regenerator to increase triethylene glycol regeneration which eventually improves dehydration efficiency. Flash gas from the flash tank or dehydrated pure CO_2 from the absorption column can be used a source of stripping gas.

Injection stripping gas to the reboiler have a significant influence in the water removal percentage. The stripping gas equivalent to the flash gas amount can dehydrate gas about twice in the same process without stripping gas. Water content below 50 ppm can be obtained by this process.

Introducing stripping gas to the extra stripping column has a further impact on dehydration efficiency. It is possible to achieve water content less than 1 ppm in the dehydrated CO_2.

Although toluene, benzene, and n-heptane showed the same dehydration capability, the deviation between the added stripping gas amount and recirculated stripping gas amount occurred maximum in toluene and then in benzene and least in n-heptane. This method was able to achieve water content less than 1 ppm.

In all simulations performed in this work, there was no significant increase in dehydration efficiency when the number of absorption stages was increased above 15 stages.

Because of the lack of literature data on the CO_2 dehydration process, conclusions drawn about this process can be taken as a reference for further researches with the purpose of emphasizing the possibilities of TEG dehydration process as a cheap and efficient alternative.

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Appendices

Appendix 1: Task description

Telemark University College

Faculty of Technology

FMH606 Master's Thesis

Title: CO_2 dehydration after CO_2 capture

TUC supervisor: Associate Professor Lars Erik Øi

Task Description:

The aim is to develop simulation models of CO_2 dehydration after CO_2 capture.

1. Literature search on dehydration methods based on glycol absorption for captured CO_2. The search should include processes from commercial suppliers of dehydration plants.
2. Process description of a dehydration process for compressed CO_2 based on absorption in triethylene glycol (TEG). The dehydration can be performed after or between CO_2 compression stages.
3. Process simulation of a dehydration process using e.g. Aspen HYSYS and possibly also other process simulation programs like Aspen Plus and ProMax.
4. Optimization of the dehydration process for different conditions.

Task background:

Several Master Projects have been performed at Telemark University College about calculation and simulation of different CO_2 capture processes. Telemark University College has collaborated with different companies (Tel-Tek, Statoil, Aker Kværner, Norcem, Skagerak and Gassnova) which work with plans for CO_2 capture. There have also been performed several Master projects at TUC on dehydration of natural gas, and also Master projects at TUC on dehydration of captured CO_2 in 2014 and 2015. A paper based on this work was presented at a conference autumn 2014.

Student category: PT or EET

Practical arrangements: The work will mainly be carried out at Telemark University College.

Signatures:

Supervisor (date and signature): *Lars Erik Øi* 4/2 – 2016

Student (date and signature): *Bjørn* 04- Feb - 2016

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Appendix 2: A conference paper "Simulation of glycol processes for CO₂ dehydration" by Lars Erik Øi and Birendra Rai

Simulation of Glycol Processes for CO₂ Dehydration

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Abstract—Water must be removed from CO_2 prior to transport or storage to avoid corrosion and hydrate formation. Absorption into triethylene glycol (TEG) followed by desorption is the traditional gas dehydration method, and is expected to be the preferred method for large scale CO_2 dehydration. There is no agreement on the level of accepted water content after dehydration, and the specifications vary normally in the range between 5 and 500 ppm (parts per million by volume). In literature, it is claimed that use of solid adsorbents is necessary to reduce the water content to below 30 or 10 ppm. In this work, simulations in Aspen HYSYS demonstrate that it is possible to obtain below 1 ppm water using a traditional glycol dehydration process including an extra stripping column. The models Peng-Robinson (PR) and Twu-Sim-Tassone (TST) with updated parameters in Aspen HYSYS version 8.0 are used. A Drizo process using a stripping gas which is later condensed and recirculated is also simulated, and this process also achieves a water content below 1 ppm.

Keywords— CO_2; glycol; dehydration; Aspen HYSYS

I. INTRODUCTION

CO_2 removed from natural gas or from CO_2 capture should be dehydrated prior to transport or storage. Water may lead to problems like corrosion and hydrate formation. The need for water removal from CO_2 and possible specifications are discussed in several references [1,2,3] and water specifications are normally in the range between 5 and 500 ppm (parts per million by volume). CO_2 for enhanced oil recovery normally requires the lowest water content.

There are several different gas dehydration methods available. The most used processes for dehydration are based on absorption, adsorption or membranes. The most traditional method for large scale dehydration to moderate water levels is by absorption into triethylene glycol (TEG). For very low water levels, adsorption processes are claimed to be necessary [4,5]. There are however simulated reasonable processes for glycol dehydration of CO_2 down to water levels below 5 ppm [6] using

stripping gas and an extra stripping column. The Drizo process makes use of a condensable stripping gas which is recirculated and is able to reduce the water content down to 1 ppm [7].

A recent study [5] have evaluated different commercial processes for CO_2 dehydration based partially on information from vendors of technology. Processes based on glycol and on solid adsorption were evaluated. It was claimed that use of solid adsorbents is necessary to reduce the water content to below 30 or 10 ppm.

The main purpose of this paper is to present updated simulations of flow-sheet models for CO_2 dehydration by absorption in triethylene glycol. The Peng-Robinson (PR) model and the Twu-Sim-Tassone (TST) model (the glycol package in Aspen HYSYS) are used. In version 8.0, the parameters in the TST model have been updated compared to version 7.2 which was used in earlier work with Aspen HYSYS [6]. Especially, it is an aim to simulate the alternative including an extra stripping column and a Drizo process achieving a water level down to 1 ppm. The simulation results in this work are mainly from the Master Thesis work of Birendra Rai [8].

II. SIMULATION PROGRAMS AND MODELS FOR GLYCOL DEHYDRATION

Commercial process simulation programs which have been used for glycol dehydration are Aspen Plus, Aspen HYSYS, Pro/II and ProMax. Process simulation programs are useful for simulation of absorption processes because complex vapour/liquid equilibrium models are available in the programs and because efficient stage to stage models for absorption and desorption columns are available.

Absorption and distillation columns are traditionally simulated as a sequence of equilibrium stages. The stages can also be specified with a Murphree efficiency. A Murphree efficiency can be defined as the change in mole fraction of a component (in this case water) from the stage below to a given stage, divided by the change if equilibrium was achieved on the given stage [4].

Vapour/liquid equilibrium data in the TEG/water system has been discussed in several papers [4,6,9]. The vapour/liquid equilibrium between CO_2 and water has also been evaluated [1,10]. This equilibrium must be included in a complete model of the TEG/water/CO_2 system. One special challenge for such a model, is to calculate the correct solubility of CO_2 in a TEG/water solution.

In Aspen HYSYS, the equilibrium models PR [11] and TST [12] are available for glycol dehydration. The TST model is claimed in the Aspen HYSYS program documentation to be the most accurate. This is however based on the assumption of dehydration of natural gas, and it is uncertain whether the TST model is accurate when CO_2 is the dominating gas. The TST model parameters have been updated from earlier versions in the Aspen HYSYS version 8.0 used in this work. The PR model has only one adjustable parameter for each binary component pair while TST has 5 adjustable parameters for each binary pair. In Aspen Plus, PRO/II and ProMax there are also models available recommended for the TEG/water system [6].

Simulations of the natural gas dehydration process in Aspen HYSYS have been performed with emphasis on glycol regeneration by Øi and Selstø [13]. A traditional TEG dehydration process was simulated using the PR equation of state. A water content of 58 ppm was specified in the dehydrated gas. Different regeneration processes were simulated, e.g. addition of stripping gas to the reboiler and adding stripping gas to an extra stripping column. Bilsbak [14] used the simulation program Pro/II to simulate TEG dehydration of CO_2 using an equation of state.

Besides the references mentioned in this work, there is very little information found in the open literature about simulation of CO_2 dehydration processes and particularly of CO_2 dehydration based on absorption in glycol.

III. PROCESS DESCRIPTION

A traditional glycol dehydration process is shown in Fig. 1. Water is absorbed from a gas in a glycol solution in an absorption column (contactor). The liquid (rich glycol) is then depressurized to a flash tank to evaporate some of the absorbed CO_2. Then the liquid is heated by regenerated glycol in a heat exchanger and sent to a desorber (regenerator). In this column, water is removed from the top and regenerated TEG is removed from the bottom. Heat is added in the reboiler. The regenerated TEG (lean glycol) is pumped through the heat exchanger and a cooler back to the absorber. More detailed process descriptions can be found in e.g. [4,6].

It is possible to reduce the water content in the glycol by adding stripping gas to the reboiler. The reduction in water content in the glycol will make it possible to improve the dehydration of the CO_2 in the treated gas. The stripping gas can come from the flash gas or from the dehydrated gas.

The water content in the glycol can be reduced further by adding an extra stripping column below the reboiler. This is shown in Fig. 2.
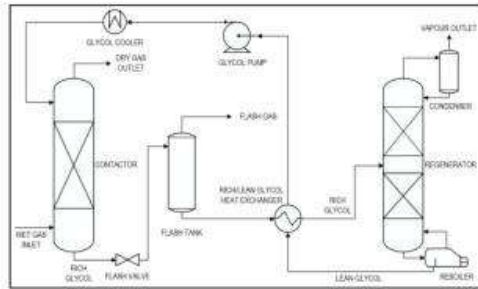

Fig. 1. A traditional TEG dehydration process

There are also different special glycol dehydration processes. One example is the Drizo process [7] where the added stripping gas is a condensable component which is condensed after the stripping column and recirculated to the bottom of the stripping column.

In earlier work [6] the possible water content achievable from these processes was calculated using Aspen HYSYS. A traditional process could achieve below 200 ppm. Using stripping gas, less than 50 ppm could be achieved. Adding an extra stripping column below the reboiler could reduce the water content to below 5 ppm.

To achieve very low water content, the height of the absorption column have to be increased compared to a traditional process. The absorption column will contain plates or packing. An expected plate efficiency is order of magnitude 50 %, and an expected packing efficiency of a packing height of 0.5 meter is also order of magnitude 50 % [15]. An increase of the packing height from 10 to 20 stages or order of magnitude from 5 to 10 meters will increase the cost significantly.

The extra stripping column will add complexity to the process. The cost for extra height in this column is however small because the diameter is very low.
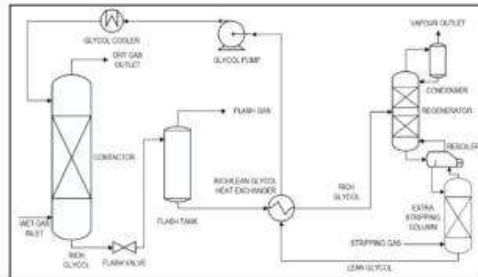

Fig. 2. A TEG dehydration process with extra stripping column

IV. PROCESS SIMULATION, RESULTS AND DISCUSSION

A. Simulation of standard process

A traditional TEG dehydration process as in Fig. 1 has been simulated in the simulation program Aspen HYSYS version 8.0 using the PR equation of state and TST model (the glycol property package). The Aspen HYSYS flow-sheet model for the base case simulation is presented in Fig. 4.

The specifications for the base case process calculation are given in Table 1. These are similar to the specifications in earlier simulations [6]. The enthalpy setting in Aspen HYSYS was changed to the Cavett model instead of the default property package EOS (equation of state) to avoid unrealistic low temperatures. Recommendations for a traditional process can be found also in [4]. The absorption column was simulated with 10 stages and with Murphree efficiency 0.5, which is assumed to be equivalent to approximately 10 actual plates or 5 meter of structured packing [15].

TABLE I.   BASE CASE SPECIFICATIONS FOR SIMULATION (FIGURE 4)

|	
Temperature, inlet gas [^{0}C]	30
Pressure, inlet gas [kPa]	3000
Gas flow, inlet gas [kmol/h]	501.1
Water in inlet gas [mol-%]	0.23
Temperature, TEG to contactor [^{0}C]	35
Pressure, TEG to contactor [kPa]	3000
Flow, TEG to contactor [kmol/h] (in first iteration)	3.583
Water in lean TEG [mass-%] (in first iteration)	1.04
Number of stages in absorber	10
Murphree efficiency for each stage in absorber	0.5
Pressure after the depressurization valve [kPa]	110
Temperature, TEG to regeneration [^{0}C]	153
Number of desorber stages (+ condenser/reboiler)	4
Murphree efficiency for each stage in desorber	1.0
Reflux ratio in desorber	0.5
Reboiler temperature [^{0}C]	200
Pressure in desorber [kPa]	101
Pressure, after TEG pump [kPa]	3000
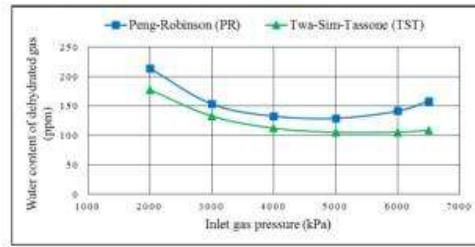
Fig. 3.   Water content in dehydrated gas as a function of absorption pressure

The calculation sequence of the process in the Aspen HYSYS flow-sheet is mostly following the real flow direction. The gas feed stream to the absorber is saturated with water. The liquid feed to the absorber has to be estimated in the first iteration. Then the absorption column and the rest of the process is calculated step by step. The cold side of the main heat exchanger is calculated based on a specified temperature on the stream to the desorber.

Using the base case specifications in Table 1, the water content in dehydrated gas was calculated to 153 ppm with the PR model and 133 ppm with the glycol package using the TST model. Compared to earlier simulations, the results from the PR model were identical. The results using the TST model were lower in water content compared to the earlier results [6]. The differences are expected to be due to updated TST parameters. The deviations between calculated water content from PR and TST are lower in this work.

The process was simulated also at other absorption pressures. The results are shown in Fig. 3. Both models gave a minimum water content and maximum dehydration efficiency at 5000 kPa. Minimum water content was 129 ppm at about 5000 kPa using the PR model and 105 ppm using the TST model. In earlier work [6] the TST model gave a minimum water content at 3000 kPa.
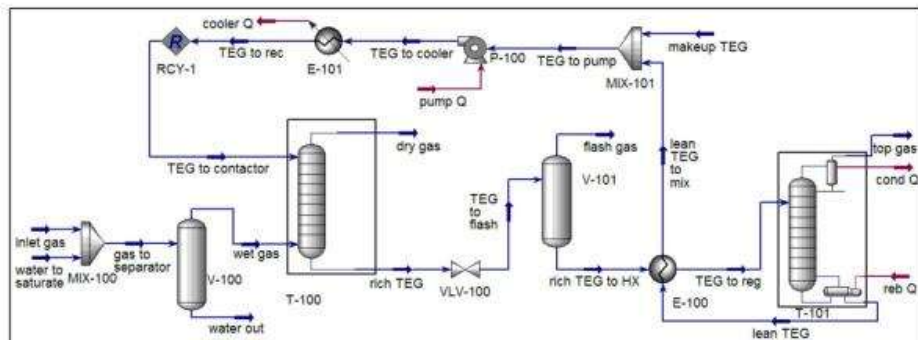

Fig. 4.   Aspen HYSYS flow-sheet model for traditional TEG dehydration process

The dehydration can be improved by increasing the TEG circulating rate and by increasing the number of absorption stages in the standard process. However, these changes gave only minor improvements in achieved water content in the dehydrated gas.

B. Simulation of stripping gas to reboiler

A process with stripping gas added to the reboiler was simulated. The stripping gas was specified with temperature 190 °C, pressure 101 kPa, 99.17 % CO_2 and 0.83 % water [6]. The composition was similar to the flash gas composition from depressurization after the absorption column in the base case calculation. Fig. 5 shows the results from the Aspen HYSYS simulations using the TST model.

Calculations comparable to the base case simulations were performed with varying the stripping gas flow and number of stages in the absorption column. Using the TST model, less than 50 ppm was achieved using 10 absorption stages and 1 kmole/h stripping gas. Using 15 stages, it was necessary to use 0.6 kmole/h to achieve less than 50 ppm in the dehydrated gas.

There is some difference in calculated flash gas amount for the two models, 0.89 kmole/h for the PR model and 0.63 kmole/h for the TST model. This indicates that the two models calculate the CO_2 solubility in the TEG/water solution differently. When using the PR model, the water amount for a given stripping gas amount is slightly higher than using the TST model. But because the PR model calculates a higher flash gas amount, the PR model also calculates about 50 ppm when the amount of stripping gas is set to the amount of available flash gas.

Increasing the number of stages and the amount of stripping gas made it possible to improve the dehydration down to about 30 ppm. This was achieved with both the PR and the TST model.
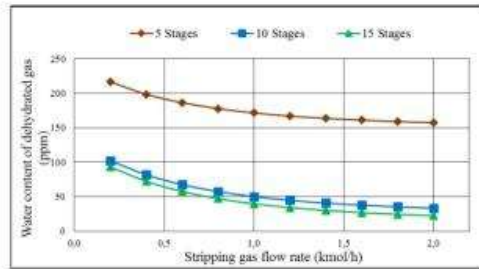

Fig. 5. Water content as a function of stripping gas and number of absorber stages with stripping gas to reboiler using the TST model

C. Simulation of stripping gas to extra column

A simulation of the process in Fig. 2 was performed. In earlier work [6] this process has been simulated using two strategies. The first strategy was to simulate the process with two columns. The second strategy was to simplify the flow-sheet model by simulating the desorption column and the extra stripping column as one column with heating at an intermediate stage as shown in Fig. 6. Similar numerical results have been achieved using these two strategies [6,13]. The strategy with only one column was chosen in this work because these simulations are easier to converge.

The extra stripping column was specified with 3 equilibrium stages. In the case of the extra stripping gas and the desorber simulated as one column, it was 3 stages between the stripping gas feed to the bottom and the reboiler heat addition. The number of stages above and below the feed in the desorber and in the extra stripping column have been varied in earlier work [16]. The results showed that increasing the number of stages in the sections above 3 (as specified in this work) did not improve the dehydration efficiency significantly.
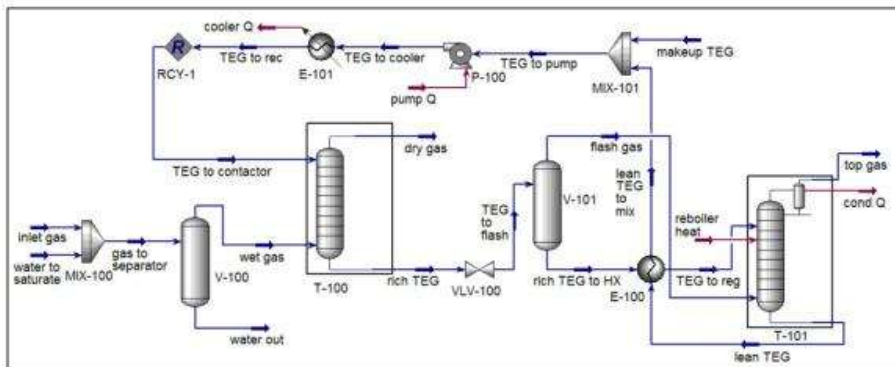

Fig. 6. Aspen HYSYS flow-sheet of dehydration process with extra stripping column
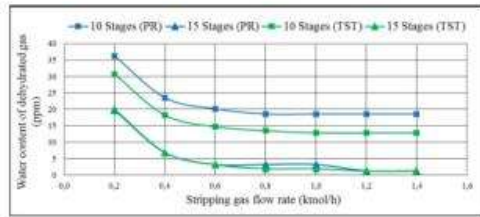
Fig. 7. Water content as a function of stripping gas and number of absorber stages with extra stripping column

Fig. 7 shows results from these calculations as a function of stripping gas and number of absorber stages for the process with an extra stripping column.

Using 10 absorber stages reduces the water content down to about 20 ppm using the PR model and down to about 15 ppm using the TST model. The water content is not reduced further by adding more stripping gas with 10 absorber stages. With 15 stages or more, less than 5 ppm was achieved using the available flash gas, and less than 2 ppm was achieved by adding more stripping gas. Using more than 15 absorber stages did not improve the dehydration efficiency significantly.

In earlier work [6] similar results were obtained using 20 absorber stages for the same conditions. With more than 1.2 kmole/h, less than 5 ppm was achieved with both the PR model and the TST model.

At very low water content (below 2 to 5 ppm) the water amount in the stripping gas is a limiting factor. Less than 1 ppm water in dehydrated gas has been calculated when pure (dehydrated) CO_2 was used as stripping gas. This was calculated using 15 absorber stages and a high stripping gas amount with both the PR and the TST model.

D. Simulation of a Drizo process

A Drizo process with n-heptane as stripping gas was simulated. The flow-sheet is shown in Fig. 8. The condensing of the stripping gas is performed in a three phase separator, and the organic phase is pumped back and evaporated in a heater before it is added to the bottom of the extra stripping column. It was checked that the amount of recirculated n-heptane was approximately equal to the n-heptane added to the desorber in the simulation.

A similar process has been simulated earlier [13]. An argument for not closing the loop in the simulation of the recirculation of the stripping gas, is that a small fraction of the added stripping gas will be dissolved in the solvent and lost e.g. in the flash gas. This is of minor interest for the evaluation of the process.

The results for n-heptane as a stripping gas using the TST model are presented in Fig. 9. Similar results were obtained using the PR model. Similar simulations were also performed with the components benzene and toluene as stripping gas. When using benzene and toluene, more of the stripping gas was dissolved in the glycol, so that the deviation between added stripping gas and recirculated stripping gas became larger.

Similar levels of water were achieved as in the simulations with flash gas or CO_2 as stripping gas. Less than 20 ppm was obtained using 10 absorption stages in the absorber and available flash gas. Less than 5 ppm was obtained with 15 stages and stripping gas amounts similar to the available flash gas and less than 1 ppm when the amount of stripping gas was increased.
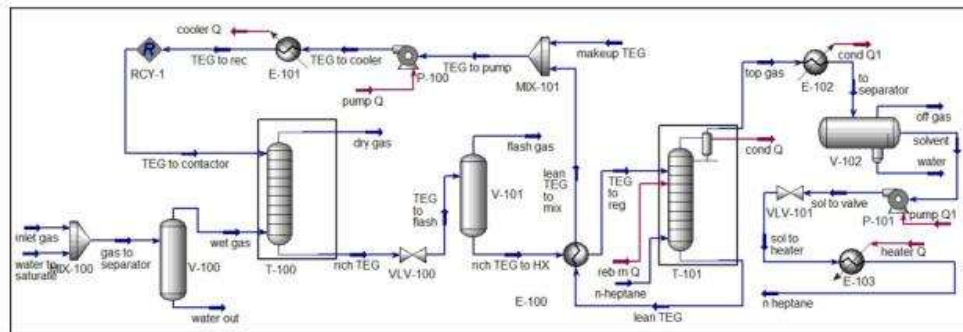

Fig. 8.   Aspen HYSYS flow-sheet of Drizo process using n-heptane as stripping gas
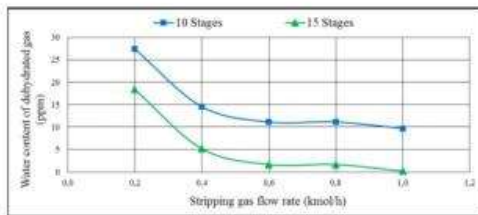
Fig. 9. Water content in Drizo process with n-heptane as a function of stripping gas and number of absorber stages using the TST equilibrium model

V. CONCLUSION

It is demonstrated that it is possible to simulate both a traditional glycol dehydration process and more advanced CO_2 dehydration processes like the Drizo process using the process simulation program Aspen HYSYS. The PR and TST models give similar results for all the simulated alternatives.

The calculated water content for different absorption pressures shows that a maximum dehydration efficiency is achieved at a pressure of about 5000 kPa.

A traditional TEG dehydration process is satisfactory to achieve a water content below 150 ppm in dehydrated CO_2. Using stripping gas, a water specification of less than 50 ppm can be achieved. In a Drizo process or a process including an extra stripping column and a high absorption column, it is possible to achieve less than 1 ppm water in dehydrated CO_2.

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Appendix 3: Moisture prediction chart [McKetta - Wehe]

Equilibrium Water Vapor Content of Sweet Natural Gas
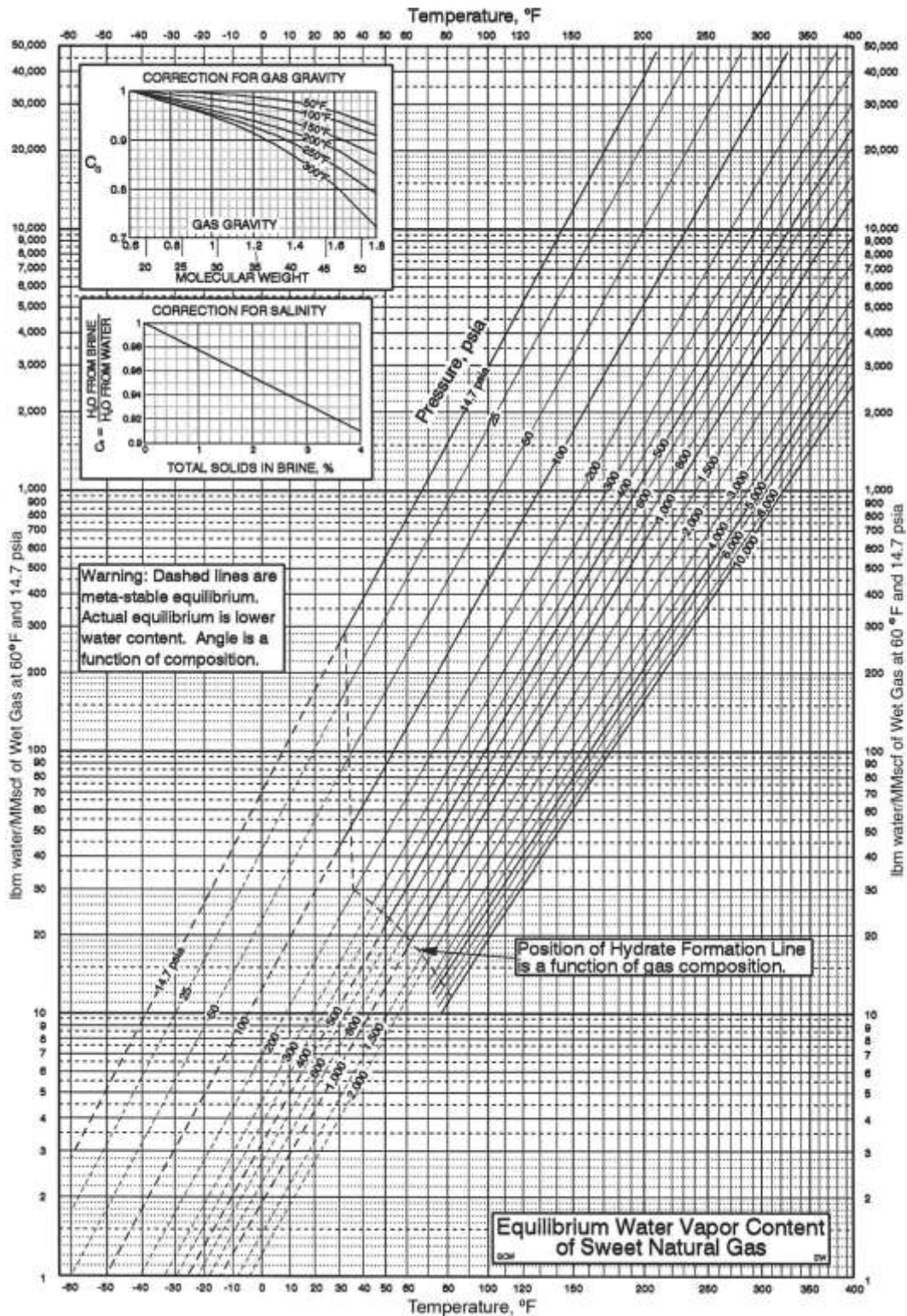
Appendix 4: Simulation results of TEG dehydration of CO_2 by introducing stripping gas to the reboiler

Stripping gas flow rate (kmol/h)	Water content of dehydrated gas (ppm)							
	5 Stages		10 Stages		15 Stages		20 Stages	
	PR	TST	PR	TST	PR	TST	PR	TST
0.2	258.6	216.7	123.4	102.3	108.3	93.2	107.7	93.2
0.4	240.9	198.4	101.9	81.9	87.6	72.3	86.9	72.3
0.6	227.9	186.4	85.3	67.5	71.8	57.5	71.1	57.5
0.8	218.7	177.7	73.7	57.4	60.4	47.2	59.6	47.2
1.0	212.1	171.5	67.1	50.1	51.9	39.7	51.2	39.7
1.2	207.1	166.9	60.9	44.7	45.5	34.2	45.1	34.2
1.4	203.5	163.6	56.1	40.7	40.5	30.0	40.0	30.0
1.6	200.9	161.0	52.5	37.6	36.6	26.8	36.2	26.8
1.8	198.9	158.9	49.5	35.2	33.5	24.4	33.2	24.4
2.0	197.3	157.3	46.8	33.3	31.0	22.4	30.8	22.4

Appendix 5: Simulation results of TEG dehydration of CO₂ by introducing stripping gas to the extra stripping column

Stripping gas flow rate (kmol/h)	Water content of dehydrated gas (ppm) - Peng Robinson & Twu-Sim-Tassone							
	5 Stages		10 Stages		15 Stages		20 Stages	
	PR	TST	PR	TST	PR	TST	PR	TST
0.2	185.6	155.7	36.3	30.7	20.0	19.7	20.0	19.4
0.4	175.0	145.0	23.5	18.2	6.9	6.8	6.9	6.8
0.6	172.7	142.0	20.2	14.7	3.2	3.2	3.2	3.2
0.8	172.7	141.0	18.6	13.5	3.2	1.9	3.2	1.9
1.0	172.7	141.0	18.6	12.8	3.2	1.9	3.2	1.9
1.2	172.7	141.0	18.6	12.8	1.3	1.3	1.3	1.2
1.4	172.7	141.0	18.6	12.8	1.3	1.3	1.3	1.2

Appendix 5: Simulation results of TEG dehydration of CO_2 by introducing portion of dehydrated CO_2 as stripping gas to the extra stripping column

Stripping gas flow rate (kmol/h)	Water content of dehydrated gas (ppm) - Peng Robinson & Twu-Sim-Tassone							
	5 Stages		10 Stages		15 Stages		20 Stages	
	PR	TST	PR	TST	PR	TST	PR	TST
0.2	185.9	154.9	36.3	29.8	20.01	18.76	20.01	18.76
0.4	175.2	144.4	22.6	17.4	5.90	5.97	5.90	5.97
0.6	172.7	141.3	19.1	13.8	2.18	2.24	2.18	2.24
0.8	172.7	140.3	19.1	12.5	2.18	0.96	2.18	0.96
1.0	172.7	140.3	17.5	12.5	2.18	0.96	2.18	0.96