

WHITE PAPER

# Acid Gas Cleaning using DEPG Physical Solvents: Validation with Experimental and Plant Data

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## Introduction

Acid gas removal is an important process in various branches of the hydrocarbon processing industry, primarily in natural gas processing and refining. Acid gas removal is also an essential part of other processes, such as coal gasification where carbon dioxide, hydrogen sulfide, carbonyl sulfides, mercaptans, and other contaminants need to be removed.

Acid gas is defined as gas containing significant amounts of contaminants, such as hydrogen sulfide ( $H_2S$ ), carbon dioxide ( $CO_2$ ), and other acidic gases. Sour gas is gas contaminated with  $H_2S$ . This term comes from the rotten smell due to sulfur content (1). Thus, “gas sweetening” refers to  $H_2S$  removal, because it improves the odor of the gas being processed, while “acid gas removal” refers to the removal of both,  $CO_2$  and  $H_2S$ .

Acid gases need to be removed in order to comply with sales gas quality regulations. These regulations are in place to minimize environmental impact and ensure gas transport pipeline integrity, avoiding undesired occurrences, such as corrosion caused by  $H_2S$  and  $CO_2$  in the presence of water. Acid gases also need to be removed due to the toxicity of compounds, such as  $H_2S$ , and the lack of the heating value of  $CO_2$ . Typically, “pipeline quality” or sales gas is required to be sweetened to contain concentrations of  $H_2S$  that’s no more than 4 parts per million (ppm), and a heating value of no less than 920 to 1150 Btu/SCF, depending on the final consumer requirements (2).

There are numerous processes developed for acid gas removal, and they typically fall into one of the five categories: chemical solvents (amines), physical solvents, adsorption, membranes, and cryogenic fractionation (3, 4).

When gas processors turn to absorption processes for acid gas removal, several factors affect their decision in choosing whether to use a chemical or physical absorption process from an economic standpoint. They take into account the required solvent circulation rate that affects capital and operating costs by strongly influencing equipment size and energy requirements for solvent regeneration (4). In this paper, we will describe acid gas cleaning via absorption processes with emphasis on the use of physical solvents.

## Acid Gas Cleaning – Brief Process Overview

A typical flow diagram of a gas treating unit is shown in Figure 1. The acid gas is sent to a separator to remove any entrained liquid or sand and then fed to the bottom of the absorber column. The absorber can be a tray or packed tower, although packing is usually preferred due to high capacity and better options for materials of construction.

The feed gas then flows upward, counter-current to the lean amines or physical solvent solution which is introduced in one or more stages around the top of the absorber. The cleaned gas exits the top of the column. The solvent with the absorbed acid gas, called rich amines (or solvent), is sent to a flash drum and a second “Stripper” column, to be regenerated by means of heating in the case of the chemical solvent. Physical solvent regeneration is completed by reducing the pressure in a couple of stages, unless deep cleaning of  $H_2S$  or  $CO_2$  is required, in which case, a stripper column will be used. As shown in Figure 1, there are many unit operations involved in this process, and operating the gas cleaning unit optimally will require control and sound engineering judgment. Process simulation is a critical tool, not only to optimize the acid gas cleaning unit alone, but for the entire gas treating facility.

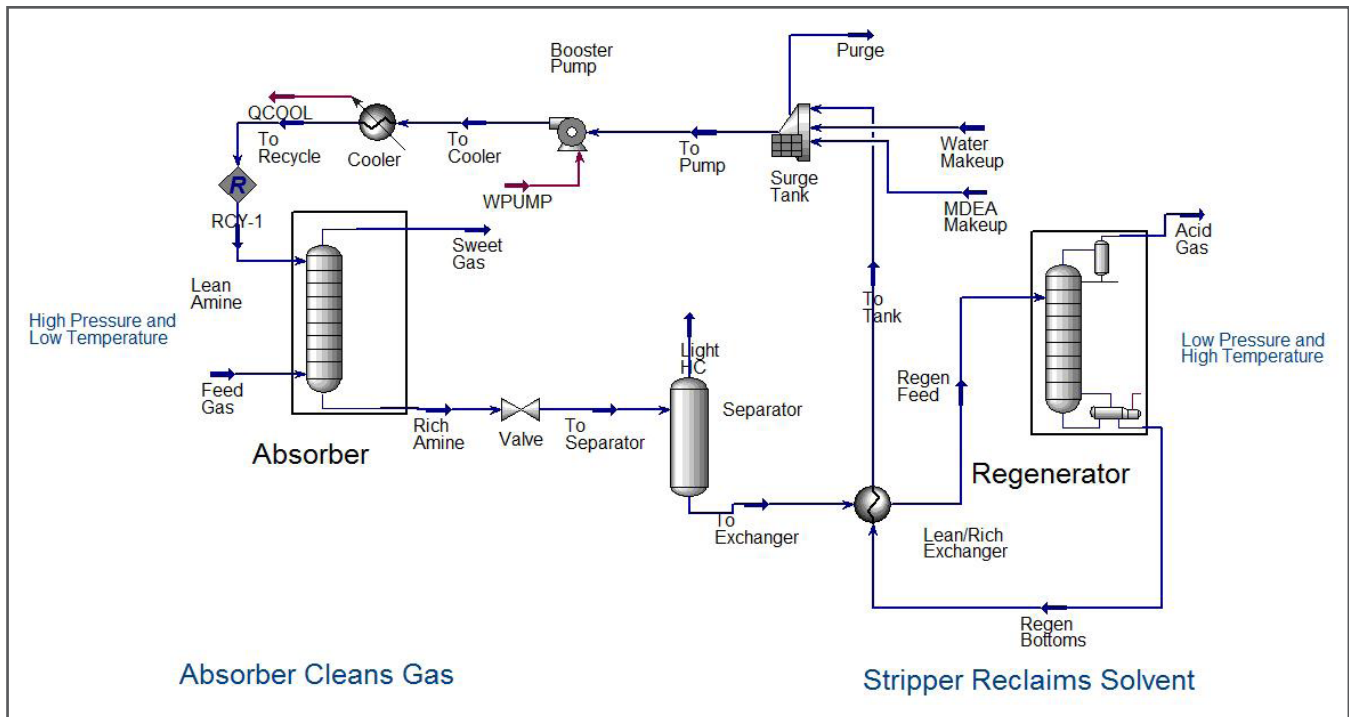


Figure 1: Typical acid gas treating unit: Absorber cleans gas, Regenerator (Stripper) reclaims solvent

Acid Gas Cleaning is an integral functionality of Aspen HYSYS® version 8.3 and higher. The “Acid Gas” property package in Aspen HYSYS provides the thermodynamics based on the Electrolyte NRTL model (12) with all the necessary aqueous-phase equilibrium and kinetics reactions required for rigorous calculations of the process. In Aspen HYSYS V8.6, the Acid Gas Cleaning functionality has been enhanced with a new property package, “Acid Gas - Physical Solvents”, based on the Perturbed Chain Statistical Association Fluid Theory (PC-SAFT) Equation Of State, which allows users to model Dimethyl Ether of Polyethylene Glycol (DEPG), a constituent of a commercially available solvent called Selexol®\*.

The DEPG generic formula is  $\text{CH}_3\text{O}(\text{C}_2\text{H}_4\text{O})_n\text{CH}_3$  with  $n$  ranging from 2 to 9. Some of the most common uses of DEPG include:

- CO<sub>2</sub> removal from natural gas
- CO<sub>2</sub> and H<sub>2</sub>S removal from natural gas
- CO<sub>2</sub>, H<sub>2</sub>S, and COS removal from gasifier product
- CO<sub>2</sub> removal in ammonia plants
- Mercaptan removal from molecular sieve regeneration gas
- Contaminant removal from landfill gas
- Controlling the hydrocarbon dew point (HCDP) to comply to pipeline specifications

\* Selexol is a registered trademark of Allied Chemical Corporation. All other trademarks cited here are the property of their respective owners.

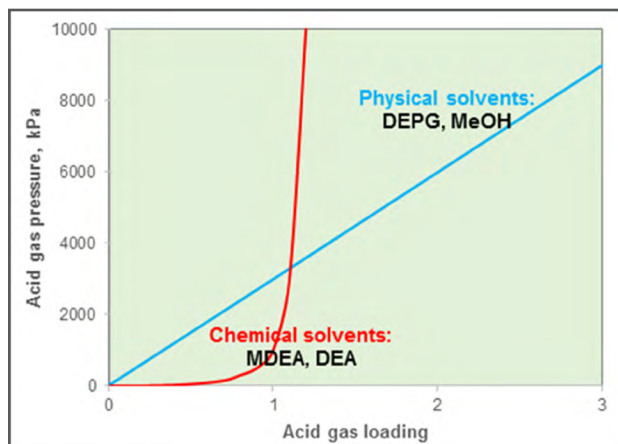


Figure 2: Comparison of effectiveness of physical vs. chemical solvents.

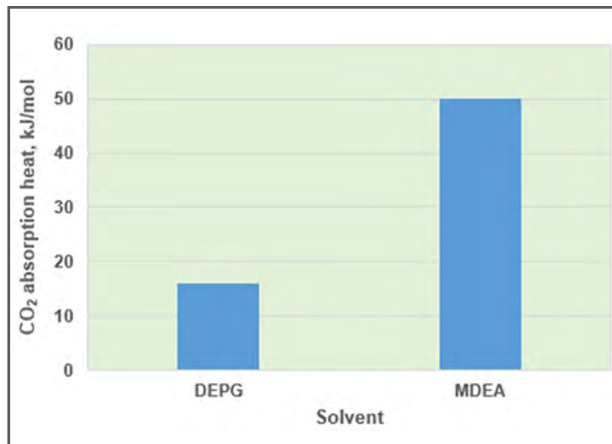


Figure 3: Relative magnitude of heat of absorption of physical vs. chemical solvent

Physical solvents are normally favored over chemical solvents in processes with very high acid gas concentration for bulk removal of sour gas. However, when high recovery of heavy hydrocarbons is desired, the use of physical solvents may not be the optimal solution, since they have a higher co-absorption of hydrocarbons. Thus, much of the research on the subject of choosing the right type of solvent is concerned with selecting a solvent with high capacity for acid gas and low capacity for hydrocarbons (3). Some of the advantages of using a physical solvent, such as DEPG, include lower solvent regeneration energy requirements, temperature for separation that is close to ambient, and partial gas dehydration during the absorption process (4). Another advantage of using a physical solvent, such as DEPG, is that it is stable and non-corrosive so that special metallurgy is not required, reducing relative capital and operating costs.

Typically, chemical solvents are most suitable at lower pressures, and physical solvents are favored for higher acid gas partial pressures, as shown in Figure 2. Figure 2 describes the acid gas loading as a function of acid gas partial pressure. The red line shows that at lower partial pressure of acid gas, the chemical solvent is very effective in cleaning the gas, up to a point where the capacity is plateaued. For the physical solvent, shown by the blue line, the relationship is linear and is more effective at higher partial pressures. Figure 3 shows lower energy per mol of CO<sub>2</sub> absorption by DEPG when compared with MDEA. This results in lower energy required to regenerate the DEPG solvent.

## DEPG Solubility Trends Affect Its Applicability

DEPG suitability for a particular range or applications, such as acid gas cleaning of natural or synthetic gas, is influenced by its solubility trends, as shown in Figure 4 (5). For example, solubility of CO<sub>2</sub>, H<sub>2</sub>S, COS, CS<sub>2</sub>, and mercaptans in DEPG is significantly higher than that of H<sub>2</sub>, CO, and methane. Thus, the former list will be preferentially absorbed.

There is a high selectivity of H<sub>2</sub>S over CO<sub>2</sub> (about a factor of 10). We can also see that heavy hydrocarbons are also quite soluble in DEPG, thus DEPG is not very suitable for natural gases with high natural gas liquids (NGL) content since valuable components can be lost with the solvent. Alternatively, this characteristic has been exploited to control the hydrocarbon dew points, which is beneficial in gas transport (5).

Relative solubility of various gases in Selexol <sup>®</sup> solvent			
Component	$R = \frac{K' \text{Component}}{K'CH_4}$	Component	$R = \frac{K' \text{Component}}{K'CH_4}$
H <sub>2</sub>	0.20	H <sub>2</sub> S	134
N <sub>2</sub>	0.30	C <sub>6</sub> H <sub>14</sub>	167
CO	0.43	CH <sub>3</sub> SH	340
CH <sub>4</sub>	1.00	C <sub>7</sub> H <sub>16</sub>	360
C <sub>2</sub> H <sub>6</sub>	7.20	CS <sub>2</sub>	360
CO <sub>2</sub>	15.2	C <sub>2</sub> H <sub>5</sub> Cl	400
C <sub>3</sub> H <sub>8</sub>	15.4	SO <sub>2</sub>	1400
<i>i</i> - C <sub>4</sub> H <sub>10</sub>	28	C <sub>6</sub> H <sub>6</sub>	3800
<i>n</i> - C <sub>4</sub> H <sub>10</sub>	36	C <sub>2</sub> H <sub>5</sub> OH	3900
COS	35	CH <sub>2</sub> Cl <sub>2</sub>	5000
<i>i</i> - C <sub>5</sub> H <sub>12</sub>	68	CH <sub>2</sub> Cl <sub>3</sub>	5000
C <sub>2</sub> H <sub>2</sub>	68	C <sub>2</sub> H <sub>2</sub> S	8200
NH <sub>3</sub>	73	H <sub>2</sub> O	11000
<i>n</i> - C <sub>5</sub> H <sub>12</sub>	83	HCN	19000

Figure 4: Relative solubility of various gases in Selexol<sup>®</sup> solvent (5)

## Modeling Chemical (Amines) & Physical Solvents

One of the strategies for selecting a chemical or physical solvent to scrub the acid gas is to determine which solvent has the needed selective affinity to the contaminants. Chemical solvents, such as alkanolamines—MDEA and DEA, are widely used in the industry, and work by reacting with the contaminants. Since heat is required for regeneration, the higher operating costs need to be accounted for in the selection process. Since physical solvents, such as DEPG, preferentially absorb the contaminants through physical means, pressure reduction can regenerate the solvent without the use of heat, thus minimizing operating costs.

The technology for modeling the chemical and physical solvent processes for removal of acid gases can be separated into two areas—the thermodynamic package and the simulation engine.

The thermodynamic package technology for chemical solvent modeling is based on the Electrolyte Non-Random Two-Liquid (Electrolyte NRTL) model for electrolyte thermodynamics and Peng-Robinson Equation of State for vapor phase properties. Regression has been performed with available

VLE and heat of absorption data for many amine solvents, including all major amine solvents used in the industry, such as: MDEA, MEA, DEA, PZ, PZ+MDEA, DGA, DIPA, Sulfolane-DIPA, Sulfolane-MDEA, and TEA (see Appendix I for abbreviations decoded).

Two models are available for the simulation of the absorber and regenerator units—Efficiency and Advanced. Both are based on AspenTech's proprietary Rate-Based technology. The Advanced model uses Maxwell-Stefan theory (8) to rigorously calculate the heat and mass-transfer rates without assuming thermal or chemical equilibrium between the vapor and liquid for each stage. The Efficiency model uses a conventional equilibrium-stage model to solve the column, but the non-equilibrium behavior inherent in acid gas systems is modeled by calculating a Rate-Based efficiency for  $\text{CO}_2$  and  $\text{H}_2\text{S}$  at each stage. The efficiencies are computed using the same underlying correlations for mass transfer and interfacial area used by the Efficiency model. The results from the Efficiency and Advanced models are comparable for most systems, but the Efficiency solves much faster due to its simplicity. The Advanced model is recommended when contaminants other than  $\text{H}_2\text{S}$  and  $\text{CO}_2$  are present in the feed gas.

Several examples of modeling amines are distributed with Aspen HYSYS. Additional information on the subject can be found using the "Search" functionality within Aspen HYSYS, where you can access webinars, Computer Based Training, Jump Start Guides, and more.

**The focus of this work is on physical solvents - DEPG.**

Physical solvent modeling in Aspen HYSYS employs the PC-SAFT equation of state, which follows the recommendations of the Final Report for Consortium of Complex Fluids (6). The PC-SAFT equation includes an association term that accounts for strong intermolecular forces which can better predict system behavior with associating compounds. It is a proven model that can represent a wide range of compounds, including hydrocarbons, inorganic gases present in natural gas streams, water, and other polar and associating components. The model can fit vapor pressure, liquid density, and liquid heat capacity very well without requiring volume translation terms. Often both VLE and LLE can be represented with the same binary interaction parameters.

The pure component parameters of the PC-SAFT model were obtained from open literature and regressed from experimental vapor pressure, liquid density, and liquid heat capacity data by AspenTech staff. Binary parameters were regressed using experimental data primarily from the NIST TDE source data archive and GPA research reports. Actual references used in data regression analysis are provided in subsequent sections. NIST TDE contains a comprehensive collection of experimental measurements of thermodynamic and thermochemical properties of pure components and mixtures and is accessible free of charge in Aspen Plus®.

# Modeling Physical Solvents – Pure Component Properties Validation

Data available from open literature, such as Coastal AGR Solvent Bulletin (7), was used to determine the pure component parameters of DEPG. Vapor pressure, liquid density, and liquid heat capacity data were used. The fit using PC-SAFT is satisfactory, as shown in Figures 5-7 below.

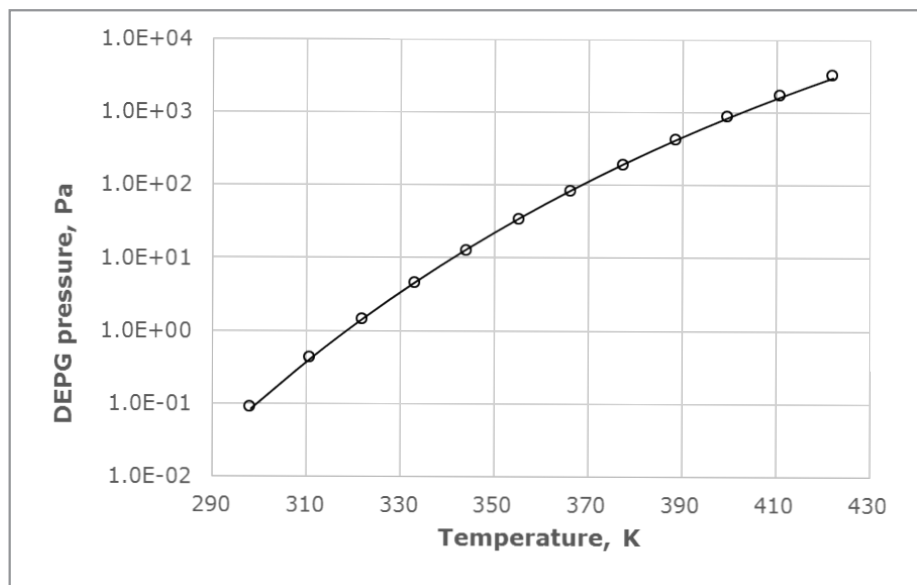


Figure 5: DEPG vapor pressure, experimental (7): (o); PC-SAFT model: (lines)

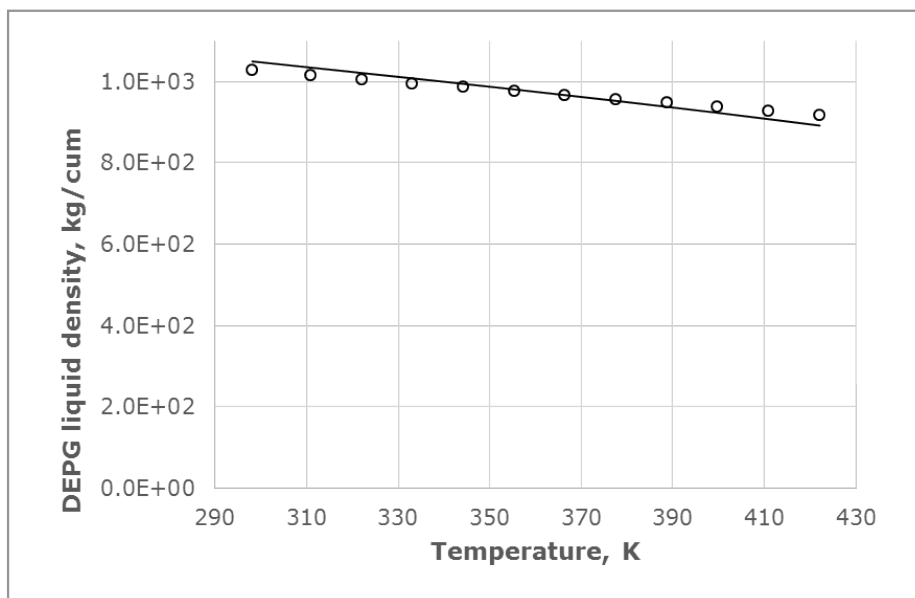


Figure 6: DEPG liquid density, experimental (7): (o); PC-SAFT model: (lines)

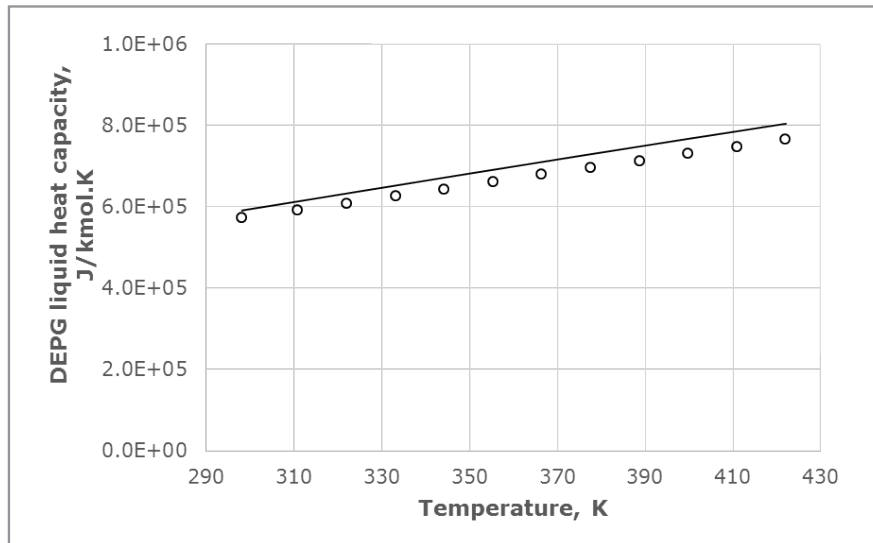


Figure 7: DEPG liquid heat capacity, experimental (7): (o); PC-SAFT model: (lines)

## Modeling Physical Solvents – VLE Validation

VLE data for DEPG in the open literature is limited and some references (9) and (10) are shown below. The fit to the data and the extrapolation to low concentration are good.

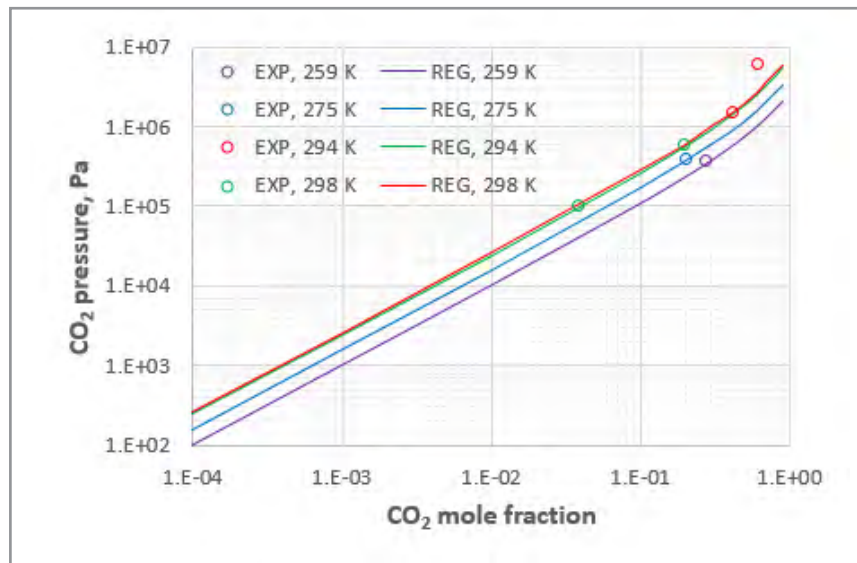


Figure 8: Experimental Data from Kutsher, et.al. (9) (points) compared to PC-SAFT model: (lines)



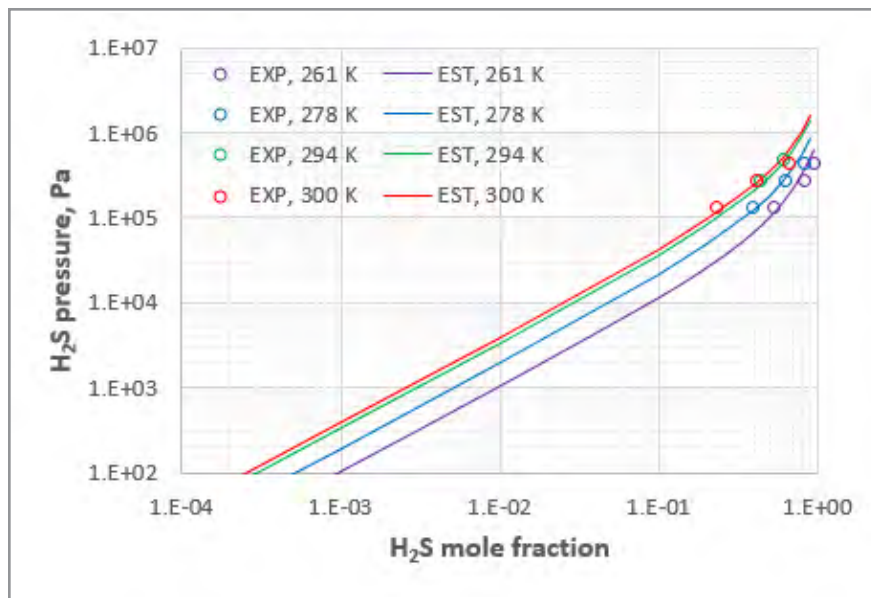


Figure 9: Experimental Data from Ameen, et. al. (10) (points) compared to PC-SAFT model: (lines)

## Modeling Physical Solvents – Solubility Trends Validation

The following components are commonly encountered in the gas removal process: CO<sub>2</sub>, H<sub>2</sub>S, COS, CO, mercaptans, hydrocarbons—light and heavy, nitrogen, water, HCN, and BTEX. Most of the pure component parameters for these components were obtained from the literature, while a few were re-regressed or had to be regressed because none could be found in the literature. Data for vapor pressure, liquid density, and heat capacity, if available, were used in the regression. Vapor pressure was given the highest weight, thus fit most accurately while maintaining reasonable accuracy for density and heat capacity. Water, ammonia, and HCN were modeled with 2B association scheme (6).

Solubilities of key components in DEPG were calculated and shown as a function of temperature and pressure in Figures 10 and 11 to show that the PC-SAFT model is able to represent the expected behavior as described earlier (see Figure 4). For example, as shown in Figures 10 and 11, CO<sub>2</sub> and H<sub>2</sub>S are much more soluble in DEPG than methane and H<sub>2</sub>. H<sub>2</sub>S is much more soluble than CO<sub>2</sub>.

We also see that higher temperature causes solubility of CO<sub>2</sub> and H<sub>2</sub>S to decrease significantly, but not so much for methane and H<sub>2</sub>. More solubility validation data and references are available in Appendix II.

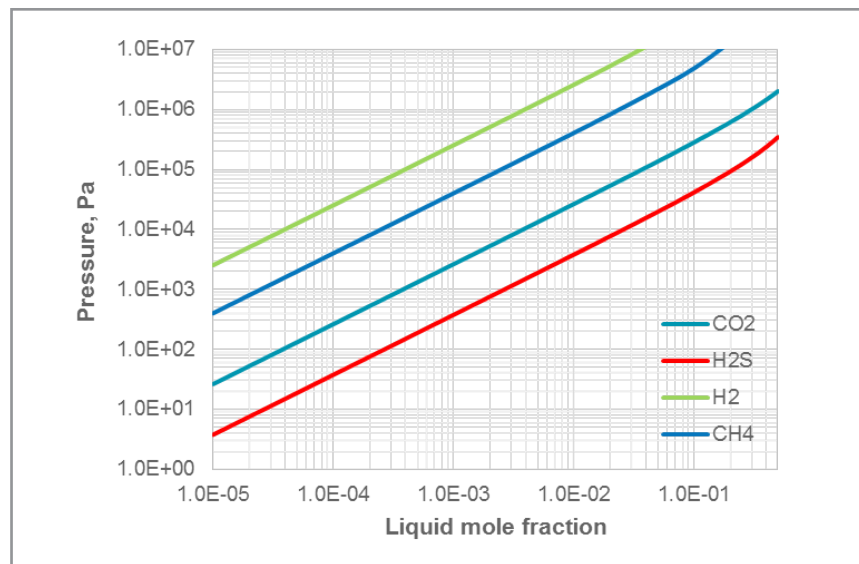


Figure 10: Solubility of  $\text{CO}_2$ ,  $\text{H}_2\text{S}$ ,  $\text{H}_2$ , and  $\text{CH}_4$  in DEPG at 298.15 K

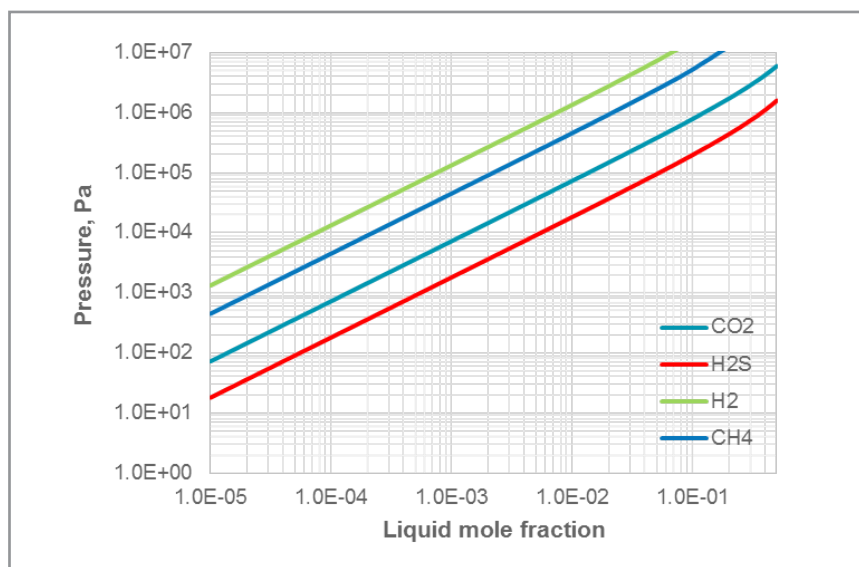


Figure 11: Solubility of  $\text{CO}_2$ ,  $\text{H}_2\text{S}$ ,  $\text{H}_2$ , and  $\text{CH}_4$  in DEPG at 373.15 K

## Modeling Physical Solvents – Flowsheet Validation

Besides validating the thermodynamic model with laboratory data, the simulation model has also been validated against published data (11). The data is not actual plant measurements, but design cases from the report of Argonne National Laboratory (11). See Figure 12 and Table 1 below for reference.

In this work, a simulation model of the acid gas cleaning process using Aspen HYSYS was developed to represent the process described in (11). The feed stream contains a significant amount of 40% nitrogen, 30% hydrogen, 25%  $\text{CO}_2$ , and some amount of  $\text{H}_2\text{S}$ . Two cases were presented, one at 995 psia the other at 245 psia with the configuration and the flowsheet as shown in Figure 12.

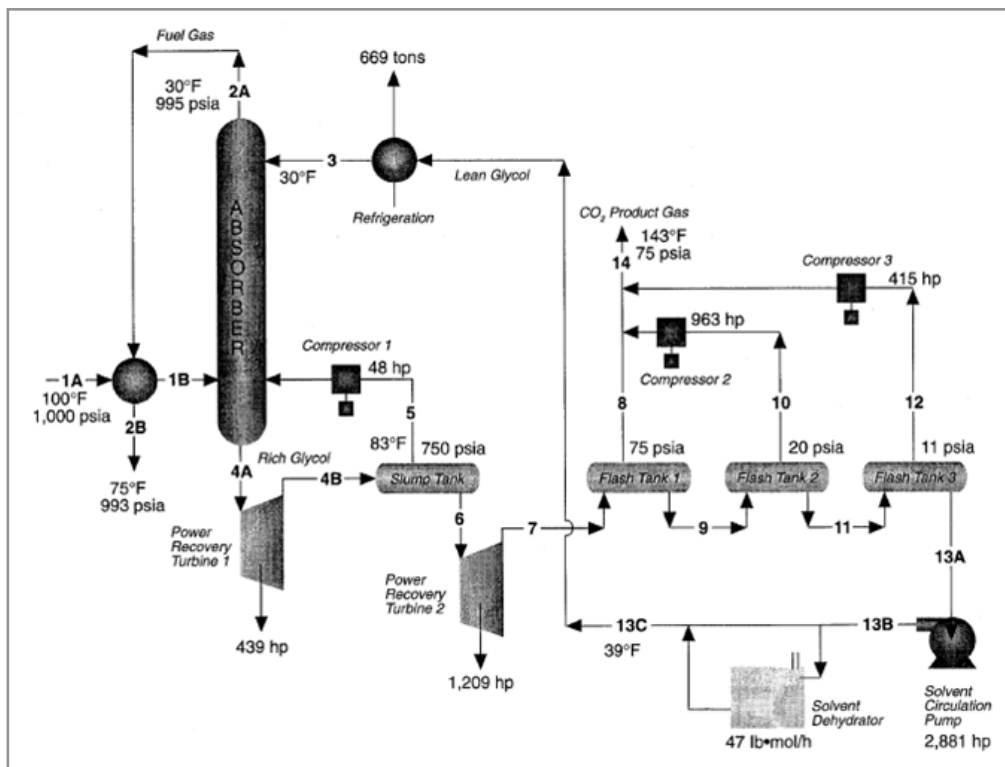


Figure 12: Flowsheet diagram of Glycol Process for CO<sub>2</sub> recovery in Case 1 of reference 11

Selected simulation results are presented in Table 1. The simulation results match reasonably well with the results from the report.

Case	Component	Sour gas, lb mol/h		Treated gas, lb mol/h		Rich solvent, lb mol/h	
		Ref. 11	HYSYS	Ref. 11	HYSYS	Ref. 11	HYSYS
High Pressure - 995 psia	CO <sub>2</sub>	4335.99	4335.99	216.93	260.24	4234.74	4191.43
	H <sub>2</sub> S	0.4	0.4	0.04	0.03	0.36	0.37
Low Pressure - 245 psia	CO <sub>2</sub>	4335.99	4335.99	218.15	220.49	4514.19	4511.85
	H <sub>2</sub> S	0.4	0.4	0.05	0.04	0.35	0.36

Table 1: Comparison of Aspen HYSYS simulation using DEPG property package with results from reference 11

## Conclusion

The Acid Gas - Physical Solvent property package has been developed for Aspen HYSYS to simulate acid gas removal processes using the PC-SAFT equation of state. PC-SAFT is a sound and well-proven model, which has been successfully used by AspenTech's customers over the years. The necessary model parameters had been developed and validated in this work using extensive laboratory and other available data. A simulation flowsheet model was also developed and shown to provide reasonable results.

The simulation model and the thermodynamic package were tested in Aspen HYSYS, the industry leader in process simulation for the energy and E&C verticals, demonstrating that the current modeling technology is suitable to serve the needs of the industry.

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Note: Irina Rumanyantseva, a former product marketing manager at AspenTech, was an original author of this paper.

# Appendix I

## Abbreviations

<b>MDEA</b>	Methylenedioxyethylamphetamine
<b>MEA</b>	Monoethanolamine
<b>DEA</b>	Diethanolamine
<b>PZ</b>	Piperazine
<b>DGA</b>	Aminoethoxyethanol (Diglycolamine)
<b>DIPA</b>	Diisopropanolamine
<b>TEA</b>	Triethanolamine
<b>DEPG</b>	Dimethyl Ether of Polyethylene Glycol

# Appendix II

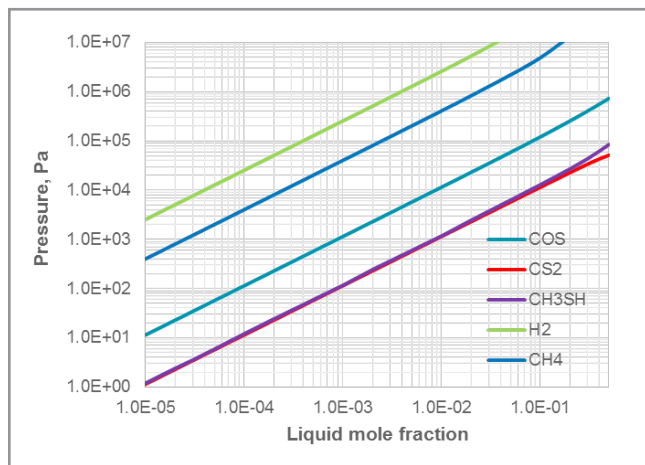


Figure 13: Solubility of COS, CS<sub>2</sub>, CH<sub>3</sub>SH, H<sub>2</sub>, and CH<sub>4</sub> in DEPG at 298.15 K. High solubility of COS, CS<sub>2</sub> and CH<sub>3</sub>SH over H<sub>2</sub> and CH<sub>4</sub>

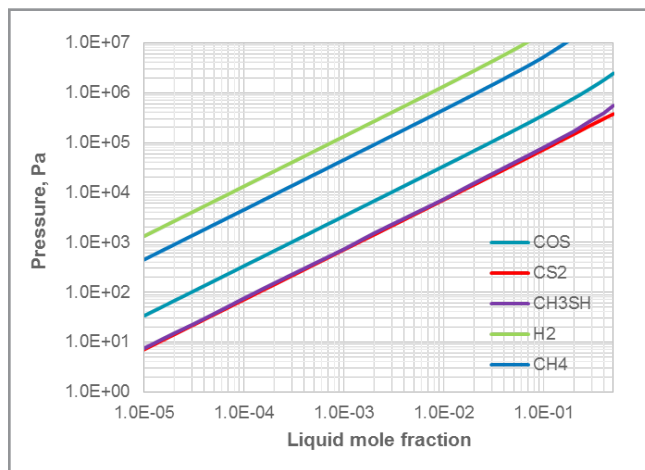


Figure 14: Solubility of COS, CS<sub>2</sub>, CH<sub>3</sub>SH, H<sub>2</sub>, and CH<sub>4</sub> in DEPG at 373.15 K. High solubility of COS, CS<sub>2</sub> and CH<sub>3</sub>SH over H<sub>2</sub> and CH<sub>4</sub>

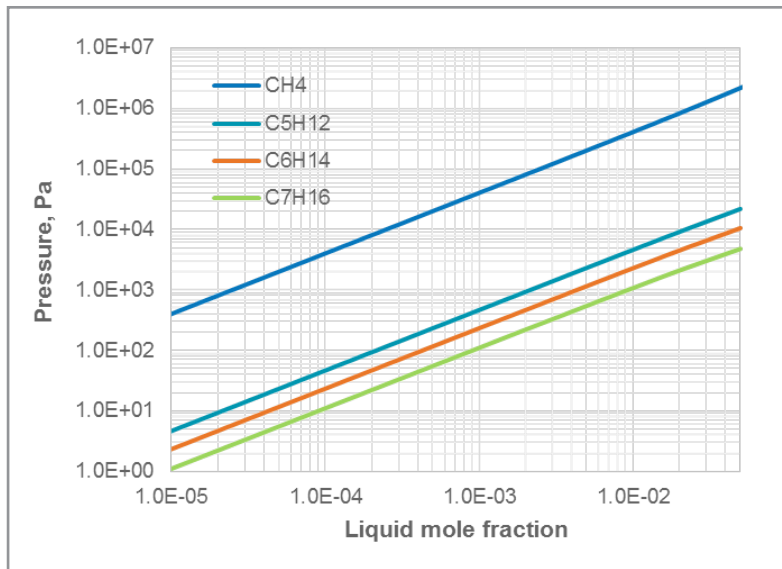


Figure 15: Solubility of heavy hydrocarbons in DEPG at 298.15 K. Solubility increases as the carbon number increases. At higher temperature, they are less soluble

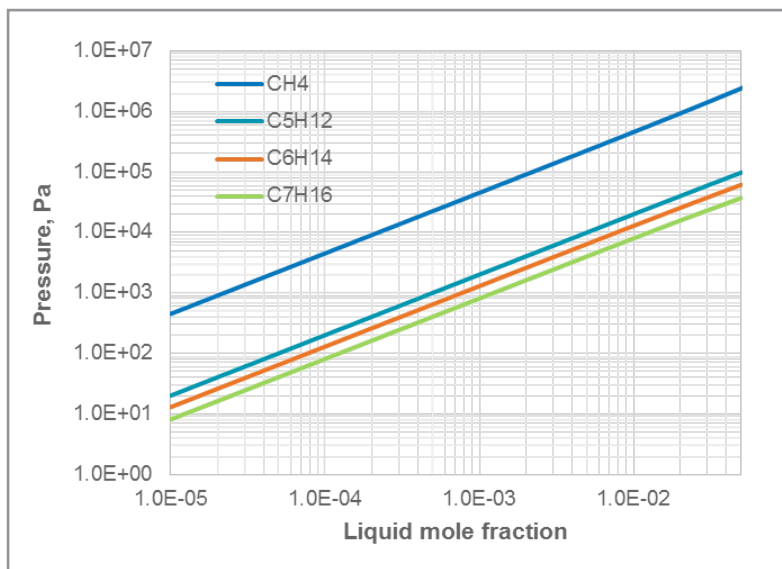


Figure 16: Solubility of heavy hydrocarbons in DEPG at 373.15 K. Solubility increases as the carbon number increases. At higher temperature, they are less soluble

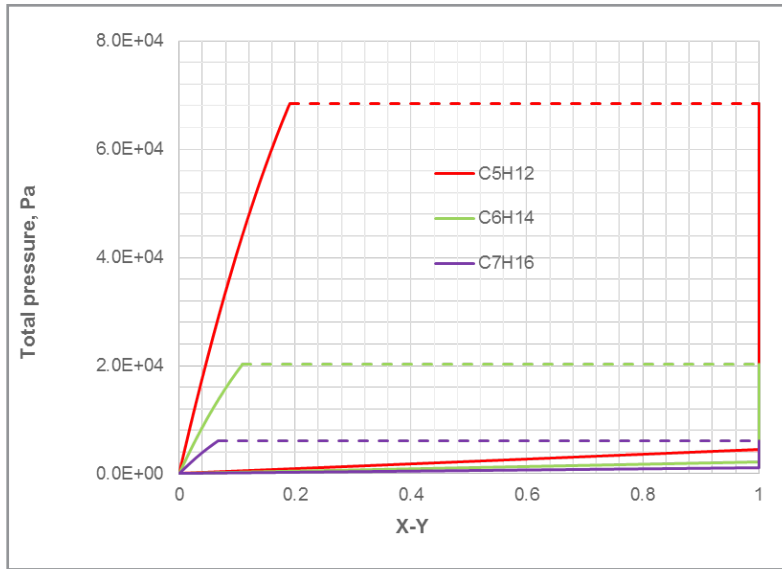


Figure 17: VLE of Hydrocarbons-DEPG at 298.15 K

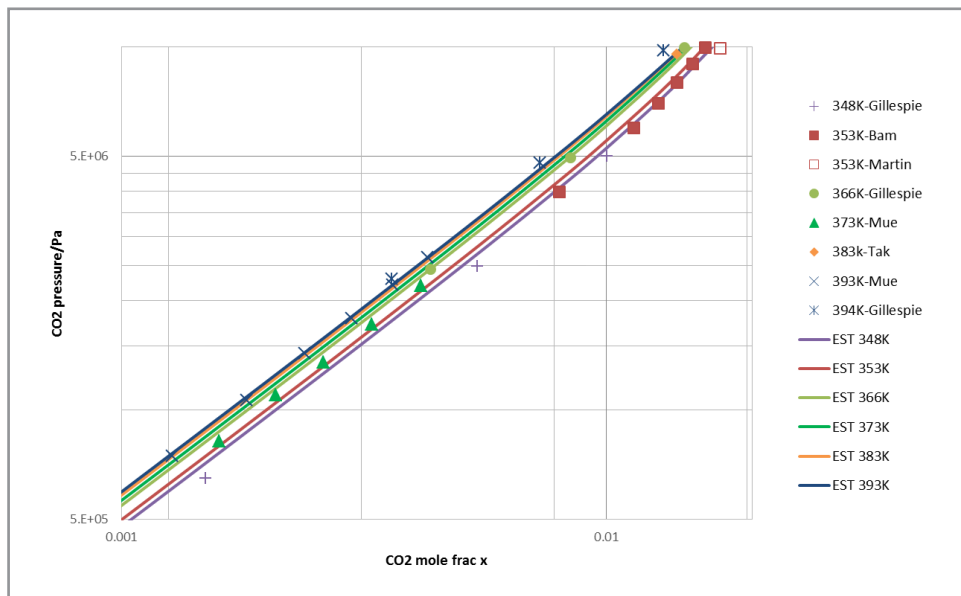


Figure 18: VLE of CO2 and H2O. The data span of wide temperature and pressure ranges

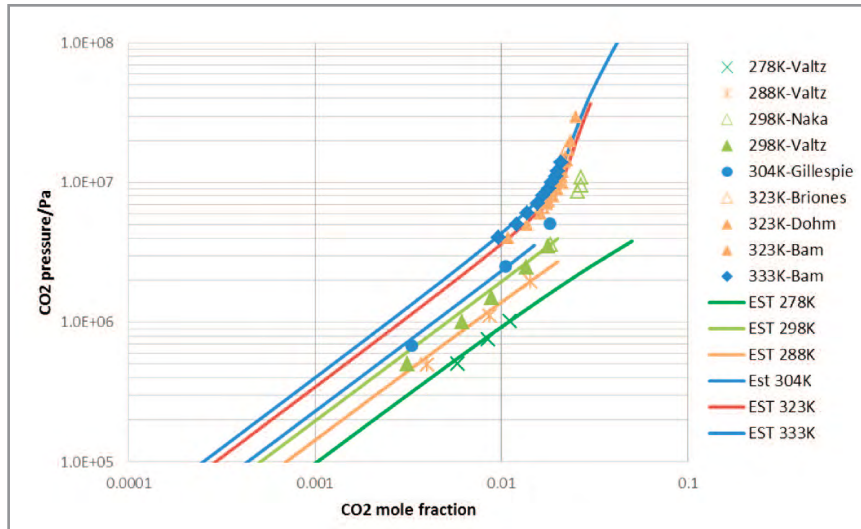


Figure 19: VLE of CO<sub>2</sub> and H<sub>2</sub>O. The data span of wide temperature and pressure ranges

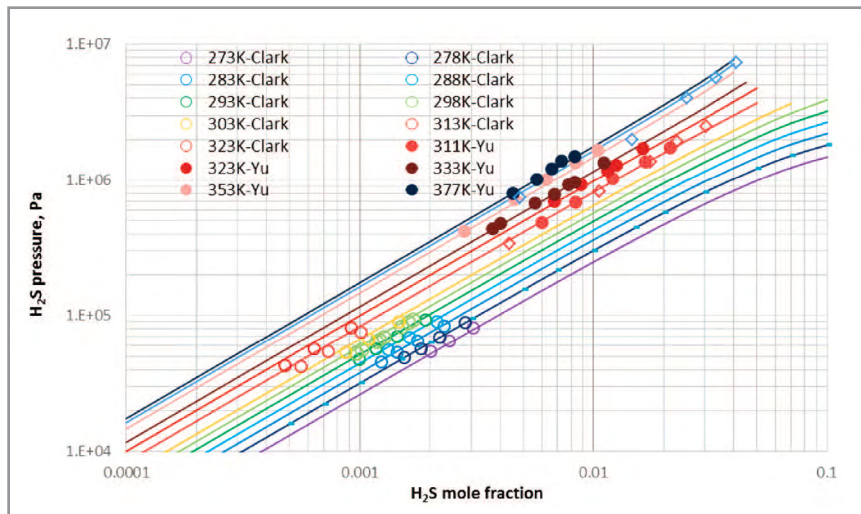


Figure 20: VLE of H<sub>2</sub>S and H<sub>2</sub>O. The data span of wide temperature and pressure ranges



## Appendix II - References

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