Recent Advances in
Petrochemical Science
ISSN: 2575-8578

Juniper Publishers
key to the Researchers

Editorial
Volume 3 Issue 1 -september 2017
DOI: 10.19080/RAPSCI.2017.03.555603

Recent Adv Petrochem Sci

Phase Identification of Natural Gas System with High CO_2 Content for Cryogenic Based Separation Process

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Submission: September 12, 2017 ; **Published:** September 20, 2017

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Abstract

The liquid-vapor and solid-vapor domains of natural gas mixture with high CO_2 content for cryogenic separation have been obtained through simulation using Aspen Hissy. Pressure-temperature phase diagram is generated for selected gaseous mixtures and the phase boundaries of both liquid-vapor region and solid-vapor region are identified. The identified domains can prove to be helpful in separation of CO_2 and other natural gas components by using appropriate cryogenic process.

Introduction

The demand for oil, natural gas and other energy sources is growing dramatically as it is estimated that worldwide energy consumption will increase by more than 40% by 2035 [1]. The growing demand is also fueled by an ever increasing population [2]. Natural gas is the largest single contributor to the world's total energy requirement as it used in domestic heating purposes and also for electricity generation and it is also considered as a clean fuel. But one third of the proven natural gas reserves are estimated to be sour and Malaysia alone have some natural gas reserves where CO_2 concentration is more than 70% [3,4]. The high content of CO_2 in natural gas enhances the formation of carbonic acid and dry ice causing corrosion and clogging of delivery pipelines. Hence, the removal of CO_2 from the natural gas is important to enhance the calorific value of natural gas [5]. With the growing population and demand for energy it is necessary to innovate technologies to develop an efficient process for carbon dioxide removal from gas streams with natural gas reserves containing high CO_2 content [6]. The cryogenic process is an emerging technology for carbon dioxide capture and purification of natural gas, which needs systematic research for future applications [7]. Cryogenics separation study involves the separation of natural gas mixture constituents at lower temperatures. Cryogenics technology is advantageous over other existing amine absorption or adsorption based processes as no chemical and solvents are required by the process, process makeup water supply is not needed, no solvent regeneration equipment are required, CO_2 is available at higher pressure and can be used for Enhanced Oil Recovery or sequestration purposes. Also, Natural Gas Liquids (NGL) are obtained as a by-product which have good market potential. At atmospheric pressure, CO_2 directly de-sublimes from gas phase to solid phase below the saturation temperature [3]. In this work, liquid-vapor and solid-vapor domains for hybrid cryogenic process have been identified through simulation. The identification of temperature and pressure ranges in hybrid application can prove to be beneficial for further design and optimization of the process.

Methodology

Table 1: Natural gas feed streams composition.

Component	Composition (Mol %)	
	Gas Mixture 1	Gas Mixture 2
CH_4	20	30
CO2	70	60
C_2H_6	5	5
C_3H_8	2.8	2.8
C_4H_{10}	1.1	1.2
C_5H_{12}	1	1

In order to identify the phase regions, feed gas includes natural gas with high CO_2 content comprising of methane to pentane as shown in Table 1. The composition is based on the maximum possible amount of heavier hydrocarbons that exists in Malaysian natural gas mixture containing high content carbon dioxide. The pressure temperature phase diagram is generated in Aspen Hissy 8.0 Peng-Robinson equation of state has been used to generate phase diagram for the selected feed streams as in the case of hydrocarbon gaseous mixture, it predicts the VLE behavior most accurately [8].

Results and Discussion

Figure 1 shows the pressure-temperature phase diagram for natural gas mixture for the selected feed stream compositions which identify the solid, liquid and vapor regions for the study. The regions in which separation can be performed are highlighted in Figure 1. It can be seen that the L-V region starts from about 0 °C to -60 °C and S-V domain lies in the temperature range of -60 °C to -120 °C. Bubble point and dew point curves have been obtained for the natural gas feed containing 60% and 70% CO_2 in feed for the maximum pressure of up to 40 bars, which is operational limit pressure for the experimental equipment. For the pressure ranges higher than that, operational problems as column choking can occur in the colder sections of the distillation column [9]. In the figure shown above, it can be seen that at higher temperatures as 40 °C to 0 °C, the gas mixture is in vapor state. It can also be noticed that the vapor-liquid region lies in the temperature range of 0 to -60 °C and beyond -60 °C to about -120 °C, the phase enters into solid-vapor region. At high pressures above 40 bar and above the CO_2 freeze out temperature, the phase lies in liquid region. At very low temperatures as -120 °C and lower than this and at moderate pressures, it enters into solid-liquid region. It is feasible to do separation in liquid-vapor and solid-vapor ranges as energy requirements to lower the temperatures to solid-liquid regions become high. Hybrid cryogenic separation setup consists of flash vessel in conjunction with packed bed. Separation of heavier hydrocarbons as butane and pentane can be done in liquid-vapor domain by using flash separator as these components liquefy at relatively lower pressures and higher temperatures. CO_2 which is still in vapor phase in L-V region can be preceded to cryogenic packed bed in S-V domain and desublimated onto the surface of the packing. Purified methane which is still in vapor form inside packed bed can be obtained and used for commercial purposes.

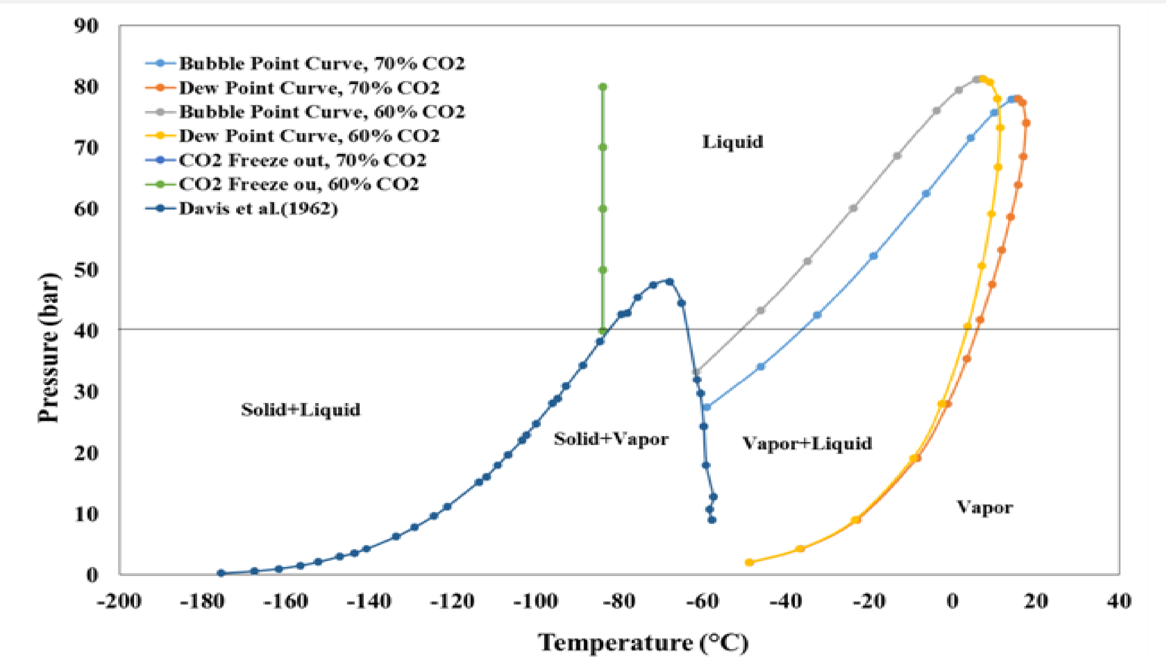

Figure 1: Pressure-temperature phase diagram for natural gas mixture.

Conclusion

Liquid-vapor (L-V) and solid-vapor (S-V) regions for high CO_2 content natural gas have been identified at cryogenic temperatures and high pressure conditions for two natural gas feed streams containing 60% and 70% CO_2 content respectively along with heavier hydrocarbons. The feed streams constituted hydrocarbon content ranging from methane to pentane. The temperature range for liquid-vapor domain extends from 0 °C to -60 °C with the pressure varying from 1 to 40 bars. At temperature of below -60 °C to -120 °C, the region is identified as solid-vapor region.

Acknowledgement

The authors would like to acknowledge University Technology PETRONAS for providing the necessary facilities and financial support for this research project work.

How to cite this article: Ali Q, Hilmi M, Suriati S, Azmi M S and B Lal. Phase Identification of Natural Gas System with High CO_2 Content for Cryogenic Based Separation Process. Recent Adv Petrochem Sci. 2017;3(1): 555603. DOI: 10.19080/RAPSCI.2017.03.555603.

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