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Synthesis and mixed integer programming based optimization of cryogenic packed bed pipeline network for purification of natural gas

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ABSTRACT

The variation of temperature, pressure and composition during cryogenic purification of natural gas using cryogenic packed bed network significantly affects separation. The present work aims to optimize cryogenic packed bed network for maximum product purity with minimum hydrocarbon losses. The separation concept between components of natural gas is based on the difference in freezing points of individual components and their composition in the mixture. The overall process of cryogenic packed bed pipeline network consists of the initial cooling of each bed, the capture of individual component and regeneration of each bed. In previous studies, the process concept of cryogenic packed bed network was validated using experimentation but the optimization study was not carried out. The simulation results and thermodynamic behavior of the individual component in the mixture are included to further explain the process concept. The sensitivity study was carried out to investigate the effect of the temperature and the pressure of each packed bed. For sensitivity study, node-edge schemes were developed and discussed in details by changing operating conditions of packed beds. Based on the node-edge scheme analysis, the optimum values for both temperature and pressures were selected for maximum separation and minimum methane loss. The energy requirements for both dehydration and CO_2 removal were calculated experimentally and compared with simulation results. For the simulation results, Aspen Hysys simulator was used along with Peng-Robinson fluid package. The energy requirements for both simulation and experimental results were calculated at atmospheric and high pressures. Both local and global optima were calculated using golden section search and depth-first strategy. The initial local optima result showed that the product purity can reach up to 77% methane, further optimization using depth-first strategy showed the product purity up to 94% with reduction of hydrocarbon losses from 39 to 16%.

1. Introduction

Natural gas is a fossil fuel, which has emerged as one of the cleanest, safest and most useful sources of energy in the world. The worldwide consumption of natural gas is expected to increase 1.6% annually till 2030. Raw natural gas consists largely of methane along with impurities such as water, carbon dioxide, and other higher hydrocarbons. Though several impurities are present in Malaysian natural gas, the major issue is the high content of CO_2 (up to 80%) (Darman and Harum, 2006).

The technologies of CO_2 separation mainly comprise of absorption, adsorption, membranes separation and cryogenic separation (Shimekit and Mukhtar, 2012). Chemical solvents based absorption is presently the most developed method used by industry. A comparative study of separation technologies for processing carbon dioxide rich natural gas in ultra-deepwater oil fields has been presented by Araújo et al. (2017). This work evaluates carbon dioxide separation alternatives for the challenging scenario in terms of technical, economic and environmental aspects. Reis et al. (2017) presented a hybrid processing scheme of optimized membrane skid with chemical absorption for the high content carbon dioxide natural gas. However, these processes are yet to be

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developed and tested to handle high CO_2 concentration in natural gas.

Cryogenic separation technology for CO_2 capture has been researched for several decades. However, for the past few years, this technology was not extensively studied due to the high expected cooling cost. In the cryogenic separation the operating temperature is usually very low which is needed to be optimized very sensibly to minimize the energy requirements and to make it good competitor with other separation technologies. In contrast, cryogenic separation does not require absorbents, and solid or liquid CO_2 can be produced directly at high pressure. With these advantages, the research on cryogenic CO_2 capture technology has made significant progress.

A detailed cryogenic processes classification based on desublimation and liquification has been presented in previous work (Maqsood et al., 2014b). The cryogenic processes were classified as conventional, non-conventional and hybrid cryogenic technologies. Northrup and Valencia (Northrop and Valencia, 2009) developed CFZ technology by modifying internal design of existing cryogenic distillation columns. Feed streams containing CO_2 from 8 to 71% have been processed below the targeted 2% pipeline quality standard. However, presence of small amount of water can cause plugging due to ice formation was the main disadvantage. In addition, the accumulation of solid CO_2 or ice on to heat exchanging surfaces causes a decrease in the heat transfer rate and overall process efficiency. Additional molecular sieve dehydration unit adds substantial cost and volume to the platform based operation. Cryocell process was proposed and tested by Hart et al. (Hart and Gnanendran, 2009). The operating conditions were maintained such that the vapor phase becomes rich in CH_4 whereas the liquid phase becomes rich in CO_2. The available literature unfortunately does not provide any details on modeling, freezing kinetics or other fundamental specifications due to commercial reasons. A process concept was developed on the basis of semi-cryogenic distillation along with mechanical centrifugation by Willems et al. (2010). Operation difficulties were present in the work regarding the formation of droplets of required sizes.

The non-conventional desublimation based packed bed was first studied by Tuinier et al. (2010). They developed a novel cryogenic process using dynamically operated packed beds to capture CO_2 and water from flue gases containing 10 vol % CO_2 and 1 vol % H_2O (Tuinier et al., 2011). The limitations of this process are that it was yet to be tested on natural gas at high CO_2 concentrations and pressure.

The cryogenic packed bed process concept for high pressure and CO_2 concentration was further experimentally validated by Abulhassan et al. (Ali et al., 2014a, b). It was concluded in this study that cryogenic packed bed is promising for high CO_2 content naturel gas due to low energy requirements. Energy requirement of cryogenic packed bed was reported to be significantly lower than the cryogenic distillation process, as the former requires 660–810 kJ/kg CO_2 energy while the later need 1472 kJ/kg CO_2 to purify natural gas with 69% of the CO_2 content. However, both cryogenic packed bed and cryogenic distillation process showed similar energy requirements with a feed with a lower concentration of CO_2. The desublimation process had been proven to be efficient for high CO_2 natural gas (Maqsood et al., 2014a). Furthermore, a detailed simulation study with experimental validation was reported using 1-dimensional pseudo homogeneous model (Ali et al., 2013) and nucleation study (Ali et al., 2014a, b).

Recently, a novel concept of desublimation based multiple packed bed pipeline network for purification of natural gas was presented by Abulhassan et al. (Ali et al., 2016) for possible offshore applications. A series of packed beds were used for simultaneous dehydration and CO_2 removal from natural gas. A novel strategy using multiple equilibrium temperature (*MET*) concept was also introduced for the simulation studies. The advantages of this cryogenic process are clean, efficient pipeline based separation with low capital cost. The detailed optimization study of multiple packed bed pipeline network was not included in the previous work.

Optimization is one of the most important engineering tools for addressing the issues related to increased cost of energy, global competition in product purity and increasingly stiff environmental regulations (Edgar and Himmelblau, 1988). The maximum utilization of the available resources for maximum profitability is essential for successful industrials operations (Basak et al., 2002).

The present study attempts to bridge the gap in existing research and literature on cryogenic packed beds by investigating the optimal operating conditions of cryogenic packed bed pipeline network to purify high CO_2 content natural gas at high pressures. The study also covers the separation of heavy hydrocarbon fractions with minimum methane losses. The separation behavior of methane, higher hydrocarbon and carbon dioxide at high pressure by using multiple equilibrium model *(MET)* is presented in this study during optimization.

2. Optimization study for cryogenic packed bed pipeline network

The optimization study comprises of the problem formulation and complete strategy to optimize the multiple packed bed pipeline network.

2.1. General schematics and process concept of cryogenic packed bed pipeline network

The basic separation principle is based on the difference of the desublimation and freezing point of each component in the mixture. The first step in the purification of the feed gas is dehydration, in which water contents were removed by using a series of beds at different initial temperatures. The cryogenic packed beds used for dehydration are the initial part of the complete schematics presented in Fig. 1. The temperature range of the beds varies from 0 °C to −30 °C. The complete study on dehydration and CO_2 removal using dual bed have been reported by Abulhassan et al. (Ali et al., 2016).

The feed continuously passes through a series of dehydration beds in order to achieve the desired pipeline specification i.e. 60–110 mg of water/MMSCF. The feed gas enters in first dehydration bed at comparatively high initial bed temperature as compared to the second or third bed. At very low initial bed temperature and high pressure, the hydrocarbon losses and energy requirements were found to be higher based on the simulation results. If the feed gas through first bed could not achieve the allowable water contents, then the second dehydration bed is inevitable which has a comparatively low initial bed temperature. For example, if the first bed is operating at −10 °C then the second bed would operate at −30 °C or below. The feed gas pressure also drops in the first bed, so the gas entering in the second bed has a lower pressure. This decrease in pressure also allows the operation of the next bed at a lower temperature because of the shift of freezing point of the natural gas components. The lower initial bed temperature allows removing the maximum remaining quantity of the water contents. After removal of the water contents, the dry gas enters in the next bed where CO_2 removes from the gas.

The beds used for CO_2 removal operates on the same principle as discussed earlier. The temperature range of CO_2 removal varies from −50 °C to −110 °C based on the feed pressure in order to minimize hydrocarbon loss and maximize the separation. The