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Refrigeration cycles in low-temperature distillation processes for the purification of natural gas

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ABSTRACT

The increasing energy demand has made low-quality natural gas reserves worthy of consideration for exploitation. As a consequence, industries have developed new process solutions in order to exploit these gas reservoirs in a profitable way. Most of these solutions are natural gas purification processes by distillation at low-temperature, involving or not solid CO_2 formation. Due to the low-temperatures reached in this type of processes, the choice of the appropriate refrigeration cycle becomes of paramount importance for limiting their energy consumptions and, thus, their operating costs. The aim of this work is to compare the performances of different types of refrigeration cycles using the coefficient of performance (COP) as discriminating factor. Several compounds (such as nitrogen, light hydrocarbons and ethylene) and their mixtures have been considered as working fluids and both non-cascade and cascade systems have been taken into account. Simulations by means of Aspen Hysys® V7.3 have led to conclude that the propane-ethylene cascade refrigeration cycle allows to attain the best performances.

1. Introduction

Projections on global energy trends show that the energy demand is expected to grow rapidly in the next twenty years. In this scenario, primary energy consumption is predicted to rise by 41% within 2035, with a great contribution coming from growing emerging economies. Among fossil fuels, natural gas is expected to have the most rapid growth (BP, 2014).

Data reported in open literature prove the existence of many gas resources which are contaminated with significant amounts of hydrogen sulfide and carbon dioxide (Northrop and Valencia, 2009). Therefore, industries have to find technologies that allow the exploitation of these fields in a profitable way.

Low-temperature processes are preferred to traditional chemical or physical absorption for gas purification when the carbon dioxide concentration in natural gas streams is high. Examples of applications of low-temperature gas purification by distillation to the natural gas industry (Fig. 1a–c) are the CFZ™ process (Haut et al., 1989; Parker et al., 2011; Northrop and Valencia, 2009; Valencia and Denton, 1985; Valencia and Victory, 1990, 1993), the Ryan-Holmes process (Holmes et al., 1983, Holmes and Ryan 1982a, b) and, more recently, a new process based on dual pressure distillation (Pellegrini, 2014; Pellegrini et al., 2015; Langè et al., 2015). Such low-temperature technology can be also applied to biogas upgrading (Pellegrini, 2014) and syngas purification (Berstad et al., 2011, 2013). In recent years, the interest in low-temperature processes for carbon dioxide removal has increased.

In this kind of processes, the refrigeration section is fundamental and attention must be devoted to its design. The system is held at the low temperatures which are characteristic of these processes by means of refrigeration cycles. These cyclical processes employed for refrigeration are typically operated by consuming mechanical or electric power for driving compressor refrigerating machines (Haaf and Henrici, 2003). The performances of a chilling cycle are evaluated in terms of the coefficient of performance (COP), which gives the ratio between the refrigerating effect and the net electrical or mechanical power supplied. Therefore, optimal solutions with high coefficients of performance are mandatory to save energy and reduce operating costs.

Several technologies are available to produce cooling duties and several compounds can be employed as working fluids in refrigeration cycles, either as pure or as mixed refrigerants. Refrigerants can be either natural (e.g., hydrocarbons, ammonia, carbon dioxide, etc.) or synthetic (e.g., chlorofluorocarbons, hydrochlorofluorocarbons, hydrofluorocarbons). However, the Montreal

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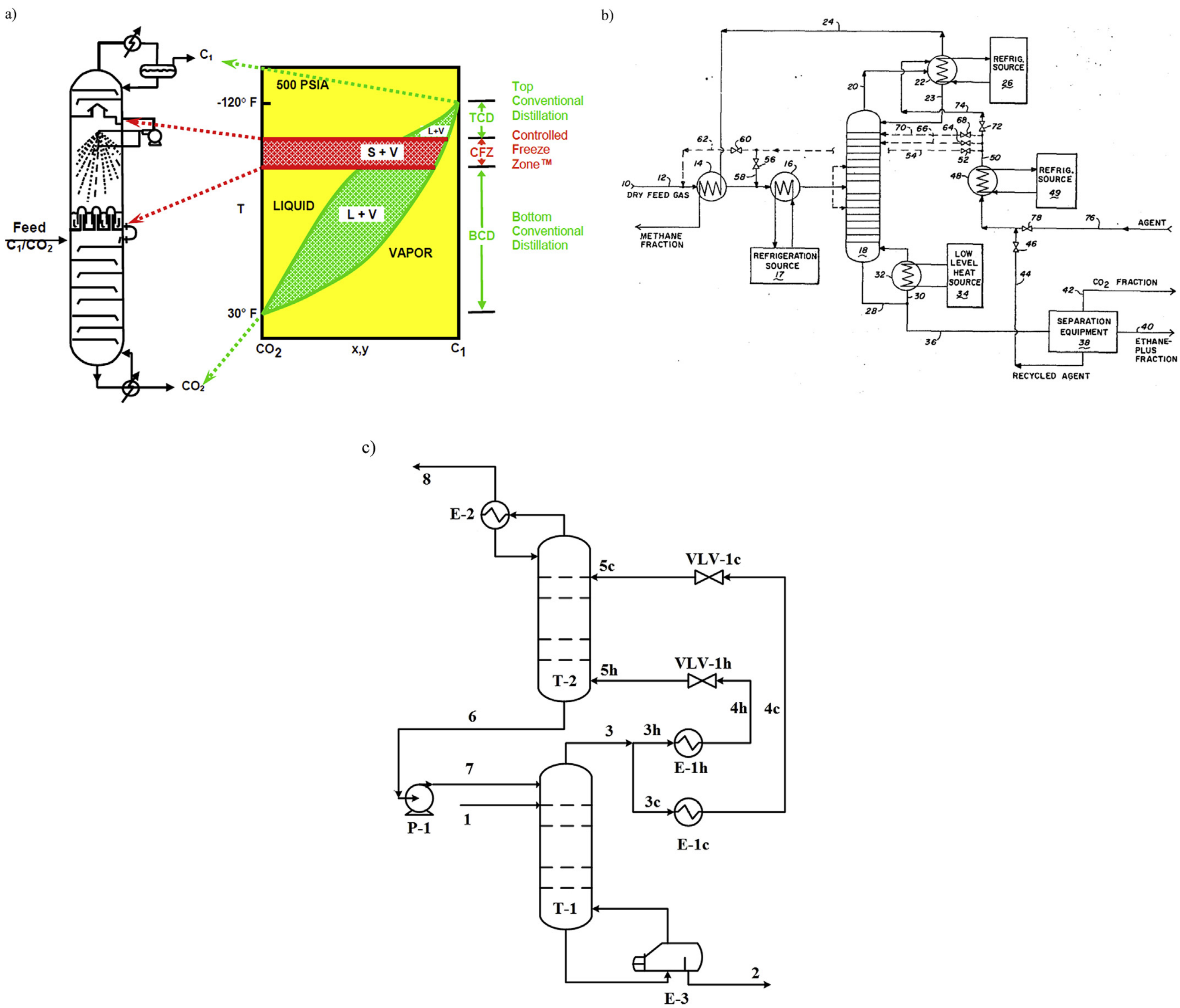

Fig. 1. Process flow diagrams for a) the CFZ™ Process (Valencia et al., 2008); b) the Ryan Holmes Process (Holmes and Ryan, 1982a); c) the dual pressure low-temperature distillation process (Pellegrini, 2014).

Protocol on Substances that Deplete the Ozone Layer has limited the use of synthetic refrigerants for environmental reasons. As a result, natural refrigerants have received a greater attention in the refrigeration industry. The choice of the most appropriate refrigerant is based on the operating conditions. Ethylene, methane, propane and ethane are used as pure fluids in several low-temperature applications, such as LNG production (Zhang and Xu, 2011; Kanoğlu, 2002; Lee et al., 2002) or the cold box section of hydrocarbons steam cracking plants (Fabrega et al., 2010). The most common application of refrigeration cycle for natural gas liquefaction is the *Phillips optimized cascade LNG process*, which consists of a three-loop cascade system that employs three pure refrigerants (typically propane, ethane or ethylene and methane) with different boiling temperatures in each loop. Also, mixed refrigerant can be used for low-temperature applications: mixtures of nitrogen and light hydrocarbons have been studied, for instance, for olefin separation (Mafi et al., 2009) and in several processes for LNG production (Wang et al., 2009).

In this work, different solutions to generate cold at temperatures around 173.15 K have been studied with the aim of defining a good solution for low-temperature upgrading processes of natural gas streams with high CO_2 content. In the open literature, no other works on the assessment of refrigeration technologies for low-temperature distillation, applied to the purification of natural gas, have been found. The work has been focused on the study of different configurations industrially used to generate cold in low-temperature processes, considering working fluids that are typically used in industrial applications for LNG production or air separation units. The studied technologies are mainly the Claude cycle (Barron, 2003), the vapor compression cycle and the cascade refrigeration cycle (Haaf and Henrici, 2003). Cascade refrigeration systems are particularly suitable for industrial applications when the evaporating temperatures are very low. For their application to low-temperature distillation processes, two different systems have been studied. The first one, which integrates the cold product of the distillation column, involves propane in the high-temperature loop