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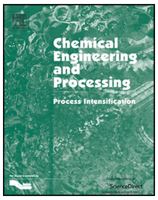

Optimal design of cryogenic distillation columns with side heat pumps for the propylene/propane separation

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ABSTRACT

Propylene is one of the most important products in the petrochemical industry, which is used as raw material for a wide variety of products. The propylene/propane separation is a very energy-intensive process because their boiling points are quite similar. In addition, at atmospheric conditions, their boiling points are −47.6 °C and −42.1 °C, respectively. To separate this mixture conventional columns which operate at high pressure and cryogenic distillation columns which operate at low pressure have been used, however, this approaches are still energy-intensive. This work presents energy-efficient and intensified distillation columns which are adiabatic such as the vapor recompression column (VRC) or diabatic such as columns with heat-integrated stages. A design and optimization procedure, which minimizes the energy consumption in the propylene/propane separation is presented. Conceptual design, superstructure representation, rigorous simulations and mathematical programming techniques are effectively combined to assess all the candidate distillation structures, refrigeration cycles, and heat integration possibilities simultaneously. Results showed that VRC and diabatic distillation columns with heat-integrated stages can reduce the energy consumption between 58 and 75% when compared with conventional distillation at high pressure. Furthermore, the proposed synthesis procedure derived simplified optimal distillation structures with few heat-integrated stages and still attained important energy savings.

1. Introduction

Propylene is used as intermediate in a wide range of chemical processes to produce fibers, foams, plastics, etc. It is mainly obtained as coproduct in the ethylene process and as byproduct in the petroleum refining industry, which in either case, it is mixed with other gases such as hydrogen, ethylene, ethane, propane, etc., but gases lighter than propylene are typically separated at an earlier stage, therefore, propylene is separated from propane in most cases. Since propylene and propane have close boiling points, and are gases at room temperature and atmospheric pressure, its separation is a very energy-intensive process.

Several separation alternatives based on solubility, diffusivity, and molecular size difference have been researched to alleviate the high energy consumption in the propylene–propane separation. Rege and Yang [1] proposed a four-step pressure swing adsorption (PSA) system, which used $AgNO_3/SiO_2$ and AlPO-14 for the separation of propylene from propane at two feed compositions, atmospheric and subatmospheric desorption pressures. Plaza et al. [2] proposed a five-step Vacuum swing adsorption (VSA) system, which used Cu-BTC because of its high pore volume and high sorption capacity, however, the results showed low recovery around 15%. Campo et al. [3] also adopted a five-step VSA system, but using X13 zeolite as sorbent. The results showed high recovery around 75% for their separation. Although membrane-based separations have been also proposed, most of them simultaneously lack high permeability and selectivity, having difficulties in scaling up, and lack of long-term stability due to poisoning of olefin-selective carriers. Pan et al. [4] studied zeolitic imidazolate frameworks for the separation of propylene/propane at several feed composition and temperature conditions. The results showed that the proposed membrane structure exhibited high permeability and selectivity for mixtures rich in propylene at temperatures lower than 22 °C.

Although the aforementioned alternatives can certainly attain energy savings, their capacity and operation time are rather constrained by equipment size and maintenance issues. To satisfy the demand of propylene, distillation is the predominant separation technology to obtain it at large scale. Intensified distillation-based

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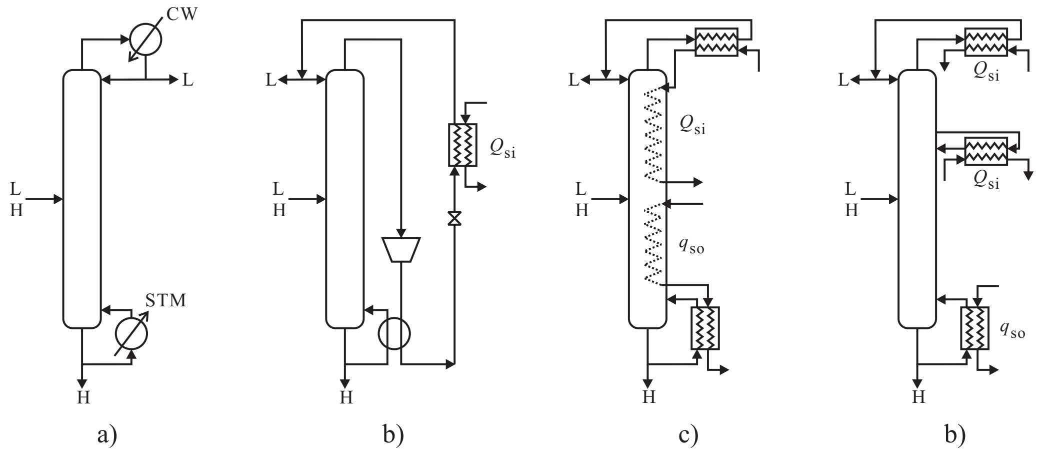

Fig. 1. Candidate distillation structures and heat integration techniques: (a) conventional distillation, (b) vapor recompression column, (c) diabatic distillation, and (d) distillation column with intermediate heat exchangers.

alternatives have been proposed to reduce energy consumption in distillation. Petterson et al. [5] reported profitable membrane-based distillation configurations where a membrane can be located close to the feed stream or at the bottom of the distillation column. Benali and Aydin [6] explored several membrane configurations which could be located at the top, middle and bottom of the distillation column. Their best reported configuration attained economic savings around 36% where one membrane at the top and one at the bottom were installed. Approaches based on mathematical programming have also been proposed [7,8], and they have shown that membrane distillation where the membrane is at an intermediate location can reduce the total annual cost up to 20%. Olujić et al. [9] studied two heat-integrated distillation columns: the vapor recompression column (VRC) and the heat-integrated distillation column (HIDiC). VRC and HIDiC were compared in terms of cost and energy consumption, in which the VRC had higher electricity consumption than the HIDiC, thus higher equipment cost since the compressor cost is predominant in this type of heat-pump assisted distillation structures. In addition, the HIDiC structure exhibited economic and energy savings of 20 and 25%, respectively, over the VRC. Ho et al. [10] presented a dynamic simulation for the HIDiC structure studied by Olujić et al. [9], and they showed that the proposed control configuration can control the HIDiC very well under various disturbances.

Despite of the aforementioned intensified alternatives, the propylene/propane mixture has been conventionally separated by means of distillation columns which operate either at high pressure (e.g., >15 bar) to ensure the use of water as cooling medium or at low temperature (e.g. <5 °C) to use a refrigerant as cooling medium. Regardless of these options, Mauhar et al. [15] have shown that the energy consumption for the separation of propylene and propane is inevitably high if the column operates between 15.95 and 12 bar.

If the separation is carried at low temperature, it is referred to as cryogenic distillation, which requires a refrigerant to provide cooling at a temperature level lower than that at the condenser, thus a distillation column must be coupled with a refrigeration system. However, when refrigerants are used as cooling utilities at the condenser, its temperature range and cost per unit amount of energy exchanged have been set at predefined conditions and treated as parameters in the synthesis problem to derive optimal distillation structures [7,9,10]. This means that the optimization of operating conditions in the refrigeration systems has not been included. In this work the operating conditions of the refrigeration system are optimized simultaneously with the distillation column in order to minimize not only the energy consumption in the latter but also in the former.

Heat integration through intermediate heat exchangers has been reported recently as an alternative to reduce the energy consumption and cost of distillation columns. It has been applied in reactive distillation systems with vapor recompression [11], heat integrated columns for the separation of binary [12,13] and ternary [14] mixtures.

The aim of this work is to propose a systematic procedure to solve the synthesis problem for the separation of the propylene/propane mixture at mild pressure and refrigeration conditions through cryogenic distillation, which embeds a refrigeration system to realize heat integration not only at the condenser or reboiler, but also at several locations in a distillation column. Therefore, the temperature levels of heat sources and heat sinks in the refrigeration system are treated as parameters, and their amount of energy exchanged at any location in the column are treated as optimization variables and embedded in the synthesis problem to find optimal cryogenic distillation columns.

2. Problem statement

Given a liquid mixture of components with low boiling point which is subject to cryogenic distillation, the structure and operating conditions of distillation columns (i.e. process side) as well as the structure and the operating conditions of refrigeration cycles (i.e. refrigeration side) are optimized simultaneously.

Fig. 1 summarizes the distillation columns subject to this research, which comprises conventional distillation columns (CC) in Fig. 1a, VRC in Fig. 1b, diabatic distillation columns (DCC) in Fig. 1c, and distillation columns with heat-integrated stages through the installation of heat pumps (HPC) in Fig. 1d. In DDC all the stages are heat-integrated because heat is supplied or removed in them. In the figure, L denotes the light component, and H the heavy component. CW denotes cooling water and STM steam. q_{so} (Q_{si}) stands for heat source (sink) in the refrigeration side. The arrows without any label entering and leaving heat exchangers denote a heat medium other than cooling water or steam.

Typically in cryogenic distillation, the stages in the column are not heat integrated, thus a refrigeration system is coupled to provide cooling at the condenser, which is at the lowest possible temperature; and to supply heating at the reboiler, which is at the highest possible temperature. Therefore, in the refrigeration side, the temperature difference must be larger than that between condenser and reboiler, which results in a large compressor work duty. Fig. 1c and d exploits the idea of removing heat at locations where the temperature is higher than that at the condenser and supplying it at locations where the temperature is lower than that at the