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Graphical method for identifying the optimal purification process of hydrogen systems

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ABSTRACT

Introducing purification devices into hydrogen systems can enhance the extent of hydrogen reuse. However, the economic performance of a purification device depends on its appropriate placement within a hydrogen system. Based on some established graphical methods, this paper explores the influences of the feed concentration on the purification process. A simple and systematic graphical method is proposed for identifying the OPP (optimal purification process) by extending the well-known pinch technology method. The proposed method can determine the OPP with the minimum feed flow rate and minimum tail gas flow rate under the condition of maximizing the HUS (hydrogen utility savings). The corresponding feed streams of the OPP also can be identified easily in the purification polygon. Furthermore, the conception of minimum separation work is used to compare different purification processes. A realistic case study is used to illustrate the applicability of the proposed method. Three different scenarios are analyzed and the results show that notable reductions in the minimum separation work consumption can be achieved (22%, 34% and 16% for the three scenarios).

1. Introduction

Hydrogen is an essential resource in refineries and is widely used in many processes, such as hydrotreating, hydrocracking and catalytic reforming, to produce high quality and clean oil products. However, along with the increasingly strict environmental regulations, demands for increasingly high quality fuels, and use of high-sulfur crude oil and heavier oil, refineries are compelled to process crude oil more deeply, causing their hydrogen consumption to rise sharply [1]. Hence, the supply and effective utilization of hydrogen has become a priority in many refineries [2].

In the past two decades, several notable studies have been carried out on the optimization of hydrogen distribution systems in refineries. Pinch technology is a powerful tool for process integration and has been widely used for heat recovery [3], water systems [4] and hydrogen systems [5]. Alves and Towler [5] applied the pinch technology concept in hydrogen distribution systems and proposed a pinch method for identifying the existence of bottlenecks and targeting the minimum flow rate of fresh hydrogen. Based on this method, Zhao et al. [6] put forward an integration methodology for hydrogen distribution systems with multiple impurities. El-Halwagi et al. [7] proposed a graphical method for determining the minimum utility consumption of a mass exchange network. However, in this method, the utility must be fresh. Subsequently, Almutlaq et al. [8] extended the method of El-Halwagi et al. [7] by applying it to systems with any utility concentration. By constructing the source and sink composite curves in the pure hydrogen load versus flow rate diagram, Zhao et al. [9] came up with a simple graphical method for determining the minimum hydrogen demand, which is similar to the method of Almutlaq et al. [8] as it also can be used in hydrogen systems with any utility concentration. Agrawal and Shenoy [10] used the NNA (nearest neighbors algorithm) to design hydrogen networks. Zhang et al. [11] generalized the NNA to hydrogen distribution systems with multiple impurities and presented a graphical approach for simultaneous targeting and design of resource conservation networks with multiple contaminants.

Mathematical programming models have also been used for the optimization of hydrogen networks. For example, Kumar et al. [12] developed a variety of mathematical models for optimum distribution of hydrogen including linear programming, nonlinear programming, mixed-integer linear programming and mixed-integer nonlinear programming models. Jia and Zhang [13] created an improved model for multi-component optimization for refinery

Abbreviations: HUS, hydrogen utility savings; OPP, optimal purification process; FSP, feed starting point; FEP, feed end point.

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hydrogen networks. Wu et al. [14] optimized refinery hydrogen networks considering the number of compressors.

As an economical and effective technology, purification has been widely used in hydrogen distribution systems. Alves [15] discussed three possible placements (above the pinch, across the pinch and below the pinch) for a purification unit in a hydrogen distribution system and found that placing purification cross the pinch was the best choice. Liu and Zhang [16] proposed a systematic methodology to select appropriate purifiers and built a superstructure model to include possible purification scenarios. Liao et al. [17] proposed a systematic approach for the retrofit design of hydrogen networks with purifiers. Subsequently, Liao et al. constructed a rigorous mathematical method for optimizing hydrogen systems both with no purification unit [18] and with one purification unit [19].

Based on the pinch approaches, Zhang et al. [20] considered the use of purification in hydrogen distribution systems, presenting a graphical method for the integration of hydrogen distribution systems with purification reuse. Subsequently, Lou et al. [21] incorporated algebraic equations into the graphical method of Zhang et al. [20], creating somewhat complicated models that can be used to find the initial location of purifiers and improve the composite curves shifting procedure. Nevertheless, in this method, the feed of the purification process is restricted to the hydrogen sources with the lowest concentration, which precludes the possibility of optimizing the purification process. Based on the method of Zhang et al. [20], Yang et al. [22] considered the constraint of feed concentration and proposed an improved graphical method for analyzing purification processes with any feed concentration. This improved graphical method is able to handle the constraints of concentration and flow rate of a purification process when targeting the maximum HUS (hydrogen utility savings). Liu et al. presented systematic methods for targeting the maximum purification feed flow rate [23] and the optimal purification feed flow rate [24] of hydrogen networks with purification reuse/recycle, respectively. However, these graphical methods can only optimize the purification process with specified purification feed, purified product, and hydrogen recovery. Another drawback is that the possible use of the tail gas from the purification device is not considered.

The main purpose of this work is to optimize the purification process in hydrogen systems by using a novel graphical method to identify the corresponding purification process based on pinch technology as well as previously published tools [20]. The proposed method can determine the maximum HUS and the minimum feed flow rate and tail gas flow rate of the purification process. The feed and tail gas flow rates of the purification process identified by the proposed method are much smaller than those obtained from the method of Zhang et al. [20]. It is noteworthy that the concentrations of the tail gas and the purified product are specified and the proposed method treats the tail gas as a hydrogen source with the lowest concentration.

2. Graphical methods for optimizing hydrogen systems with purification reuse

2.1. Purification process

Nowadays, commonly used purification technologies mainly include PSA (pressure swing adsorption) [25], membrane technology [26] and cryogenic process [27]. These purification processes rely on different separation principles and have different operating characteristics, as shown in Table 1.

A purification process can separate a feed stream into two streams, one is purified product and the other one is tail gas, as shown in Fig. 1.

Table 1
Selection guide for hydrogen purification process [16].

Factor	PSA	Membrane	Cryogenic
Feed purity/%	>40	>25	15–80
Maximum product purity/%	99.9+	98+	97
Maximum hydrogen recovery/%	Up to 90	Up to 95	Up to 98
Feed pressure/psig	150–1000	200–2000	200–1200
Product pressure/psig	Similar to feed	Much less than feed	Similar to feed
Flexibility	Very high	High	Average
Reliability	High	High	Average

The mass balances of a purification process can be described as follows [20]:

$$F_{\text{in}} = F_{\text{pur}} + F_{\text{tail}} \tag{1}$$

$$F_{\text{in}}C_{\text{in}} = F_{\text{pur}}C_{\text{pur}} + F_{\text{tail}}C_{\text{tail}} \tag{2}$$

where F and C represent the flow rate and hydrogen concentration, respectively; subscripts in, pur and tail denote the feed, purified product and tail gas, respectively.

2.2. Triangle rule of purification process

Based on the method of Zhao et al. [9], Zhang et al. [20] expressed Eqs. (1) and (2) in the hydrogen load versus flow rate diagram and proposed the triangle rule of purification process, as shown in Fig. 2a. When the feed is a mixture of several streams, the triangle rule also can be generalized to a polygon rule, as shown in Fig. 2b. For specified C_{pur} and C_{tail}, the corresponding F_{pur} and F_{tail} can be directly determined for any feed.

2.3. Graphical method of Zhang et al. [20]

In order to study hydrogen distribution systems with purification reuse systematically, Zhang et al. [20] combined the source and sink composite curves and the purification triangle in the hydrogen load versus flow rate diagram, proposing a novel graphical method for analyzing hydrogen distribution systems with purification reuse.

For a hydrogen distribution system without purification reuse, Fig. 3a shows how the source and sink composite curves can be depicted according to the method of Zhao et al. [9]. In the figure, P is the pinch point, AM represents the minimum hydrogen utility consumption and BW represents the discharged gas.

When introducing purification process into the hydrogen distribution system shown in Fig. 3a, for the case of $C_{\text{pur}} = C_{\text{utility}}$ (hydrogen concentration of utility), the method of Zhang et al. [20] for identifying the purification polygon is followed, as shown in Fig. 3b. Firstly, straight line BD with C_{pur} as its slope is constructed to represent the purified product, and similarly straight line GF with C_{tail} as its slope is constructed to represent the tail gas. Then, GF is shifted along the vertical direction until it intersects the sink composite curve and the whole sink composite curve lies above GF. For the case of $C_{\text{pur}} = C_{\text{utility}}$, the purified product can replace the

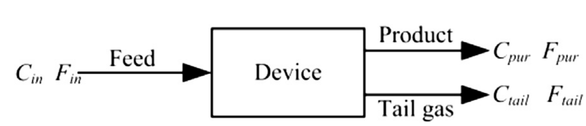

Fig. 1. Purification process.

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