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## The integration based method for identifying the variation trend of fresh hydrogen consumption and optimal purification feed

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#### ABSTRACT

Hydrogen purifier is widely used in refinery. In the view of hydrogen network integration, the Purification Feed Purity (PFP) and Purification Feed Flow Rate (PFFR) influence each other and should be optimized simultaneously. Based on the hydrogen network integration, a graphical method is developed for identifying the feasible purification feed region, the optimal purification feed and the variation trend of the fresh hydrogen consumption. With the quantitative relation between the limiting PFP and limiting PFFR analyzed, a diagram is built to identify the limiting line and feasible region of the purification feed. Furthermore, the relationship among the optimal PFP, optimal PFFR and the fresh hydrogen consumption is derived, and a two-dimensional diagram is developed to identify the variation trend line of the optimal PFP, the saved fresh hydrogen (or Hydrogen Utility Savings, HUS) and the pinch point along PFFR, as well as that of the optimal PFFR along PFP. Based on this, the optimal purification feed can be identified, as well as the maximum HUS. A case is studied to illustrate the applicability of the proposed method.

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#### 1. Introduction

Along with the increase of processing inferior crude oil, the improvement of environmental requirements and product standard, the hydro-processing ability increase rapidly in China. Hydroprocessing is the most attractive route for upgrading heavy crudes and residue, and aims to increase the H/C ratio in products and achieve higher yield. There are numerous hydro-processing technologies designed for specific objectives, such as hydrodesulphurisation (HDS), hydro-demetallization (HDM), hydrodenitrogenation (HDN), hydro-deasphaltenization (HDA) et al. In general, large amount of hydrogen is consumed by hydroprocessing to hydrogenate oil. Because of this, hydrogen is an important utility and is of significant concern of refiners. Decreasing the consumption of hydrogen utility, which is generally termed as fresh hydrogen in refinery, is an important way to increase the energy efficiency.

In the hydro-processing process, hydrogen is mainly consumed in reactors. To achieve higher conversion of the feed oil, an excess of hydrogen is generally used in these reactors. Unreacted hydrogen can be separated and reused. Besides, the hydrogen produced as

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http://dx.doi.org/10.1016/j.energy.2016.11.031 0360-5442/© 2016 Elsevier Ltd. All rights reserved. byproduct, such as that of the naphtha catalytic reformer, can be reused. All the hydrogen streams that can supply hydrogen (hydrogen source, including the fresh hydrogen) and that represent the hydrogen demand of the hydrogen consuming reactors (hydrogen sink) compose a hydrogen network. The fresh hydrogen consumption can be minimized with the match between sinks and sources optimized. This can be achieved by integrating the hydrogen network as a whole [1]. However, the amount of reuse is limited as the hydrogen purity of some sources is too low to be reused. To overcome this problem and decrease the fresh hydrogen consumption further, purification can be applied to upgrade the hydrogen purity of source streams.

Purification modifies the source flow rates and purity distribution of the hydrogen network. Its application is expected to increase the process hydrogen reuse and conserve fresh hydrogen. However, it is not always the case. The reason is that, the influence of purification changes significantly as its feed changes; sometimes the reverse results might be achieved if the purification feed is not appropriate. Hence, it is necessary to identify the variation trend of the fresh hydrogen consumption along the purification feed and optimize the purification feed. Target of the fresh hydrogen, which is the minimum fresh hydrogen consumption of the whole hydrogen network, can be applied to assess the influence of purification feed. This allows it to be screened quickly and conveniently, without actually having to carry out the network design.

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Alves et al. [1] developed the hydrogen surplus method for targeting the fresh hydrogen, compared the three possible placements of the purifier, and identified that the purifier across the pinch is the best choice. Thereafter, a unified pinch-based conceptual method [2] is developed based on the concept of limiting composite curve. This method is non-iterative and can be applied for targeting and designing both water networks and hydrogen networks with and without regeneration/purification. Foo et al. [3] established the Property Cascade Analysis (PCA) technique to set targets for material reuse networks via a tabular approach. Based on this, Ng et al. presented an optimization-based automated procedure to determine the minimum resource consumption/target(s) for a single-impurity resource conservation network (RCN) with direct material reuse/recycle [4], and extended this method to RCNs with waste-interception (regeneration) placement [5] and the property-based network [6]. Thereafter, they [7] developed an automated targeting technique to identify the minimum resource usage or total cost of a concentration- or property-based total resource conservation networks. This approach provides the same benefits as conventional pinch analysis techniques, and offers more advantages through its flexibility in setting an objective function for reuse/recycle and waste treatment networks. Zhang et al. developed a pinch-based graphical method for targeting the minimum fresh hydrogen consumption of the system with purification [8], and later extended this method with the separation performance of purifiers considered [9]. Yang et al. extended this method further to target the optimal and economical purification performance by considering the feed location and flow rate [10], and identify the optimal purification process [11]. Liu et al. studied the integration of hydrogen network with purification [12] and revealed the relationship between fresh hydrogen consumption and purification feed flow rate (PFFR) [13], as well as that between fresh hydrogen consumption and purification feed purity (PFP) [14], and that between fresh hydrogen consumption and hydrogen recovery [15]. Besides, it is verified that the limiting PFFR and limiting PFP exist; the purification beyond their limitation cannot benefit the fresh hydrogen consumption. By taking different types of purifier into consideration, Lou et al. [16] developed a pinch sliding graphical solution to target the hydrogen and water networks with purification/regeneration reuse.

There are two parameters determining the purification feed, PFFR and PFP. The optimal PFFP is affected by the PFP, and vice versa. To optimize purification feed, it is necessary to consider the variations of PFP and PFFR together. Although the above mentioned graphical-based methods can target the hydrogen network with purification reuse and identify the corresponding optimal PFFR or PFP. None of them consider the interaction between PFFR and PFP. Consequently, these methods cannot guarantee the identified fresh hydrogen target to be the minimal one.

In addition to graphical methods, the mathematical optimization method can also be used to synthesize the hydrogen systems with purification reuse/recycle. Based on the superstructure, Hallale and Liu [17] presented a mathematical approach to design the refinery hydrogen network. This method can handle pressure constraints and account for the existing equipment, and hence is suited for retrofitting real processes. Zhang et al. [18] developed a MINLP method for the overall refinery optimization through integrating the hydrogen network and the utility system with the material processing system. Van den Heever and Grossmann [19] proposed a multi-period mixed integer nonlinear programming (MINLP) model for the optimization of a hydrogen supply network consisting of five plants, four inter-connected pipelines and 20 customers. Khajehpour et al. [20] developed the reduced superstructure to get the feasible results faster and more justifiable. Tan et al. [21] put forward an optimization model for the synthesis of industrial water networks with regenerators. This model integrates a regenerator with a source-sink superstructure, and can be applied to solve the integration problems of the hydrogen network with purification reuse/recycle. Kumar et al. [22] developed a mathematical model for hydrogen networks accounting the complexity of real refinery system, such as pressure constraints, compressor flow rate recycle and flow combinations. Jia and Zhang [23] presented an improved model for hydrogen networks with the light hydrocarbon production and flash calculation incorporated and an optimization framework to solve the resulting NLP problem. Liao et al. [24] proposed a state-space superstructure based approach to incorporate all possible placements of existing compressors and purifiers. Later, they developed a mathematically rigorous systematic targeting approach with the pinch insight combined [25]. This method addresses both threshold and pinch problems, and is extended to hydrogen networks with purification reuse/recycle [26]. Kuo and Chang [27] developed a shortcut calculation method to calculate the inlet and outlet flow rates and purities of hydrogen users, and built a MINLP model for multi-period hydrogen network designs. Wang et al. [28] proposed a mathematical model to identify the optimal hydrogen network with the variation of hydrogen sink considered. Deng et al. proposed different superstructure based methods considering with the purifier, compressor and fuel system [29], the intermediate hydrogen header [30], and the interplant integration [31]. Wang et al. [32] developed an exergy-based method to optimize the energy consumption of the hydrogen network.

Umana et al. [33] presented an integrated approach for refinery process and hydrogen network design. In this method, empirical correlations are modified and adopted to predict hydrogen consumption in hydro-treaters; the light hydrocarbon yields in hydrodesulphurisation reactions are also predicted and integrated in the network model. Birjandi et al. [34] presented a new optimization method with the MINLP and NLP problems solved simultaneously. Lou et al. [35] introduced a robust optimization framework to optimize hydrogen network of refineries under uncertainty. These mathematical programming methods can optimize either the PFFR or PFP, and sometimes both. However, none of them can determine the variation trend of the fresh hydrogen consumption along the purification feed.

Based on the integration of hydrogen network, this paper aims to develop a systematic method for identifying the variation trend of the fresh hydrogen consumption and the optimal purification feed. Firstly, the characteristics of limiting purification feed is analyzed; the quantitative relationship between the limiting PFP and PFFP is derived, and the feasible region of purification feed is identified. Then, the quantitative relationship among the optimal PFP, optimal PFFR and the fresh hydrogen consumption is derived, the two-dimensional diagram is proposed to analyze the variation trend of fresh hydrogen target and the pinch point along the purification feed, and identify the optimal purification feed. A case study is presented to illustrate the targeting procedure.

## 2. Quantitative relation between purification and fresh hydrogen savings

The hydrogen purification technologies widely used in industries are pressure swing adsorption, membrane separation and cryogenic separation. No matter which method is used, the purification feed (with purity  $y_{pur}$ ) is split into purified product (with purity  $y_g$ ) and tail gas (with purity  $y_w$ ).

Hydrogen Utility Savings (HUS) is applied to evaluate the benefit of applying purification, and is defined as the decrement of fresh hydrogen consumption, which is resulted from the application of purification [13]. In the hydrogen flow rate versus purity profile [1],

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