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# Synthesis of hydrogen network with hydrogen header of intermediate purity

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## ABSTRACT

In order to simplify the network configuration and enhance the expandability and flexibility of the hydrogen network, one or two hydrogen utility headers are typically set with the consideration of requirement of hydrogen consumers. This paper proposed a superstructure-based mathematical programming model for the synthesis of hydrogen network with intermediate hydrogen header. The comprehensive superstructure is embedded with hydrogen utility, internal hydrogen sources and sinks, hydrogen headers, fuel system, compressors, purifiers and all the feasible interconnections between them. Two case studies are utilized to illustrate the feasibility and applicability of the proposed approach. The results show that the optimal flow rate of hydrogen utility will be decreased with the increase of the total number of connections as well as the increase of the number of hydrogen headers. The minimum flow rate of hydrogen utility for direct reuse/recycle without any intermediate hydrogen header can be achieved with the emplacement of two intermediate hydrogen headers. Besides, there is no direct connection among the hydrogen sources and hydrogen sinks. The Pareto front is made for the comparison on the flowrate of hydrogen utility and number of connections. The purification reuse/recycle scheme is investigated with the installation of purifier and the flowrate of hydrogen utility is reduced further.

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## Introduction

The amount of high-sulfur, heavy and inferior crude oil processed in industry has been increasing continuously. For

instance, Sinopec, a Chinese oil and gas company imported 70 Mt of high sulfur crude oil in 2010 and this corresponded to an yearly growth of 17% [1]. Concurrently, the government's environmental policy on sulfide and aromatics contents has

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led to higher standards of oil products for refineries. In order to improve the quality of oil products, refineries need to increase the loads of hydro-treating and hydrocracking, which require much fresh hydrogen. However, the production capacity of traditional continuous catalyst reforming, an important hydrogen producing process, is being reduced with the shrinking market demand for reforming products. The increasing deficit in hydrogen supply makes fresh hydrogen an increasingly expensive resource in refineries. Therefore, it is desirable for refineries to make efficient use of hydrogen. The important role of hydrogen management in synthetic crude oil refinery was first addressed by Simpson [2]. Nowadays, synthesis of hydrogen networks is widely accepted as a useful tool to improve hydrogen utilization in refineries. Generally, the methodologies developed for the synthesis and retrofit of refinery hydrogen networks can be classified into pinch analysis and optimization-based mathematical approaches.

Based on the analogy of pinch analysis for heat exchanger networks [3], Alves and Towler [4] proposed the hydrogen surplus diagram to identify the pinch and locate the minimum flow rate of hydrogen utility prior to detailed network design. Later, many other pinch-based approaches, such as material recovery pinch diagram [5] and its extensions [6–8], source composite curve [9], gas cascade analysis [10], material surplus composite curve [11] and extended limiting composite curve [12], have been developed to establish the targets for hydrogen networks. The cases with multiple resources [13], multiple impurities [7,14,15] and pressure constraints [16] have also been addressed. Recently, Liao et al. [17,18] deduced the optimal conditions for locating the targets for hydrogen networks without [17] and with one purification process [18], and developed a rigorous systematic targeting approach based on mathematical deduction. More recently, according to the characteristics of the pinch point, Liu et al. [19] developed a graphical method for identifying the upper bound of the purification feed flow rates. This technique was then extended to determine the optimal purification feed flow rates for hydrogen networks with purification reuse/recycle [20]. In addition, Yang et al. [21] introduced an iterative procedure for targeting and design of hydrogen networks involving purification reuse/recycle. A hydrogen network fulfilling the flow rate targets can then be obtained using the nearest neighbors algorithm [22], and the preliminary network can be evolved with the evolution strategies [23]. Although pinch analysis is useful in targeting the hydrogen network, it is not capable of handling multiple objectives as well as pressure and connection constraints. This calls for the development of various optimization-based mathematical techniques.

Hallale et al. [24] built up a superstructure with compressors and optimized it mathematically to maximize hydrogen recovery in the clean fuels production process. Later, many other mathematical programming approaches were developed. These include the automated targeting technique [25,26] and its modification [27], the reduced superstructure [28], overall refinery optimization [29,30], systematic methodology for selecting appropriate purifiers [31], multi-period optimization models [32,33], optimization under uncertainty [34,35]

and multiple operating scenarios [36], the state-space superstructure [37], superstructure-based formulation integrated with flash calculation [38], hydrogen sulfide removal [39] and unit models for fuel cells and steam reforming plants [40], multiple objectives [40,41], multiple components [38,42], and minimizing the total exergy consumption of the hydrogen utility and compressor work [43] and comparative analysis of different scenarios [44]. The retrofit of the hydrogen network for an existing refinery plant has also been addressed [27,37,45–47].

However, in these earlier works, hydrogen sources are directly connected to hydrogen sinks for reuse/recycle (with or without purification). The resulting network may thus be relatively complex, and any variations in hydrogen flowrate and purity at an upstream hydrogen unit (hydrogen source) will affect the downstream hydrogen sinks. In order to simplify the network configuration and enhance the controllability of the hydrogen network, hydrogen utility headers are commonly used. It should be noted that similar work has been conducted to simplify the piping network while facilitating the operation and control of water networks in large process plants. Feng and Seider [48] proposed a systematic approach for the design of water networks with internal mains, of which the concentrations are determined with pinch analysis. This approach was later extended for multi-contaminant water networks with a single internal water main [49] and involving regeneration reuse/recycle [50]. Ma et al. [51] introduced a rule-based design methodology for the design of water networks with internal mains. Zheng et al. [52] proposed an optimization-based approach for the design of multi-contaminant water networks with multiple internal mains. On the analogy of the water network, Zhang et al. [53] proposed a mathematical programming model for the optimization of intermediate pressure levels in hydrogen networks and addressed the trade-off between utility cost and complexity of the network. Later, Liang et al. [54] extended the previous model [53] by incorporating a central purification unit. However, in their work too many intermediate levels were used of which the pressures were set too high (10 MPa). Practically, at least one hydrogen header (i.e. hydrogen utility pipe) with a pressure typically around 2 MPa should be used in the hydrogen network. Jia [55] proposed a mathematical model including the mass balance for the hydrogen header. However, the influence of the placement of hydrogen headers on the target (i.e. the optimal flowrate of hydrogen utility) or network structure (i.e. the number of connections) has not well analyzed.

In this paper, a comprehensive superstructure-based mathematical programming model for the synthesis of hydrogen networks with intermediate hydrogen headers is proposed. The comprehensive superstructure incorporates hydrogen utility, internal hydrogen sources and sinks, hydrogen headers, the fuel system, compressors, purifiers and all feasible interconnections between them. The flowrate of hydrogen utility and the number of connections of the hydrogen network with up to two intermediate hydrogen headers are to be optimized. In addition, the optimal placement of compressors can also be determined using the proposed model. Two case studies are used to

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