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Synthesis of hydrogen network with hydrogen header of intermediate purity

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ARTICLE INFO

Article history:
Received 12 May 2014
Received in revised form
23 June 2014
Accepted 24 June 2014
Available online 19 July 2014

Keywords:
Hydrogen network synthesis
Process integration
Hydrogen header
Superstructure
Mathematical programming

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.

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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http://dx.doi.org/10.1016/j.ijhydene.2014.06.129

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 illustrate the feasibility and applicability of the proposed approach.

Problem statement

The problem can be expressed as follows. Given a set of hydrogen utilities with a total number of NHU, each hydrogen utility ($u \in NHU$) has its maximum capacity (FHU_u^{UB}), concentration of the cth component ($y_{u,c}$; $c \in NC$) and pressure (P_u). The outlet gas streams from hydrogen utilities, so-called external hydrogen sources, can only be allocated to hydrogen utility header ($h \in NH$). Typically, the outlet gas streams of continuous catalyst reforming, hydrocracking and hydro-treating are considered process hydrogen sources with a total number of NSR. Each process hydrogen source ($s \in NSR$) is characterized by its maximum flowrate (FSR_s^{UB}), concentration of the cth component ($y_{s,c}$; $c \in NC$) and pressure (P_s). With appropriate placement of gas compressors ($i \in NI$) for pressure lifting, hydrogen sources can be allocated to the hydrogen utility header and/or intermediate hydrogen headers ($h \in NH$) and the fuel system. Typically, the inlets of hydrocracking and hydro-treating are taken as process hydrogen sinks with a total number of NSK. Each sink ($k \in NSK$) has its own inlet gas flowrate requirement (FK_k^{in}), minimum inlet concentration limit for hydrogen ($y_{k,c}^{in}$, $\forall c = H_2$), maximum allowable inlet concentration of impurity ($y_{k,c}^{in}$) and pressure specification (P_k). Each sink can only receive hydrogen from hydrogen utility and intermediate hydrogen headers. In order to reduce the consumption of hydrogen utilities, a set of purifiers are placed to upgrade the hydrogen streams. The purifiers can only receive hydrogen streams from certain internal hydrogen sources. The product stream of a purifier will be allocated to hydrogen utility or intermediate headers and the residual stream is directed to the fuel system. The interconnections between hydrogen utilities, hydrogen consumers, purifiers, hydrogen headers and the fuel system are illustrated in Fig. 1. The objective is to optimize the hydrogen flow rates from hydrogen plants as well as the network complexity.

Mathematical model

The superstructure of the problem embedding potential configurations of interest is shown in Fig. 1. The mathematical formulations for the superstructure are presented as follows.

Formulations for hydrogen utilities

Hydrogen production processes, such as steam reforming, serve as hydrogen utilities in refinery plants. Other possible external hydrogen sources include ethylene plants, fertilizer plants and coal gasification sections. As the pressure of the hydrogen utility may not fulfill the pressure requirement of the hth hydrogen utility header, hydrogen compressors would be installed for pressure lifting. Typically, because of its high quality, the hydrogen utility will not be allocated to purifiers. In addition, the hydrogen utility is not allowed to be discharged to the fuel system.

As shown in Fig. S1, the gas stream from the uth hydrogen utility can be allocated to the hth hydrogen utility header or to the ith hydrogen compressor to lift its pressure to fulfill the pressure requirement of the hydrogen utility header.

The flowrate balance is made on the splitting node after the uth hydrogen utility (i.e. hydrogen plant),

$$FHU_u = \sum_{h \in NSH} FUH_{u,h} + \sum_{i \in NI} FUI_{u,i} \quad \forall u \in NHU \tag{1}$$

In addition, the distributed flowrate from the uth hydrogen utility should not exceed its maximum capacity,

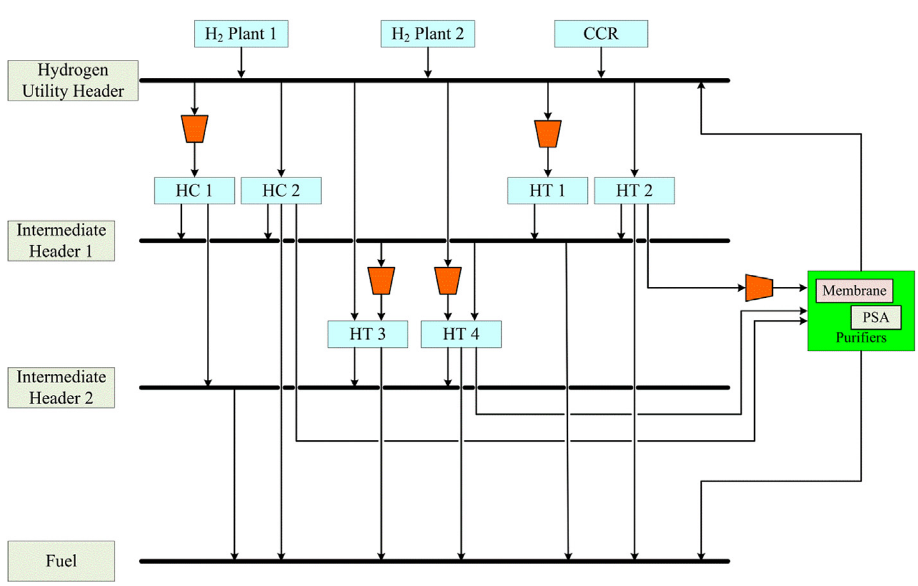

Fig. 1 – Superstructure of hydrogen network with hydrogen headers of intermediate purities.

$$FHU_u \leq FHU_u^{UB} \tag{2}$$

Formulations for process hydrogen sources

The continuous catalytic reforming (CCR) process is a typical hydrogen source in the refinery. In addition, the purges of hydro-treating and hydrocracking processes contain certain amount of hydrogen. They are categorized as process or internal hydrogen sources. As shown in Fig. S2, they can be directed to intermediate hydrogen headers, or allocated to purifiers (pressure swing absorption (PSA) or membrane) to upgrade the quality and then assigned to the hth hydrogen utility header. The compressors can be installed to lift the pressures of the hydrogen sources. The surplus hydrogen with low purity can be discharged to the fuel system.

The flow rate balance is made for the splitting node after the sth hydrogen source,

$$FSR_s = \sum_{h \in NSH} FSH_{s,h} + \sum_{p \in NP} FSP_{s,p} + \sum_{f \in NF} FSF_{s,f} + \sum_{i \in NI} FSI_{s,i} \quad \forall s \in NSR \tag{3}$$

It implies that the gas stream from the sth hydrogen source would be allocated to the hth hydrogen header, the pth purifier, the fth fuel system and/or the ith compressor.

Besides, the total flow rate for the sth hydrogen source cannot exceed its maximum capacity,

$$FSR_s \leq FSR_s^{UB} \tag{4}$$

Formulations for hydrogen headers

Typically, there exists a hydrogen utility header in each refinery plant. There will also be other hydrogen headers of intermediate hydrogen purities to simplify the hydrogen network configuration. As shown in Fig. S3, the inlet streams of hth hydrogen header can be allocated from uth hydrogen utility, sth hydrogen source, ith compressor and pth purifier. The outlet of hth hydrogen header can be directed to kth hydrogen source, ith compressor.

The flow rate balance is made for the mixing node before the hth hydrogen header,

$$FH_h^{in} = \sum_{u \in NFU} FUH_{u,h} + \sum_{s \in NSR} FSH_{s,h} + \sum_{s \in NP} FPH_{p,h}^{pord} \quad \forall h \in NH, \ c \in NC \tag{5}$$

It means that the inlet flowrate of the hth hydrogen header is the summation of all the allocated flowrate from the uth hydrogen utility, sth hydrogen source and the product of pth purifier.

The mass balance for cth component is made for the mixing node before the hth hydrogen header,

$$FH_h^{in} \cdot y_{h,c}^{in} = \sum_{u \in NFU} FUH_{u,h} \cdot y_{u,c} + \sum_{s \in NSR} FSH_{s,h} \cdot y_{s,c} + \sum_{s \in NP} FPH_{p,h}^{prod} \cdot y_{p,c}^{prod} \quad \forall h \in NH, \ c \in NC \tag{6}$$

The flow rate balance is made for the splitting node after the hth header,

$$FH_h^{out} = \sum_{k \in NSK} FHK_{h,k} + \sum_{i \in NI} FHI_{h,i} + \sum_{f \in NF} FHF_{h,f} \quad \forall h \in NH \tag{7}$$

It means that the outlet flowrate of the hth hydrogen header would be allocated to the hydrogen sink, the compressor and the fuel system. On the basis of practical consideration, the process hydrogen sources will be directly allocated to the purifier while the allocation from the hth hydrogen header to the purifiers is avoided.

The concentration of the cth component is assumed to be constant thus the outlet concentration of the cth component is equal to its inlet concentration.

$$y_{h,c}^{out} = y_{h,c}^{in} \quad \forall h \in NH \quad \forall c \in NC \tag{8}$$

Formulations for compressors

The inlet gas stream of the ith compressor can only be allocated from hydrogen sources and hydrogen header as shown in Fig. S4. Besides, its outlet would be directed to hydrogen sinks and purifiers.

The flow rate balance is made for the mixing node before the ith compressor,

$$FI_i^{in} = \sum_{h \in NH} FHI_{h,i} + \sum_{s \in NSR} FSI_{s,i} \quad \forall i \in NI \tag{9}$$

The mass balance for cth component is made on the mixing node before the ith compressor,

$$FI_i^{in} \cdot y_{i,c}^{in} = \sum_{h \in NH} FHI_{h,i} \cdot y_{h,c}^{out} + \sum_{s \in NSR} FSI_{s,i} \cdot y_{s,c} \quad \forall i \in NI, \ c \in NC \tag{10}$$

The flow rate balance is made around the outlet and inlet of the ith compressor,

$$FI_i^{out} = FI_i^{in} \quad \forall i \in NI \tag{11}$$

The concentration of cth component is assumed to be consistent, thus the outlet concentration of cth component is equal to its inlet concentration.

$$y_{i,c}^{out} = y_{i,c}^{in} \quad \forall i \in NI, \ c \in NC \tag{12}$$

The flow rate balance is made for the splitting node after the ith compressor,

$$FI_i^{out} = \sum_{k \in NSK} FIK_{i,k} + \sum_{p \in NP} FIP_{i,p} \quad \forall i \in NI \tag{13}$$

For the retrofit problem or design problem with several available compressors, the limitation for the maximum capacities for existing compressors should be taken into account,

$$FI_i^{in} \leq FI_i^{in,UB} \quad \forall i \in NI \tag{14}$$

For the grass-root design problem, the judgment on where the compressor should be placed is much more important and the modeling for the judgment of the placement of the compressors is presented later. Once the judgment of the placement of the compressors is determined, the Equations (9)–(13) will be utilized to calculate the inlet flowrate and purity after the optimization.

If there are several compressors are available for use in the grass-root design, Equations (9)–(14) can be included into the optimization model.

Formulations for hydrogen sinks

As shown in Fig. S5, the inlet gas stream for kth hydrogen sink would be supplied by hydrogen header and compressor.

The flow rate balance is made on the mixing node before the kth hydrogen sink,

$$FK_k^{in} = \sum_{u \in NHU} FHK_{h,k} + \sum_{i \in NI} FIK_{i,k} \quad \forall k \in NSK \tag{15}$$

It means that the inlet flowrate of the kth hydrogen sink is the summation of all the allocated flowrate from the hth hydrogen header and ith compressor.

Component mass balance for the mixing node before the kth hydrogen sink

$$FK_k^{in} \cdot y_{k,c}^{in} = \sum_{u \in NHU} FHK_{h,k} \cdot y_{h,c}^{out} + \sum_{i \in NI} FIK_{i,k} \cdot y_{i,c}^{out} \quad \forall k \in NSK \quad \forall c \in NC \tag{16}$$

Component concentration limitations for the kth hydrogen sink,

$$y_{k,c}^{in} \geq y_{k,c}^{\min} \quad \forall k \in NSK \quad \forall c = H_2 \tag{17}$$

$$y_{k,c}^{in} \leq y_{k,c}^{\max} \quad \forall k \in NSK \quad \forall c \in NC,\ c \neq H_2 \tag{18}$$

Formulations for the purification system

The schematic diagram for a typical purifier is illustrated in Fig. S6. Note that the inlet gas stream for the purifier would involve gas streams from hydrogen sources and compressors. Typically, the purifier (i.e. PSA or Membrane) has two outlets: product and residual streams. The product of higher hydrogen purity can be allocated to hydrogen headers or compressors if necessary. However, the residual with low hydrogen purity is assigned to fuel system.

The flow rate balance is made on the mixing node before pth purifier,

$$FP_p^{feed} = \sum_{s \in NSR} FSP_{s,p} + \sum_{i \in NI} FIP_{i,p} \quad \forall p \in NP \tag{19}$$

It means that the inlet flowrate of the purifier is the summation of all the allocated flowrate from the sth hydrogen source and ith compressor.

The mass balance for cth component can be made on the mixing node before pth purifier,

$$FP_p^{feed} \cdot y_{p,c}^{feed} = \sum_{s \in NSR} FSP_{s,p} \cdot y_{s,c} + \sum_{i \in NI} FIP_{i,p} \cdot y_{i,c}^{out} \quad \forall p \in NP,\ \ c \in NC \tag{20}$$

The flow rate and component mass balance are be made around the pth purifier,

$$FP_p^{feed} = FP_p^{prod} + FP_p^{resd} \quad \forall p \in NP \tag{21}$$

$$FP_p^{feed} \cdot y_{p,c}^{feed} = FP_p^{prod} \cdot y_{p,c}^{prod} + FP_p^{resd} \cdot y_{p,c}^{resd} \quad \forall p \in NP\ c \in NC \tag{22}$$

The cth component flow rate contained in the product over that contained in the feed of pth purifier is defined as recovery ratio ($RR_{p,c}$). Typically, the hydrogen recovery ratio is specified for each purifier.

$$FP_p^{prod} \cdot y_{p,c}^{prod} = RR_{p,c} \cdot FP_p^{feed} \cdot y_{p,c}^{feed} \quad \forall p \in NP\ \ c = H_2 \tag{23}$$

The flow rate balance is made for the splitting node after the PSA,

$$FP_p^{prod} = \sum_{h \in NH} FPH_{p,h}^{prod} + \sum_{i \in NI} FPI_{p,i}^{prod} \quad \forall p \in NP \tag{24}$$

$$FP_p^{resd} = \sum_{f \in NF} FPF_{p,f}^{resd} \quad \forall p \in NP \tag{25}$$

Formulations for the fuel system

The surplus internal hydrogen source, the residual gas stream of purifier would be sent to fuel system. The schematic diagram for the mixing node for the fuel system is illustrated in Fig. S7.

The flow rate and component mass balances are made on the mixing node before the fuel system,

$$FF_f^{in} = \sum_{s \in NSR} FSF_{s,f} + \sum_{h \in NH} FHF_{h,f} + \sum_{p \in NP} FPF_{p,f}^{resd} \quad \forall f \in NF \tag{26}$$

$$FF_f^{in} \cdot y_{f,c}^{in} = \sum_{s \in NSR} FSF_{s,f} \cdot y_{s,c} + \sum_{h \in NH} FHF_{h,f} \cdot y_{h,c}^{out} + \sum_{p \in NP} FPF_{p,f}^{resd} \cdot y_{p,c}^{resd} \quad \forall f \in NF,\ c \in NC \tag{27}$$

Note that, the inlet flow rate of fuel system (FF_f^{in}) and inlet concentration for cth component ($y_{f,c}^{in}$) are two variables and it makes the term ($FF_f^{in} \cdot y_{f,c}^{in}$) be a bilinear term. Since there are no concentration limits for the fuel system, Eq. (27) is not needed and can be left out from the model. However, this equation can be used after the optimization to calculate the inlet concentration to the fuel system.

Connection constraints

Binary variables $z_{a,b}$ are introduced to indicate if the connection exists between a supplier (hydrogen utility, hydrogen source, hydrogen header, outlet of compressor, product and residual of purifier) and a receiver (hydrogen sink, hydrogen header, inlet of compressor, inlet of purifier, fuel system). The necessary and sufficient condition for a connection to exist is that the flow rate is non-zero. The continuous flow rate variables are related with binary variables by Equation (28).

$$\begin{aligned} & F_{a,b} - z_{a,b} \cdot F_{a,b}^{UB} \leq 0 \\ & F_{a,b} - z_{a,b} \cdot F_{a,b}^{LB} \geq 0 \\ & z_{a,b} \in \left\{ \begin{array}{l} zUH_{u,h} \\ zSH_{s,h}, zSP_{s,p}, zSI_{s,i}, zSF_{s,f} \\ zPH_{p,h}^{prod}, zPF_{p,f}^{resd} \\ zIH_{i,h}, zIP_{i,p} \end{array} \right\} \\ & F_{a,b} \in \left\{ \begin{array}{l} FUH_{u,h} \\ FSH_{s,h}, FSP_{s,p}, FSI_{s,i}, FSF_{s,f} \\ FPH_{p,h}^{prod}, FPF_{p,f}^{resd} \\ FIH_{i,h}, FIP_{i,p} \end{array} \right\} \end{aligned} \tag{28}$$

where $F_{a,b}^{UB}$ and $F_{a,b}^{LB}$ are the upper and lower bounds for the flow rate variable $F_{a,b}$.

The total number of connections (N_{total}) is an important parameter associated with network complexity, given by the summation of all the connection variables as in Equation (29):

$$\begin{aligned}N_{total} = &\sum_u \sum_h zUH_{u,h} + \sum_s \sum_h zSH_{s,h} + \sum_s \sum_p zSP_{s,p} \\ &+ \sum_s \sum_i zSI_{s,i} + \sum_s \sum_f zSF_{s,f} + \sum_h \sum_k zHK_{s,h} \\ &+ \sum_h \sum_i zHI_{h,i} + \sum_h \sum_f zHF_{h,f} + \sum_p \sum_h zPH^{prod}_{p,h} \\ &+ \sum_p \sum_f zPF^{resd}_{p,f} + \sum_i \sum_k zIK_{i,k} + \sum_i \sum_p zIP_{i,p}\end{aligned} \tag{29}$$

Judgment of the placement of compressors

For grass-root design of hydrogen networks, the compressors should be placed between hydrogen sources and hydrogen sinks if necessary. The modeling technique for the judgment on the placement of compressors introduced by Liu and Zhang [31] is adopted here. Once the pressure level of a hydrogen sink or purifier is higher than that of a hydrogen source, the installation of hydrogen compressor is necessary. Once both the conditions that the flowrate between hydrogen suppliers (hydrogen utility, process hydrogen source and hydrogen header) and hydrogen receivers (process hydrogen sink, purifier, hydrogen header) is greater than zero and the pressure difference is less than zero are fulfilled, the hydrogen compressor must installed. An additional binary variable $z^{\Delta P}_{a,b}$ is introduced and it would be set to one if the pressure difference between a and b is less than zero and vice versa.

$$\begin{aligned}&z_{a,b} = 1 \text{ and } z^{\Delta P}_{a,b} = 1 \rightarrow z_{a,i} = 1 \text{ and } z_{i,b} = 1 \\ &z_{a,b} \in \left\{\begin{matrix} zUH_{u,h} \\ zSH_{s,h}, zSP_{s,p}, zSI_{s,i}, zSF_{s,f} \\ zPH^{prod}_{p,h}, zPF^{resd}_{p,f} \\ zIH_{i,h}, zIP_{i,p} \end{matrix}\right\}\end{aligned} \tag{30}$$

The binary variable $z^{\Delta P}_{a,b}$ is related with the pressure difference by Equation (31),

$$\begin{aligned}&P_b - P_a - z^{\Delta P}_{a,b} \cdot \Delta P^{UB} \le 0 \\ &P_b - P_a + \left(1 - z^{\Delta P}_{a,b}\right) \cdot \Delta P^{UB} \ge \Delta P^{LB} \\ &z_{a,b} \in \left\{\begin{matrix} zUH_{u,h} \\ zSH_{s,h}, zSP_{s,p}, zSI_{s,i}, zSF_{s,f} \\ zPH^{prod}_{p,h}, zPF^{resd}_{p,f} \\ zIH_{i,h}, zIP_{i,p} \end{matrix}\right\}\end{aligned} \tag{31}$$

Then Equation (30) can be reformulated as follows,

$$\begin{aligned}&z_{a,b} + z^{\Delta P}_{a,b} - z_{a,i} \le 1 \\ &z_{i,b} - z_{a,i} \ge 0 \\ &z_{a,b} \in \left\{\begin{matrix} zUH_{u,h} \\ zSH_{s,h}, zSP_{s,p}, zSI_{s,i}, zSF_{s,f} \\ zPH^{prod}_{p,h}, zPF^{resd}_{p,f} \\ zIH_{i,h}, zIP_{i,p} \end{matrix}\right\}\end{aligned} \tag{32}$$

The flow rate of hydrogen utility is strongly related to the capacity of hydrogen plant and its operating cost. Therefore, the objective function can be formulated to minimize the consumption of hydrogen utility.

$$\min FHU = \sum_{u \in NHU} FHU_u \tag{33}$$

In addition, the minimum total number of connections can be determined by setting its upper bound. The upper bound is reduced until the optimal flow rate of hydrogen utility increases. The critical number of connections is considered the minimum.

Three scenarios, i.e. Direct reuse/recycle, Purification reuse/recycle without/with the judgment of the placement of compressors are explored. The relationship between the minimum flow rate of hydrogen utility and number of connections is investigated.

***Scenario 1* – Direct reuse/recycle**

Objective function: Equation (33)

Subjected to Equations (1)–(8), (15)–(18), (26) and connection constraint (28).

Note that the equations and terms related to the compressors and purifiers are omitted. In addition, the component mass balance for the fuel system as shown in Equation (27) is neglected and the bilinear term ($FF^{in}_f \cdot y^{in}_{f,c}$) is removed from the model. The bilinear terms ($FH^{in}_h \cdot y^{in}_{h,c}$, $FHK_{u,k} \cdot y^{out}_{h,c}$) and the binary variables ($z_{a,b}$) are included in the model. It makes the model an MINLP.

***Scenario 2* – Direct reuse/recycle with the judgment of the placement of compressors**

Objective function: Equation (33)

Subjected to Equations (1)–(8), (15)–(18), (26) and connection constraint (28) and pressure constraints (31)–(32).

Compared with Scenario 1, the equations and terms related to the judgment of the placement of compressors are included.

***Scenario 3* – Purification reuse/recycle with the judgment of the placement of compressors**

Objective function: Equation (33)

Subjected to Equations (1)–(8), (15)–(26) and connection constraint (28) and pressure constraints (31)–(32). Compared with Scenario 1, the equations and terms related to the purifier and the judgment of the placement of compressors are included.

Case study

Case 1

A refinery hydrogen network for case 1 is taken from Alves and Towler [4] and the existing network is shown in Fig. 2. As shown, a certain extent of hydrogen integration is included in the existing network and the current external (fresh or imported) hydrogen consumption is reported as 277.2 mol/s. The original process data for this example are listed in Table 1. As shown, there are four hydrogen consuming processes, namely, hydrocracker unit (HCU), naphtha hydrotreater (NHT), cracked naphtha hydrotreater (CNHT), and diesel hydrotreater (DHT). The inlets for those hydrogen consumers are taken as internal hydrogen sinks and their outlets are considered to be internal hydrogen sources. In addition, a catalytic reforming unit (CRU) and a steam reforming unit (SRU) are two internal hydrogen producing facilities which are

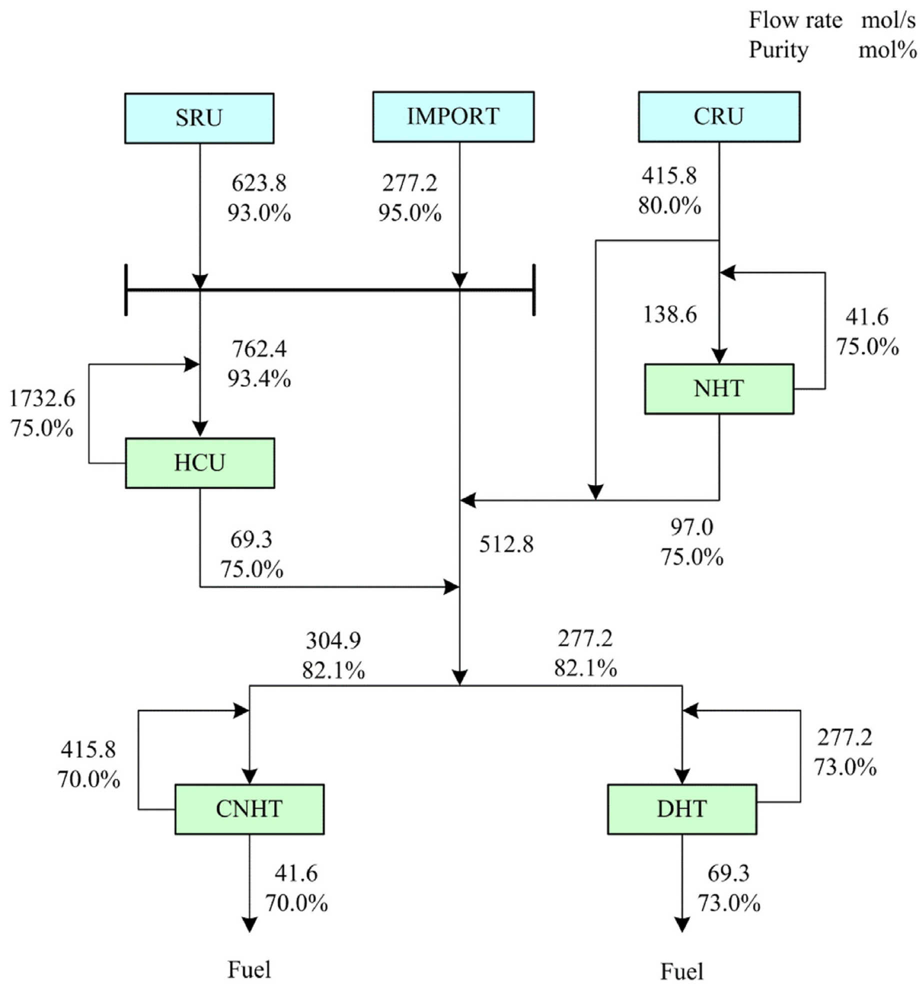

Fig. 2 – Existing refinery hydrogen network for Case 1 [4].

available in this plant. To minimize the operating cost, the internal hydrogen sources should be fully utilized before any purchase of external fresh hydrogen is considered. In this case, the imported hydrogen supply has a hydrogen purity content of 95 mol %.

Table 1 – Current process data for hydrogen consumers and sources [4].

Process	Make-up		Purge		Recycle
	Flow rate (mol/s)	Purity (mol% H_2)	Flow rate (mol/s)	Purity (mol% H_2)	Flow rate (mol/s)
HCU	762.4	93.36	69.3	75	1732.6
NHT	138.6	80	97	75	41.6
CNHT	304.9	82.14	41.6	70	415.8
DHT	277.2	82.14	69.3	73	277.2
Source	Flow rate			Purity (mol% H_2)	
	Maximum (mol/s)	Minimum (mol/s)	Current (mol/s)		
SRU	623.8	0	623.8	93	
CRU	415.8	415.8	415.8	80	
import	346.5	0	277.2	95	

The process data and extracted limiting data are listed in Table 2. As shown, the data in the last two rows of Table 2 specifies the flow rate and minimum purity requirement for hydrogen sinks. The data in the two columns on the right side of Table 2 indicates the maximum available flow rate and purity for hydrogen sinks.

To determine the minimum hydrogen utility as well as the minimum number of connections, the mathematical model for ***Scenario 1*** is utilized. The MINLP model is solved in the GAMS software [56] using DICOPT [57] as solver for MINLP, CPLEX [57] for MIP and KNITRO [57] for NLP (the PC specification: Intel D CPU 3.00 GHz, 4 GB RAM).

Firstly, only one hydrogen header so-called hydrogen utility header is placed. Once those limiting data in Table 2 is given, the model is solved in 0.01 CPUs and the fresh hydrogen from the import is determined as 506.611 mol/s. The total number of connections is determined as 12. The hydrogen purity for the hydrogen header is targeted as 80.61 mol%. Fig. 3 illustrates the optimal hydrogen network of Case 1 with one hydrogen header. The results are also shown in Table S1 in the Supplementary file.

Next, two hydrogen headers are installed. To keep the total number of connections as 12, the minimum flow rate of fresh

Table 2 – Limiting data for Case 1 [4].

Sources	Sinks					Current flow rate (mol/s)	Maximum flow rate (mol/s)	Purity (mol% H_2)
	HCU	NHT	CNHT	DHT	Fuel			
Import	138.6		72.6	66		277.2	346.5	95
SRU	623.8					623.8	623.8	93
CRU		138.6	145.2	132		415.8	415.8	80
HCU	1732.6		36.3	33		1801.9	1801.9	75
NHT		41.6	50.8	46.2		138.6	138.6	75
CNHT			415.8		41.6	457.4	457.4	70
DHT				277.2	69.3	346.5	346.5	73
Flow rate (mol/s)	2495	180.2	720.7	554.4	110.9			
Purity (mol% H_2)	80.61	78.85	75.14	77.57				

hydrogen from the import is determined as 457.400 mol/s. With the placement of one more intermediate hydrogen header, 9% of reduction for the minimum flow rate of fresh hydrogen is achieved. The hydrogen purities for two intermediate hydrogen headers are determined as 81.095 mol% and 75.769 mol%, respectively.

In order to reduce the minimum flow rate of fresh hydrogen from the import, the total number of connections would be increased. By setting the upper bounds for the total number of connections to be 13, 14, 15 and 16, the minimum flow rate of fresh hydrogen from the import is determined as 359.889 mol/s, 294.178 mol/s, 279.630 mol/s and 268.821 mol/s. The optimized results and the optimal hydrogen network of Case 1 with two hydrogen headers are shown Table S2 and Fig. S8 ($N_{total} = 12$), Table S3 and Fig. S9 ($N_{total} = 13$), Table S4 and Fig. S10 ($N_{total} = 14$), Table S5 and Fig. S11 ($N_{total} = 15$). Besides, Fig. 4 illustrates the optimal hydrogen network of Case 1 with two hydrogen headers ($N_{total} = 16$) and the optimized results are shown in Table S6. If the total number of connections increase further (greater than 16), the minimum flow rate of fresh hydrogen from the import is kept to be 268.821 mol/s, which agrees with that reported in the literature [4,5,10,23]. The comparison for minimum flow rates for hydrogen utility versus number of connections for Case 1 is illustrated in Table 3 and the Pareto front can be plotted (see Fig. 5) on the basis of the data shown in Table 3. Note that, the minimum flow rate of hydrogen utility (268.821 mol/s) is achieved when two hydrogen headers are installed. The judgment of the optimal flowrate of hydrogen utility and the complexity of the network (i.e. number of connections) can be made according the resulted Pareto. Although the same model and solving strategy are applied for the synthesis of hydrogen network with more than two hydrogen headers, the flow rate of hydrogen utility would not be reduced further for Case 1 while the number of connections will increase.

Case 2

Fig. 6 shows the current hydrogen network for case 2 which is adopted from the base case of Elkamel et al. [30]. As shown, the current fresh hydrogen consumption from the hydrogen plant is reported as 80 MMSCFD. The current process data for this example are listed in Table 4. As shown, there are five hydrogen consuming processes, namely, hydrocracker unit (HCU), gas oil hydrotreater (GOHT), residue hydrotreater (RHT), diesel hydrotreater (DHT), and naphtha hydrotreater (NHT). The inlets for those hydrogen consumers serve as internal hydrogen sinks and their outlets would be considered as internal hydrogen sources. The purity and pressure requirements for the sinks and the purity and pressure conditions for the sources are listed in Table 4. Note that all the hydrogen consumers have internal recycle compressors. The flow rates of internal recycle hydrogen streams would not be involved in either those of hydrogen sinks or sources. In addition, a catalytic reforming unit (CRU) is an internal hydrogen source with specified hydrogen purity of 80%, outlet pressure of 300 psi and maximum capacity of 14.5 MMSCFD. Its maximum capacity is assumed in this paper according to

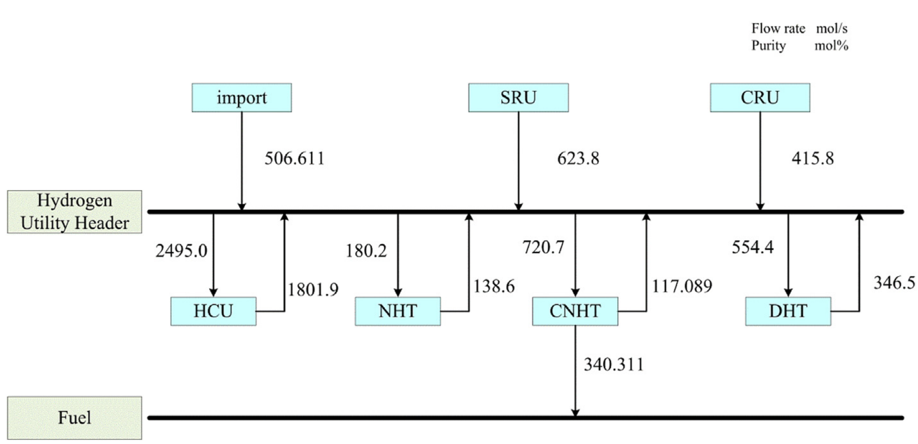

Fig. 3 – An optimal hydrogen network of Case 1 with one hydrogen header.

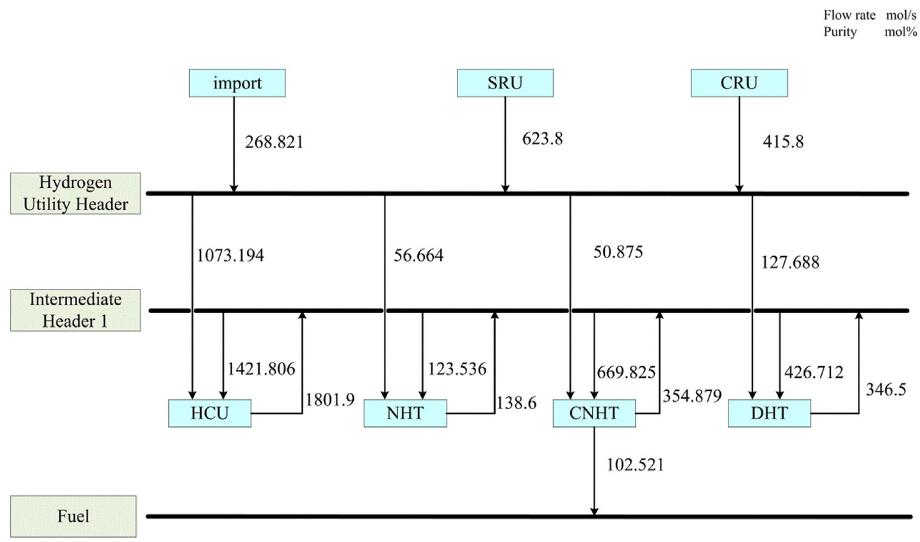

Fig. 4 – An optimal hydrogen network of Case 1 with two hydrogen headers (N_{total} = 16).

Table 3 – Comparison for flow rates of hydrogen utility verse number of connections for Case 1.

One hydrogen header		Two hydrogen headers	
No. of connections	Hydrogen utility flow rate (mol/s) Proposed model	No. of connections	Hydrogen utility flow rate (mol/s) Proposed model
12	506.611	12	457.400
		13	359.889
		14	294.178
		15	279.630
		16	268.821

the optimized results in the literature of Elkamel et al. [30]. In order to reduce the operating cost for hydrogen plant, the internal hydrogen sources should be fully utilized before the hydrogen utility from hydrogen plant is considered. In this case, the hydrogen utility is specified with a hydrogen purity content of 95 mol%, outlet pressure of 300 psi and maximum capacity of 80 MMSCFD. Besides, the fuel system operates at low pressure (200 psi) which would receive unused internal hydrogen sources. The case will be used to investigate the influences of the placement of compressors and the purifier on the consumption of hydrogen utility.

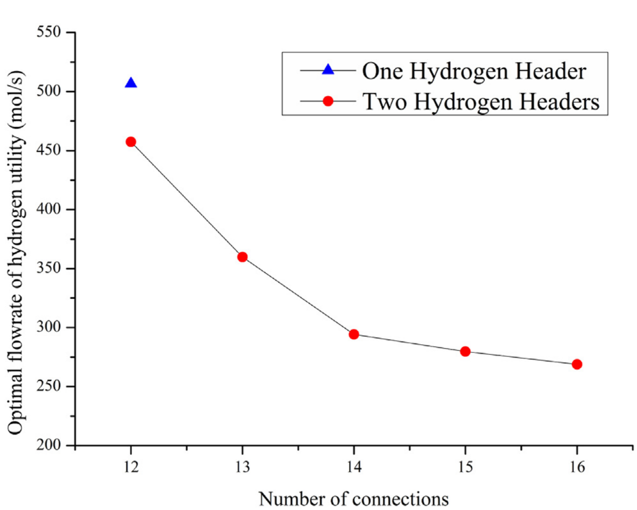

Fig. 5 – Pareto solution for the trade-off between the optimal hydrogen utility and number of connections.

In order to investigate the influence of placement of compressor on the hydrogen utility, the models without and with the consideration of placement of compressor will be solved separately. To determine the minimum hydrogen utility of hydrogen network for direct reuse/cycle without the placement of compressor, the mathematical model for Scenario 1 is utilized). It is solved in GAMS software [56] using DICOPT [57] as solver for MINLP, CPLEX [57] for MIP and KNITRO [57] for NLP (based on the PC specification: Intel D CPU 3.00 GHz, 4 GB RAM).

The extracted flow rates and purities for hydrogen sinks and maximum flow rate and purities for hydrogen sources are shown in Table 4 and they were used in the model. Based on the results of case 1, two hydrogen headers are set up. The best solution for the flow rate of hydrogen utility is found at 79.909 MMSCFD with the minimum number of connections of 15. The flow rate of hydrogen utility is same with that determined by hydrogen pinch. The optimized results are listed in Table S7 and Fig. 7 illustrates an optimal hydrogen network for Case 2 without the placement of compressors.

In addition, on the basis of the data listed in Table 4, the inlet pressure requirements for sinks HC, GOHT, RHT and DHT are 2000 psi, 500 psi, 600 psi and 500 psi. In order to reduce the

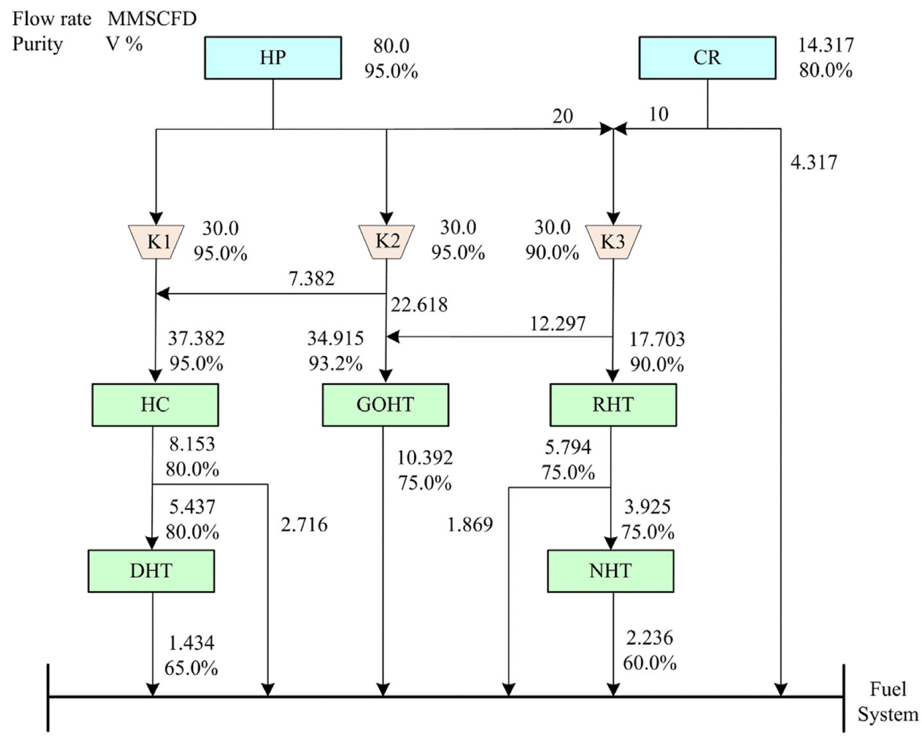

Fig. 6 – Current hydrogen network for Case 2 [30].

total number of compressors, compressors should not be set before the hydrogen header. Hence the pressure of the hydrogen headers are set to 300 psi. Hence it is necessary to install the compressors between the hydrogen headers and the hydrogen sinks.

To consider the placement of compressors, the mathematical model for ***Scenario 2*** is utilized. It is solved in GAMS software [56] using DICOPT [57] as solver for MINLP, CPLEX [57] for MIP and KNITRO [57] for NLP (based on the PC specification: Intel D CPU 3.00 GHz, 4 GB RAM).

The extracted flow rates, purities and pressures for hydrogen sinks and maximum flow rate, purities and pressures for hydrogen sources are shown in Table 4. Based on the result of case 1, two hydrogen headers are set up. The best solution for the flow rate of hydrogen utility is found at 79.909 MMSCFD with the minimum number of connections of 19. The optimized results are listed in Table S8 and Fig. S12 shows the optimized hydrogen network for direct reuse/recycle with the placement of compressors. As shown, the hydrogen headers (Header 1 and Header 2) are assigned to compressors and then allocated to hydrogen sinks HC, GOHT, RHT and DHT. However, the optimal results are close to the optimized results without the placement of compressors. It implies that the placement of compressors have nothing to do with the flow rate of the hydrogen utility. However, the total number of connections is increased due to the installation of compressors. In addition, the purifier would be installed to upgrade the quality of several internal hydrogen sources for further utilization.

In order to further reduce the consumption of hydrogen utility, the purification unit (i.e. PSA) is incorporated into the hydrogen network to upgrade the quality of internal hydrogen

Table 4 – Limiting data for Case 2 [30].

Sources	Sinks					Fuel	Current flow rate (MMSCFD)	Maximum flow rate (MMSCFD)	Purity (V% H_2)	Pressure (psi)
	HCU-K	GOHT-K	RHT-K	DHT-K	NHT-K					
HP	37.382	34.915	7.703				80	80	95	300
CRU			10			4.317	14.317	14.5	80	300
HCU				5.437		2.716	8.153	8.153	80	1200
GOHT						10.392	10.392	10.392	75	350
RHT					3.925	5.794	9.719	9.719	75	400
DHT						1.434	1.434	1.434	65	350
NHT						2.236	2.236	2.236	60	300
Flow rate (MMSCFD)	37.382	34.915	17.703	5.437	3.925	26.889				
Purity (V% H_2)	95.00	93.20	90.00	80.00	75.00					
Pressure (psi)	2000	500	600	500	300					

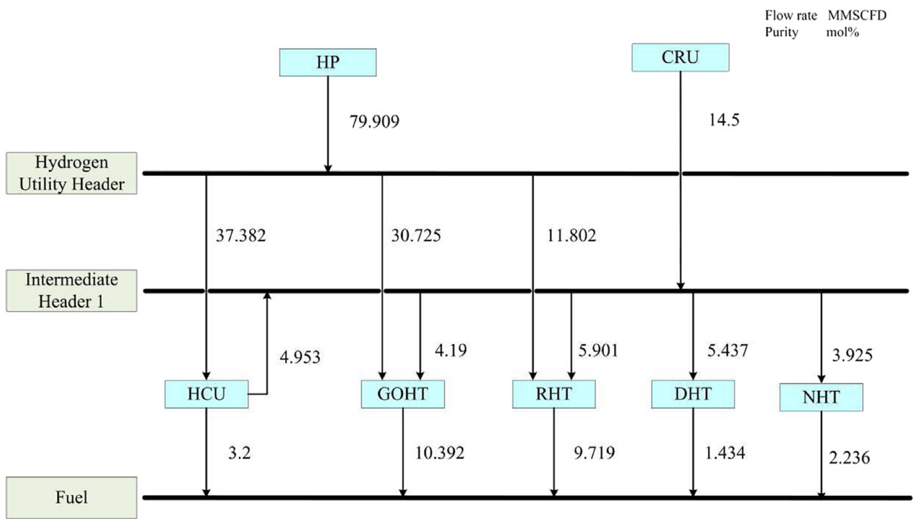

Fig. 7 – Optimal hydrogen network for Case 2 without the placement of compressors (Direct reuse/recycle).

sources and the product of the PSA will be assigned to hydrogen sinks if the pressure requirement is fulfilled. To model the placement of compressor and purifier, the mathematical model for ***Scenario* 3** is utilized. It is solved in GAMS software [56] using DICOPT [57] as solver for MINLP, CPLEX [57] for MIP and KNITRO [57] for NLP (based on the PC specification: Intel D CPU 3.00 GHz, 4 GB RAM).

The process data for the purification unit PSA are assumed in this paper. The inlet pressure for PSA is set as 300 psi. The pressure for the product of PSA is assumed to be 300 psi and the slight pressure drop is neglected. The hydrogen purity for its product is given as 95% and the hydrogen recovery is defined as 90%. Besides, the pressure for its residue is set as 200 psi. The maximum inlet flow rate for PSA is assumed to be 32 MMSCFD. The optimal flow rate of hydrogen utility is found at 61.4 MMSCFD with the minimum number of connections of 22. The optimized results are listed in Table S9 and Fig. 8 shows the optimized hydrogen network with the placement of compressors and purifier.

Conclusions

This paper proposed a superstructure-based optimization model for the synthesis of hydrogen networks with hydrogen headers. The comprehensive superstructure is embedded with hydrogen utility, internal hydrogen sources, hydrogen sinks, hydrogen headers, fuel system, compressors, purifiers and all the feasible interconnections between them. The comparative analysis is conducted for several scenarios:

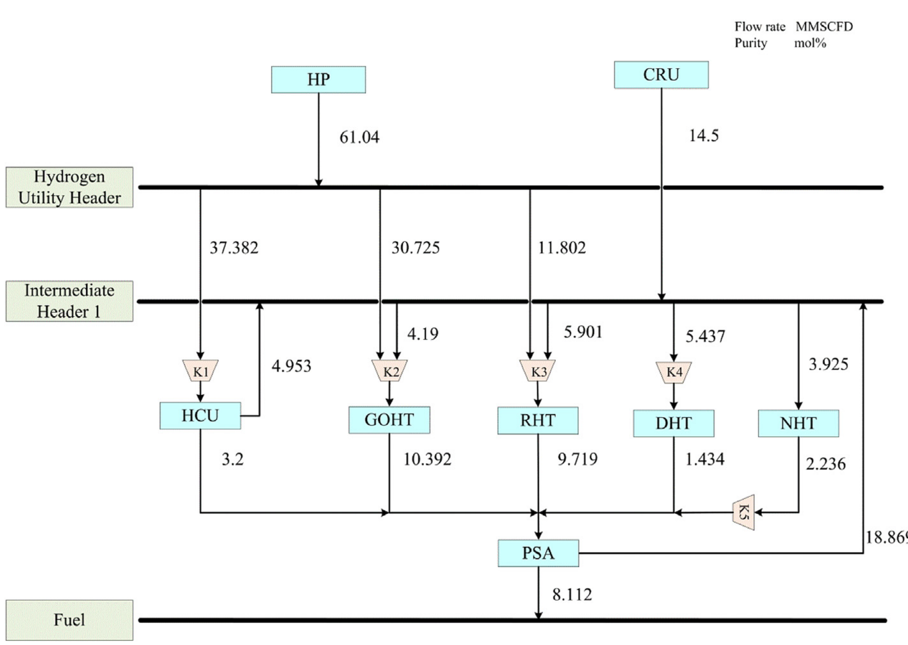

Fig. 8 – Optimal hydrogen network for Case 2 with the placement of compressors (Purification reuse/recycle).

hydrogen utility versus number of hydrogen headers, hydrogen utility versus number of connections, placement of compressors and purifiers. Two case studies are analyzed 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 decision can be made according to the resulted Pareto front. The results show that the minimum hydrogen utility without hydrogen header can be achieved for the network with two hydrogen headers and there is no direct connection between hydrogen sources and sinks. With the installation of purifier, the flowrate of the hydrogen utility can be further reduced.

Acknowledgments

Financial support provided by the National Basic Research Program of China (No. 2012CB720500) and National Natural Science Foundation of China (No. U1162121) are gratefully acknowledged. The research is also supported by Science Foundation of China University of Petroleum, Beijing (No. YJRC-2011-08 and LLYJ-2011-61).

Notation

Sets and indices

NHU	set of hydrogen utilities
NSR	set of hydrogen sources
NSK	set of hydrogen sinks
NI	set of compressors
NC	set of components
NH	set of hydrogen headers
NP	set of purifier
NF	set of fuel system
u	index for hydrogen utility
s	index for hydrogen source
k	index for hydrogen sink
i	index for compressor
c	index for component
h	index for hydrogen header
p	index for purifier
f	index for fuel system

Parameters

FHU_u^{UB}	upper bound for flow rate allocated from hydrogen utility u
FSR_s^{UB}	upper bound for flow rate allocated from hydrogen source s
$FI_i^{in,UB}$	upper bound for flow rate allocated for the inlet of compressor i
FK_k^{in}	inlet flow rate for hydrogen sink k
P_u	outlet pressure for hydrogen utility u
P_s	outlet pressure for hydrogen source s
P_k	inlet pressure for hydrogen sink k
P_h	pressure for hydrogen header h
P_p^{in}	inlet pressure for purification system
P_p^{prod}	product pressure for purifier p
P_p^{resd}	residue pressure for purifier p
P_f	inlet pressure for fuel system
ΔP^{UB}	upper bound for pressure difference between a and b
ΔP^{LB}	lower bound for pressure difference between a and b
$RR_{p,c}$	recovery ratio for cth component in purifier
$y_{u,c}$	outlet concentration of component c for hydrogen utility u
$y_{s,c}$	outlet concentration of component c for hydrogen source s
$y_{k,c}^{max}$	maximum inlet concentration of component c for hydrogen sink k
$y_{k,c}^{min}$	minimum inlet concentration of component c for hydrogen sink k

Continuous variables

FHU_u	flow rate allocated from hydrogen utility u
$FUH_{u,h}$	flow rate from hydrogen utility u to hydrogen header h
$FUI_{u,i}$	flow rate from hydrogen utility u to hydrogen compressor i
FSR_s	flow rate allocated from hydrogen source s
$FSH_{s,h}$	flow rate from hydrogen source s to hydrogen header h
$FSI_{s,i}$	flow rate from hydrogen source s to hydrogen compressor i
$FSF_{s,f}$	flow rate from hydrogen source s to fuel system
$FSP_{s,p}$	flow rate from hydrogen source s to purifier p
FH_h^{in}	inlet flow rate for hydrogen header h
FH_h^{out}	outlet flow rate for hydrogen header h
$FHK_{h,k}$	flow rate from hydrogen header h to hydrogen sink k
$FHI_{h,i}$	flow rate from hydrogen header h to compressor i
$FHF_{h,f}$	flow rate from hydrogen header h to fuel system
FI_i^{in}	inlet flow rate for compressor i
FI_i^{out}	outlet flow rate for compressor i
$FIK_{i,k}$	flow rate from compressor i to hydrogen sink k
$FIP_{i,p}$	flow rate from compressor i to purifier p
FP_p^{feed}	inlet flow rate for purifier p
FP_p^{prod}	product flow rate for purifier p
$FP_p^{residue}$	residue flow rate for purifier p
$FPH_{p,h}^{prod}$	flow rate from product of purifier p to hydrogen header h
$FPI_{p,i}^{prod}$	flow rate from product of purifier p to hydrogen compressor i
$FPF_{p,f}^{resd}$	flow rate from residue of purifier p to fuel system
FF_f^{in}	inlet flow rate for fuel system
$F_{a,b}$	flow rate between supplier a and receiver b
N_{total}	total number of connections
$y_{h,c}^{in}$	inlet concentration of component c for hydrogen header h
$y_{h,c}^{out}$	outlet concentration of component c for hydrogen header h
$y_{i,c}^{in}$	inlet concentration of component c for compressor i
$y_{i,c}^{out}$	outlet concentration of component c for compressor i
$y_{k,c}^{in}$	inlet concentration of component c for hydrogen sink k
$y_{p,c}^{feed}$	inlet concentration of component c for purifier p
$y_{p,c}^{prod}$	product concentration of component c for purifier p
$y_{p,c}^{resd}$	residue concentration of component c for purifier p
$y_{f,c}^{in}$	inlet concentration of component c for fuel system

Binary variables

$z_{a,b}$ connection between supplier *a* and receiver *b*

$Z^{\Delta p}_{a,b}$ pressure difference between supplier *a* and receiver *b*

Subscripts/Superscripts

LB Lower bound

UB Upper bound

min minimum

max maximum

in inlet

out outlet

mix mix

Abbreviations

CNHT cracked naphtha hydrotreater

CCR continuous catalytic reforming

DHT diesel hydrotreater

GOHT gas oil hydrotreater

HCU hydrocracker unit

MILP mixed integer linear programming

MINLP mixed integer non-linear programming

MRPD material surplus composite curve

NHT naphtha hydrotreater

PSA pressure swing adsorption

RHT residue hydrotreater

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.ijhydene.2014.06.129

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