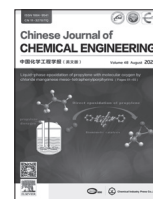




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Fuzzy optimization design of multicomponent refinery hydrogen network

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ABSTRACT

Hydrogen and light hydrocarbon components are essential resources of the refinery. The optimization of the refinery hydrogen system and recovery of the light hydrocarbon components contained in the gas streams are key strategies to reduce the operating costs for sustainable development. Many research efforts have been focused on the optimization of single impurity hydrogen network, and the flowrates of the hydrogen sources and sinks are assumed to be constant. However, their flowrates vary along with the quality of crude oil and refinery processing plans. A general superstructure of multicomponent refinery hydrogen network is proposed, which considers four components, namely H_2 , H_2S , CH_4 and C_{2+} , as well as the flowrate variations of hydrogen source and hydrogen sink. The mathematical model based on the superstructure is developed with objective functions, including the minimization of total annualized cost and the maximization of overall satisfaction of the hydrogen network. Moreover, the model considers the removal of hydrogen sulfide and the recovery of light hydrocarbon components (*i.e.*, C_{2+}) in the optimization. To verify the applicability of the proposed mathematical model, a simplified industrial case study with four scenarios is solved. The optimization results show that the economic benefit can be maximized by considering both the direct reuse of gas streams from high-pressure separator (HP gas stream) and from low-pressure separator (LP gas stream) and the recovery of the light hydrocarbon streams. The fuzzy optimization method can be used to guide the optimal design of the refinery hydrogen system with multi-period variable flowrates.

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1. Introduction

The quantity of inferior crude oil bought and processed by modern refineries is increasing year by year. Simultaneously, environmental legislation on the composition of sulfur content in clean oils has become more stringent. The proportion of hydrotreating and hydrocracking processes is increasing, which requires a large amount of hydrogen consumption. The sharp increase in hydrogen consumption in refineries makes the problem of hydrogen shortage more prominent. Besides, the refinery gas produced in the hydrogenation process contains many highly valued components such as light hydrocarbon components (*i.e.*, C_2H_6 , C_3H_8 , $n-C_4H_{10}$, $n-C_5H_{12}$). It is more critical to fully recycle the effective resources

(*i.e.*, light hydrocarbon components) in refinery off-gas streams. Once the light hydrocarbon components are recovered, a hydrogen-rich stream can be generated and further reused to the hydrogen system, thereby improving the hydrogen utilization ratio. From another perspective, the hydrogen-rich gas streams from the refinery can be further purified to produce high-purity hydrogen, which can be hydrogen sources for the development of hydrogen economics.

At present, the widely accepted methods for improving the utilization ratio of hydrogen are integrating and optimizing the refinery hydrogen network. The methods are mainly the pinch method and mathematical programming. Alves and Towler [1] first introduced the Hydrogen Surplus Diagram, which was used to determine the pinch point of the hydrogen system and the minimum target of hydrogen utility. Later, many studies based on the pinch method to solve the minimum flowrate of hydrogen utility have been proposed, such as Material Recovery Pinch Diagram [2],

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Source Composite Curve [3], Gas Cascade Analysis [4], Composite Algorithm Table [5], and Improved Problem Table [6]. Some research also extended the pinch method to the integrated purification and reuse hydrogen network. For example, Liu *et al.* [7,8] considered the impact of multiple impurities and the inlet flowrate of purification on hydrogen utility in hydrogen networks. Liao *et al.* [9,10] deduced the optimal conditions for hydrogen network allocation with and without purification, and developed a rigorous solution method based on mathematical derivation. Deng *et al.* [11] presented the hydrogen network integrated with the shared purifier in the hydrogen production plant. In addition, nearest neighbor algorithm (NNA) [12] is a widely accepted design method for hydrogen networks that meets flowrate requirements and is extended to design hydrogen networks with minimal compression work [13]. However, the pinch method has certain limitations when solving problems including cost factors, multiple impurities, or pressure limitations.

Hallale and Liu [14] firstly developed a method for optimizing hydrogen distribution networks based on superstructures to solve the minimal annual total costs in the refinery. Then, superstructure-based mathematical programming methods were used to solve problems with different constraints, such as multi-component flash evaporation [15], hydrogen header [16,17], and shortcut model [18]. Wu *et al.* [19] proposed a multi-component hydrogen purification optimization model and considered the light hydrocarbon recovery process in the purification unit. Li *et al.* [20] developed a superstructure model for the integration of refinery hydrogen network and hydrogen-storage based purifier, which can reduce the consumption of hydrogen utility. The light hydrocarbon recovery methods in the refinery mainly include cryogenic separation, absorption separation, adsorption separation [21], membrane separation [22], and hydrate separation. Recently, the simulation and modeling of unit modules during the hydrogenation process (*i.e.*, hydrogenation reactor [23], hydrogen separator [24], hydrogen purifier [25], H_2S removal [26,27] and hybrid hydrogen network of the chemical plant [28] are integrated into the optimization of the refinery hydrogen network.

Multi-period [29,30] and uncertainty problems in refinery also can be solved by mathematical programming methods. At present, most studies on hydrogen system optimization regard the outlet of the hydrogenation unit as a hydrogen source stream and consider its flowrate as a fixed value. The uncertainty design of hydrogen networks includes chance constrained programming [31], robust optimization [32], and stochastic programming [33]. For the hydrogen network, Liao *et al.* [34] proposed the concept of mixed potential to represent the disturbance resistance of the hydrogen network. Sardashti Birjandi *et al.* [35] developed a method based on steady-state flexibility to study the flexibility of hydrogen networks under operational uncertainty. Liang *et al.* [36] considered a multi-period discrete operation scheme and fluctuations, and presented a multi-period hydrogen network flexible design method that can meet the fluctuation flexibility of each sub-period. Kang *et al.* explored the impact of different integration schemes on the economics of hydrogen networks [37] and a multi-period hydrogen network design method based on flexible analysis [17]. Considering the multi-scale uncertainty of refinery hydrogen network, Chen *et al.* [38] proposed a stochastic programming model under flexible constraint framework. In the actual operation, the hydrogenation process will generate several hydrogen streams, such as gas streams from high-pressure separator (HP gas stream) and from low-pressure separator (LP gas stream), and dry gas. At the same time, the flowrate of these streams is generally not fixed and may change with the differences of the actual processing oil properties and processing plans, which affects the optimal design of the hydrogen network. It makes sense to take into account the dynamic flow of streams in the hydrogen network [39].

Compared with the deterministic model, fuzzy optimization considers both the objective function and the uncertainty in the actual process, and obtains the more robust solutions. Zimmermann [40] applied the fuzzy linear programming to the vector maximum problem and always achieved effective solutions. Aviso *et al.* [41] presented a novel fuzzy mixed integer linear program model to optimize the biomass co-firing network, which considered the uncertainty of biomass types and regional biochar capacity. Tan and Cruz [42] proposed a fuzzy method for water network design and introduced parameter uncertainty into the mathematical model constraints. The fuzzy method has also proven to be suitable for water network problems with multiple objectives or multiple decisions [43,44]. Later, Tan [45] proposed a fuzzy model that considered the uncertainty of concentration and flowrate, as well as the integration of water networks for single and multiple plants. Considering the fuzzy parameters and the flexible constraints of the hydrogen network, Birjandi *et al.* [46] developed a fuzzy-based multiobjective nonlinear program model to optimize the hydrogen network of the actual refinery with the objective function of minimizing TAC and carbon emissions. Impurities in the hydrogen stream such as sulfide content [47] have a great influence on the performance of the hydrogen network. By extending the concept of single impurity system mixed potential to multi-impurity hydrogen network and combining with mathematical programming model, [48] the disturbance resistance ability of the refinery hydrogen network can be optimized. In recent years, fuzzy optimization has been increasingly applied in the technological, environmental and economic aspects of energy systems [49].

However, to the best of our knowledge, there are few studies conducted on the optimization of multi-impurities refinery hydrogen networks considering the flowrate variation of hydrogen sources and sinks, which is common situation in the practical refinery operation. To overcome the limitation, this paper presents a superstructure for the optimization of hydrogen network integrated with light hydrocarbon recovery. It is assumed that hydrogen streams contain H_2 , H_2S , CH_4 and C_2+ . The superstructure model includes all possible connections between external hydrogen source (catalytic reforming, hydrogen production units), purifier (*i.e.*, pressure swing adsorption, PSA), desulfurization unit, light hydrocarbon recovery unit, hydrogenation unit, and fuel system. In addition, a fuzzy optimization model of the multi-component hydrogen network is established by adding fuzzy constraints and taking the overall satisfaction of the hydrogen network as the objective function. The hydrogen network of a simplified industrial case is designed, and comparative analysis for multiple scenarios is conducted to illustrate the applicability of the proposed method.

2. Problem Statement

The general superstructure of the multi-component hydrogen network in a refinery can be illustrated in Fig. 1. A refinery hydrogen system mainly includes hydrogenation units (*i.e.*, hydrocracking, hydrotreating, *etc.*) and hydrogen supply units (*i.e.*, hydrogen production plant, continuous catalytic reforming (CCR), *etc.*). Hydrogen utility is generated from the hydrogen production plant and hydrogen-rich stream produced by the catalytic reforming unit can be regarded as external hydrogen sources ($u \in U$). The flowrate variation range of the external hydrogen sources is F_u^L (lower bound) to F_u^U (upper bound). The hydrogen header ($h \in H$) is mainly used to receive external hydrogen sources and supply hydrogen to the hydrogenation unit, and the pressure is defined as P_h . The inlet of the hydrogenation unit can be regarded as a process hydrogen sink ($k \in K$), and the inlet flowrate is required to be F_k^{in} . The upper and lower limit of flowrate is F_k^U and F_k^L , respectively. The minimum inlet hydrogen concentration of the hydrogenation unit is required

as $Y_{k,H_2}^{\min}$, and pressure is required as P_k^{in} . The outlet streams of the hydrogenation unit mainly include high-pressure gas (HP gas) and low-pressure gas (LP gas), which can be considered as the process hydrogen source ($i \in I$). The outlet flowrate F_i is between F_i^L and F_i^U . The hydrogen concentration and pressure are specified as Y_{i,H_2} and P_i . The desulfurization unit inside the hydrogenation unit can remove sulfur of HP gas and LP gas streams. The HP gas that meets the concentration constraint can be recycled directly or sent to other hydrogenation units, purification units, and fuel systems. If it does not meet the requirements, it needs to be utilized after desulfurization treatment. The LP gas needs to be desulfurized before being sent to other hydrogenation units, fuel systems, or purification. Generally, the process hydrogen source is rich in light hydrocarbon components such as C_{2+} . Also, the light hydrocarbon components in the hydrogen source of the recovery process can generate hydrogen-rich streams, which can be recycled to the process hydrogen sink or sent to the purification unit.

According to the refinery processing scheme and preliminary design data, each hydrogen source and hydrogen sink has a corresponding flowrate variation range, and this variation range is used to estimate the fuzzy demand of natural gas for hydrogen production. The model covers extreme solutions ranging from optimistic (high-risk) to conservative (low risk). It aims to determine an optimal hydrogen resource reduction and cost reduction and avoids the manufacture of low-quality oil products. The final hydrogen network would meet all flowrate and concentration balances in the fuzzy interval, as well as connection constraints, and cost constraints.

3. Mathematical Model

A list of indexes, sets, parameters, and variables is given in the Notation, where all parameters and all variables are respectively represented in uppercase and lowercase symbols.

3.1. Fixed flowrate constraints

3.1.1. External hydrogen sources

A typical hydrogen production technique is steam methane reforming (SMR), which includes three main steps: reforming, shift conversion, and purification. The shift gas is further upgraded in a purifier (such as PSA [50]) to generate product gas, which is commonly referred to as hydrogen utility or fresh hydrogen. The inlet stream of the PSA in hydrogen production plant includes shift gas, process hydrogen sources, and the outlet streams of light hydrocarbon recovery unit. The relationship between the inlet flowrate and the component balance of the PSA in hydrogen production plant is shown as Eqs. (1)–(3).

$$f_u = fup_{u,p} \quad \forall u \\ = \text{HPlant (hydrogen production plant), } p = \text{PSA1} \quad (1)$$

$$f_p^{\text{in}} = fup_{u,p} + \sum_{i \in I} fip_{i,p} + \sum_{r \in R} frp_{r,p} \quad \forall u = \text{HPlant}, p = \text{PSA1} \quad (2)$$

$$f_p^{\text{in}} \cdot y_{p,c}^{\text{in}} = fup_{u,p} \cdot Y_{u,c} + \sum_{i \in I} (fip_{i,p} \cdot y_{i,c}) + \sum_{r \in R} (frp_{r,p} \cdot y_{r,c}^{\text{out}}) \\ \forall u = \text{HPlant}, p = \text{PSA1}, c \in C \quad (3)$$

where, f_u denotes the flowrate of shift gas from the hydrogen production plant, and $fup_{u,p}$ is the flowrate of shift gas allocated to purifier p . f_p^{in} represents the inlet flowrate of purifier p , $fip_{i,p}$ and $frp_{r,p}$ denote the flowrate from process hydrogen sources i and outlet streams of Light Hydrocarbon Recovery r to purifier p . $Y_{u,c}$, $y_{i,c}$,

and $y_{r,c}^{\text{out}}$ denote the concentration of component c for shift gas, process hydrogen sources and outlet stream of the LHR, respectively.

Shift gas is obtained from the shift conversion of natural gas for hydrogen production. Thus, the flowrate of natural gas can be calculated by Eq. (4).

$$fu_{ng} = \beta \cdot f_u \quad \forall u = \text{HPlant} \quad (4)$$

where the conversion coefficient β is specified as 0.2282, which is taken from petrochemical plant in Northeast China.

The hydrogen-rich gas from the continuous catalyst reforming (CCR) unit can be sent to the purifier (PSA in CCR) for upgrading. The inlet flowrate of the PSA in CCR is as shown in Eqs. (5)–(7).

$$f_u = fup_{u,p} \quad \forall u = \text{CCR}, p = \text{PSA2} \quad (5)$$

$$f_p^{\text{in}} = fup_{u,p} \quad \forall u = \text{CCR}, p = \text{PSA2} \quad (6)$$

$$f_p^{\text{in}} \cdot y_{p,c}^{\text{in}} = fup_{u,p} \cdot Y_{u,c} \quad \forall u = \text{CCR}, p = \text{PSA2}, c \in C \quad (7)$$

The outlet streams of the PSA unit include product gas and residual gas streams. The inlet and outlet flowrate balance and the component balance of component c of purifier can be expressed as Eqs. (8)–(11). The product gas stream of the PSA unit is sent to the hydrogen header, while its residual gas stream is sent to the fuel system. The hydrogen recovery ratio of the PSA is defined as Eq. (12).

$$f_p^{\text{in}} = f_p^{\text{prod}} + f_p^{\text{resd}} \quad \forall p \in P \quad (8)$$

$$f_p^{\text{in}} \cdot y_{p,c}^{\text{in}} = f_p^{\text{prod}} \cdot y_{p,c}^{\text{prod}} + f_p^{\text{resd}} \cdot y_{p,c}^{\text{resd}} \quad \forall p \in P, c \in C \quad (9)$$

$$f_p^{\text{prod}} = \sum_{h \in H} fph_{p,h} \quad \forall p \in P \quad (10)$$

$$f_p^{\text{resd}} = fpf_{p,f} \quad \forall p \in P \quad (11)$$

$$f_p^{\text{in}} \cdot y_{p,c}^{\text{in}} \cdot R_{p,c} = f_p^{\text{prod}} \cdot Y_{p,c}^{\text{prod}} \quad \forall p \in P, c = H_2 \quad (12)$$

where, f_p^{in} is the total inlet flowrate of PSA, f_p^{prod} is the product flowrate of PSA, and f_p^{resd} is the residual flow rate of the PSA, $fph_{p,h}$ is the flow rate of PSA to the hydrogen network, and $fpf_{p,f}$ is the flow rate of PSA discharged to the fuel gas system, $R_{p,c}$ is the hydrogen recovery ratio of PSA unit, $y_{p,c}^{\text{in}}$, $y_{p,c}^{\text{prod}}$, and $y_{p,c}^{\text{resd}}$ respectively represent the composition of component c of PSA inlet, outlet product gas, and residual gas streams.

3.1.2. Hydrogen header

It assumes that there is only one hydrogen header in the hydrogen system and its inlet flowrate is equal to its outlet flowrate. The inlet streams of the hydrogen header are the product gas streams of purifier p . The flowrate balance can be described as Eqs. (13)–(15).

$$f_h^{\text{in}} = \sum_{p \in P} fph_{p,h} \quad \forall h \in H \quad (13)$$

$$f_h^{\text{in}} \cdot y_{h,c} = \sum_{p \in P} (fph_{p,h} \cdot Y_{p,c}^{\text{prod}}) \quad \forall h \in H, c \in C \quad (14)$$

$$f_h^{\text{out}} = \sum_{k \in K} fhk_{h,k} \quad \forall h \in H \quad (15)$$

where f_h^{in} and f_h^{out} represent the inlet and outlet flowrate of hydrogen header, $fph_{p,h}$ represents the flowrates from purifier to hydro-

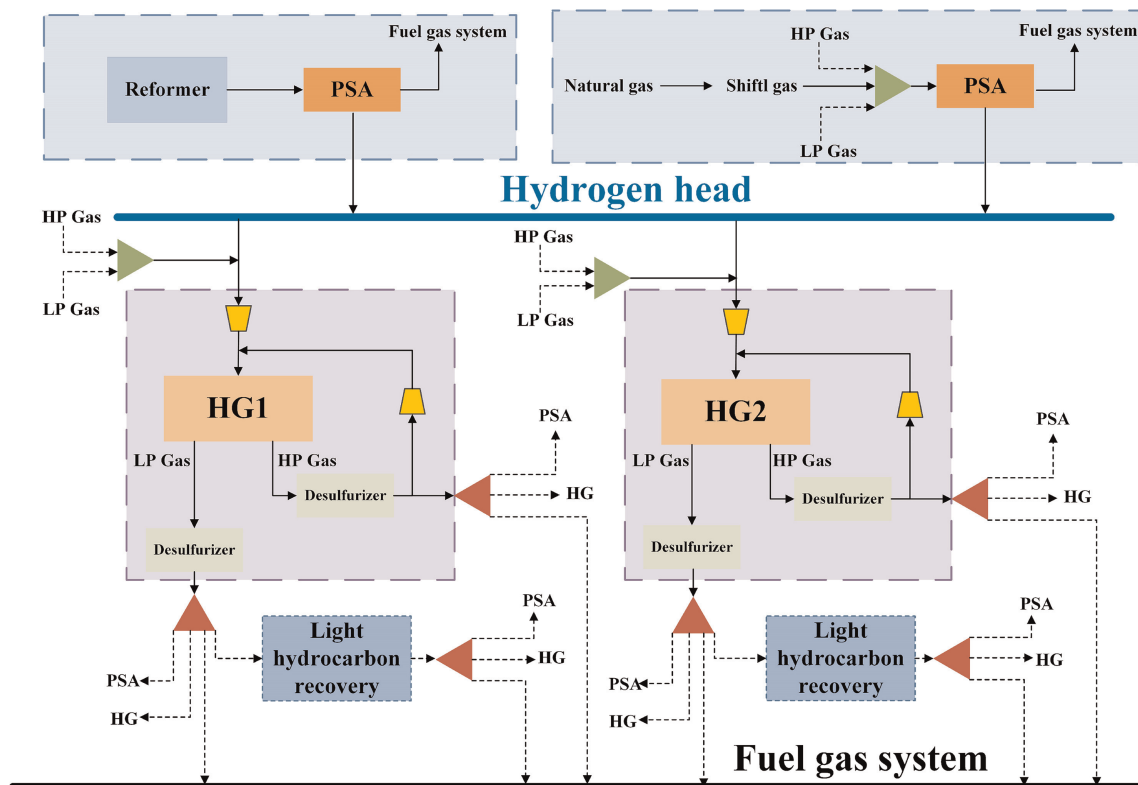


Fig. 1. Superstructure of optimal design of multi-component refinery hydrogen network.

gen header, and $f_{hk_{h,k}}$ represents the flowrates from hydrogen header to hydrogenation unit, $y_{h,c}$ denotes the concentration of component c of hydrogen header.

3.1.3. Hydrogenation unit

The hydrogenation unit can receive hydrogen streams from hydrogen header, process hydrogen sources, and light hydrocarbon recovery unit. The inlet flowrate and component balance for the hydrogenation unit are shown in Eqs. (16) and (17).

$$f_k^{\text{in}} = \sum_{h \in H} f_{hk_{h,k}} + \sum_{i \in I} f_{ik_{i,k}} + \sum_{r \in R} f_{rk_{r,k}} \quad \forall k \in K \quad (16)$$

$$f_k^{\text{in}} \cdot y_{k,c}^{\text{in}} = \sum_{h \in H} (f_{hk_{h,k}} \cdot y_{h,c}) + \sum_{i \in I} (f_{ik_{i,k}} \cdot y_{i,c}) + \sum_{r \in R} (f_{rk_{r,k}} \cdot y_{r,c}^{\text{out}}) \quad \forall k \in K, c \in C \quad (17)$$

The inlet hydrogen concentration of the hydrogenation unit ($y_{k,c}^{\text{in}}$) should not be less than the minimum concentration requirement of hydrogen ($Y_{k,c}^{\text{min}}, \forall k \in K, c = \text{H}_2$), and the H_2S concentration should not be greater than the maximum concentration requirement of H_2S ($Y_{k,c}^{\text{max}}, \forall k \in K, c = \text{H}_2\text{S}$). The constraints can be expressed via Eqs. (18) and (19).

$$y_{k,c}^{\text{in}} \geq Y_{k,c}^{\text{min}} \quad \forall k \in K, c = \text{H}_2 \quad (18)$$

$$y_{k,c}^{\text{in}} \leq Y_{k,c}^{\text{max}} \quad \forall k \in K, c = \text{H}_2\text{S} \quad (19)$$

The outlet streams (HP gas and LP gas) of the hydrogenation unit, which can be treated as process hydrogen sources, should be sent to the corresponding desulfurization unit if necessary. The desulfurization unit inside of the hydrogenation unit can remove H_2S . When the H_2S concentration of the stream is higher than the maximum concentration requirement, it needs to be

desulfurized. However, while the H_2S concentration meets the inlet requirements of the hydrogenation unit, desulfurization treatment is not considered.

It is assumed that the desulfurization rate is $R_{k,c}^{\text{DS}}$, which is expressed by Eq. (20). When the outlet stream does not need desulfurization treatment, the desulfurization rate ($R_{k,c}^{\text{DS}}$) is set as zero. The outlet flowrate of the desulfurization unit can be determined by Eq. (21). The inlet and outlet streams satisfy the mass balance except for H_2S , as shown in Eq. (22).

$$F_k^{\text{out}} \cdot Y_{k,c}^{\text{out}} \cdot (1 - R_{k,c}^{\text{DS}}) = f_i \cdot y_{i,c} \quad \forall k \in K, c = \text{H}_2\text{S} \quad (20)$$

$$f_i = F_k^{\text{out}} - F_k^{\text{out}} \cdot Y_{k,c}^{\text{out}} \cdot R_{k,c}^{\text{DS}} \quad \forall k \in K, i \in I, c = \text{H}_2\text{S} \quad (21)$$

$$f_i \cdot y_{i,c} = F_k^{\text{out}} \cdot Y_{k,c}^{\text{out}} \quad \forall k \in K, i \in I, c \neq \text{H}_2\text{S} \quad (22)$$

where F_k^{out} denotes the outlet flowrate of the hydrogenation unit, which includes HP gas and LP gas, f_i and $y_{i,c}$ denote the flowrate and the concentration of component c of process hydrogen source, respectively.

The process hydrogen sources can be sent to other hydrogenation units, the purifier (i.e., PSA) or fuel system, and the flowrate balance is expressed in Eq. (23).

$$f_i = \sum_{k \in K} f_{ik_{i,k}} + \sum_{p \in P} f_{ip_{i,p}} + \sum_{r \in R} f_{ir_{i,r}} + f_{if_{i,f}} \quad \forall i \in I \quad (23)$$

where, $f_{ik_{i,k}}$, $f_{ip_{i,p}}$ and $f_{if_{i,f}}$ respectively represent the flowrate of the process hydrogen source to the hydrogenation unit, the purifier, and the fuel system.

3.1.4. Light hydrocarbon recovery

The desulfurized LP gas can be sent to a light hydrocarbon recovery unit (LHR) to recover light hydrocarbon components and generate hydrogen-rich streams. Assuming that the light hydrocarbon recovery unit can recover all C_{2+} in the stream, then

the C_{2+} concentration of the outlet stream is zero. The outlet flowrate of the light hydrocarbon recovery unit can be determined by Eq. (24). Other components except for C_{2+} satisfy the mass balance of the light hydrocarbon recovery unit, as shown in Eq. (25).

$$f_r^{\text{out}} = f_i - f_i \cdot y_{i,c} \quad \forall r \in R, c = C_{2+} \quad (24)$$

$$f_r^{\text{out}} \cdot y_{r,c}^{\text{out}} = f_i \cdot y_{i,c} \quad \forall r \in R, c \neq C_{2+} \quad (25)$$

The outlet hydrogen-rich stream of the light hydrocarbon recovery unit can be sent to the purifier (i.e., PSA), other hydrogenation units, or the fuel system, as shown in Eq. (26).

$$f_r^{\text{out}} = \sum_{p \in P} f_{rp} + \sum_{k \in K} f_{rk} + f_{rf} \quad \forall r \in R \quad (26)$$

3.1.5. Fuel system

The inlet stream of the fuel system mainly includes the residual gas of the purifier, process hydrogen sources, and the outlet stream of the light hydrocarbon recovery unit. The flowrate balance is shown as Eq. (27).

$$f_{\text{Fuel}} = \sum_{p \in P} f_{pf} + \sum_{i \in I} f_{if} + \sum_{r \in R} f_{rf} \quad (27)$$

3.1.6. Connection constraints

Binary variable $z_{a,b}$ is introduced to determine the connections between the supplier (a) and the receiver (b). The sufficient and necessary condition for the existence of the connection of the stream is that the flowrate is positive. Therefore, when the connection between the streams exists, the value of the binary variable equals one. Furthermore, the value of the binary variable is zero, which indicates the connection does not exist. The relationship between binary variables and continuous variables is shown in Eq. (28).

$$\begin{aligned} f_{a,b} - z_{a,b} \cdot F_{a,b}^{\text{UB}} &\leq 0 \\ f_{a,b} - z_{a,b} \cdot F_{a,b}^{\text{LB}} &\geq 0 \\ z_{a,b} &\in \left\{ \begin{array}{l} z_{ph,p,h}, z_{pf,p,h}, z_{hk,h,k} \\ z_{ik,i,k}, z_{ip,k,p}, z_{ir,i,r}, z_{if,k,f} \\ z_{rk,r,k}, z_{rp,r,p}, z_{rf,r,f} \end{array} \right\} \\ f_{a,b} &\in \left\{ \begin{array}{l} f_{ph,p,h}, f_{pf,p,h}, f_{hk,h,k} \\ f_{ik,i,k}, f_{ip,i,p}, f_{ir,i,r}, f_{if,i,f} \\ f_{rk,r,k}, f_{rp,r,p}, f_{rf,r,f} \end{array} \right\} \end{aligned} \quad (28)$$

where $F_{a,b}^{\text{UB}}$ and $F_{a,b}^{\text{LB}}$ represent the upper and lower bounds for the flowrate variable $f_{a,b}$, respectively.

The compressor may need to be installed when the pressure level of the receiver is greater than the pressure of the supplier. An additional binary variable $z_{a,b}^{\Delta P}$ is introduced to indicate the pressure difference between the two streams. The relationship between the pressure difference of the binary variable $z_{a,b}^{\Delta P}$ is shown as Eq. (29).

$$\begin{aligned} (P_b - P_a) - \Delta P^{\text{max}} \cdot z_{a,b}^{\Delta P} &\leq 0 \\ (P_b - P_a) + \Delta P^{\text{max}} \cdot (1 - z_{a,b}^{\Delta P}) &\geq \Delta P^{\text{min}} \\ z_{a,b}^{\Delta P} &\in \left\{ \begin{array}{l} z_{ph,p,h}^{\Delta P}, z_{pf,p,h}^{\Delta P}, z_{hk,h,k}^{\Delta P} \\ z_{ik,i,k}^{\Delta P}, z_{ip,k,p}^{\Delta P}, z_{ir,i,r}^{\Delta P}, z_{if,k,f}^{\Delta P} \\ z_{rk,r,k}^{\Delta P}, z_{rp,r,p}^{\Delta P}, z_{rf,r,f}^{\Delta P} \end{array} \right\} \end{aligned} \quad (29)$$

where P_a and P_b represent the outlet pressure levels of supplier a and receiver b , while ΔP^{min} and ΔP^{max} represent the minimum and maximum pressure difference.

When $z_{a,b}$ and $z_{a,b}^{\Delta P}$ are both 1, the value of the binary variable $z_{a,b}^{\text{comp}}$ is one, which can be expressed by Eq. (30). The compressor needs to be installed between supplier a and the receiver b when $z_{a,b}^{\text{comp}}$ is one.

$$1 - z_{a,b} - z_{a,b}^{\Delta P} + z_{a,b}^{\text{comp}} \geq 0 \quad (30)$$

The calculation formulations of compression work $W_{a,b}^{\text{comp}}$ are adopted from Deng *et al.* [13]. The compression work $W_{a,b}^{\text{comp}} = fab_{a,b} \cdot (\mu_b - \mu_a)$ can be obtained when $z_{a,b}^{\text{comp}}$ equals one, and when $z_{a,b}^{\text{comp}}$ is zero, the compression work $W_{a,b}^{\text{comp}}$ is equal to zero. The calculations can be described as Eqs. (31)–(33).

$$W_{a,b}^{\text{comp}} = fab_{a,b} \cdot (\mu_b - \mu_a) \quad (31)$$

$$W_{a,b}^{\text{comp}} \leq M_{\text{comp}}^{\text{max}} \cdot z_{a,b}^{\text{comp}} \quad (32)$$

$$\begin{aligned} W_{a,b}^{\text{comp}} - fab_{a,b} \cdot (\mu_b - \mu_a) &\leq (1 - z_{a,b}^{\text{comp}}) \cdot M_{\text{comp}}^{\text{max}} \\ W_{a,b}^{\text{comp}} - fab_{a,b} \cdot (\mu_b - \mu_a) &\geq (z_{a,b}^{\text{comp}} - 1) \cdot M_{\text{comp}}^{\text{max}} \end{aligned} \quad (33)$$

$$\begin{aligned} W_{a,b}^{\text{comp}} &\in \{W_{p,h}^{\text{comp}}, W_{h,k}^{\text{comp}}, W_{i,k}^{\text{comp}}, W_{i,p}^{\text{comp}}, W_{i,r}^{\text{comp}}, W_{r,k}^{\text{comp}}, W_{r,p}^{\text{comp}}\} \\ z_{a,b}^{\text{comp}} &\in \{z_{p,h}^{\text{comp}}, z_{h,k}^{\text{comp}}, z_{i,k}^{\text{comp}}, z_{i,p}^{\text{comp}}, z_{i,r}^{\text{comp}}, z_{r,k}^{\text{comp}}, z_{r,p}^{\text{comp}}\} \\ fab_{a,b} &\in \{f_{ph,p,h}, f_{hk,h,k}, f_{ip,i,p}, f_{ik,i,k}, f_{ir,i,r}, f_{rk,r,k}, f_{rp,r,p}\} \end{aligned} \quad (34)$$

where $fab_{a,b}$ represents the flowrate between supplier a and receiver b , μ_a and μ_b denote the pressure index.

3.2. Fuzzy constraints

The overall degree of fuzzy constraint satisfaction is defined as λ , and the fuzzy level of satisfaction is between zero and one (Eq. (35)). The value zero is corresponding to unsatisfactory, one is completely satisfactory, and fractional values indicate partial satisfaction.

$$0 \leq \lambda \leq 1 \quad (35)$$

In fuzzy optimization using “max-min” aggregation, every fuzzy constraint should be associated with the degree λ . Note that the hydrogen consumption is at the upper bound when $\lambda = 0$, at the lower bound when $\lambda = 1$. The consumption of shift gas in hydrogen production (f_u) is specified fuzzy optimization goals for the network; the linear membership function is using to expressed as Eq. (36), which is adopted from Tan *et al.* [45]

$$f_u - F_u^{\text{U}} = \lambda(F_u^{\text{L}} - F_u^{\text{U}}) \quad (36)$$

where F_u^{L} and F_u^{U} respectively represent the lower and upper limits of the external hydrogen source.

The flowrate balance of the process hydrogen source is shown in Eq. (37), which can be sent to the hydrogenation unit k , PSA unit p , light hydrocarbon recovery unit r , and the fuel system f . It can be seen that when $\lambda = 0$, the flowrate of the process hydrogen source is F_i^{U} , while when $\lambda = 1$, the flowrate of the process hydrogen source is F_i^{L} . This relationship means that lower values of λ are inherently more optimistic (and thus riskier), while higher values of λ are inherently more conservative. Therefore, the objective function forces the model towards a more conservative solution with a

robust network. If the hydrogen available from source i is known precisely, then the fuzzy interval has zero width, namely $F_i^U = F_i^L$.

$$F_i^U + \lambda(F_i^L - F_i^U) = \sum_{k \in K} f_{ik,i,k} + \sum_{p \in P} f_{ip,i,p} + \sum_{r \in R} f_{ir,i,r} + \sum_{f \in F} f_{if,i,f} \quad \forall i \in I \quad (37)$$

Eqs. (38) and (39) describe the flowrate and component balance of each hydrogenation unit. It can be seen that the inlet flowrate for the hydrogenation unit k is required to be F_k^L when $\lambda = 0$. While $\lambda = 1$, the inlet flowrate for the hydrogenation unit k is required to be F_k^U . The logic used is similar, with higher λ denoting more conservative (or less risky) conditions. In addition, if the hydrogen demand of sink k is known precisely, the width of the fuzzy interval is zero.

$$F_k^L + \lambda(F_k^U - F_k^L) = \sum_{h \in H} f_{hk,h,k} + \sum_{i \in I} f_{ik,i,k} + \sum_{r \in R} f_{rk,r,k} \quad \forall k \in K \quad (38)$$

$$\begin{aligned} [F_k^L + \lambda(F_k^U - F_k^L)] \cdot y_{k,c}^{\text{in}} &= \sum_{h \in H} (f_{hk,h,k} \cdot y_{h,c}) + \sum_{i \in I} (f_{ik,i,k} \cdot y_{i,c}) \\ &+ \sum_{r \in R} (f_{rk,r,k} \cdot y_{r,c}^{\text{out}}) \quad \forall k \in K, c \in C \end{aligned} \quad (39)$$

3.3. Economic models

The total annualized cost (TAC) of the hydrogen system in the refinery, including operating costs and equipment investment costs, is expressed as Eq. (40). The annual working time is 8000 h.

$$\begin{aligned} \zeta_{\text{TAC}} &= OC_{\text{H}_2} + OC_{\text{Power}} + OC_{\text{PSA}} + OC_{\text{LHR}} - OE_{\text{Fuel}} - OE_{\text{LHR}} \\ &+ Af \left(\sum_{\text{new}} C_n + \sum_{\text{new}} C_{\text{pipe}} + \sum_{\text{new}} C_{\text{LHR}} \right) \end{aligned} \quad (40)$$

where Af represents the annualized factor of equipment investment cost, as shown in Eq. (41), fi denotes the interest rate fraction per year, and ny denotes the number of years of depreciation. (i.e. $fi = 5\%$, $ny = 5$)

$$Af = fi \cdot (1 + fi)^{ny} / [(1 + fi)^{ny} - 1] \quad (41)$$

3.3.1. Operation cost

The operating cost mainly includes the cost of natural gas used as raw material for hydrogen production, the electricity charge generated by the compressor, the operating cost of PSA, the operating cost of the light hydrocarbon recovery, the fuel benefit, and the light hydrocarbon recovery benefit.

The operating cost of hydrogen utility can be calculated as Eq. (42). Note that, because the flowrate of rich-hydrogen stream of CCR is not an optimized target of hydrogen system, the hydrogen cost of CCR is not considered.

$$OC_{\text{H}_2} = \sum e_{\text{H}_2} \cdot f_{hu,u} \cdot t \quad \forall u = \text{HPlant} \quad (42)$$

$$e_{\text{H}_2} = 0.06 + 0.31e_{\text{ng}} \quad (43)$$

where $f_{hu,u}$ is the flowrate of the hydrogen utility, and t denotes the annual operating time. Eq. (43) is used to calculate the unit hydrogen production cost (e_{H_2}), e_{ng} is the price of natural gas (0.26 USD·m⁻³) as the raw material for hydrogen production.

The operating cost OC_{Power} of the compressor can be calculated by Eq. (44).

$$OC_{\text{Power}} = \sum_{n \in N} e_{\text{Power}} \cdot \text{Power}_n \cdot t \quad (44)$$

where, e_{Power} represents the unit price of power (0.057 USD·(kW·h)⁻¹), Power_n represents the compression work.

In an isothermal or adiabatic process, the compression work of the compressor n can be expressed by Eq. (45), $\mu(\mu_{\text{out}}$ and $\mu_{\text{in}})$ represent the pressure index, in an isothermal process, $\mu = P_0 \ln\left(\frac{P}{P_0}\right)$, and in an adiabatic process $\mu = \left(\frac{\gamma}{\gamma-1}\right)P_0 \ln\left(\frac{P}{P_0}\right)^{\frac{\gamma-1}{\gamma}}$ [51].

$$\text{Power}_n = \dot{f}_n^{\text{in}} \cdot (\mu_{\text{out}} - \mu_{\text{in}}) \quad (45)$$

where, γ is the adiabatic index, P_0 represents the standard pressure (101.325 kPa), P_{out} and P_{in} represent the outlet and inlet pressure of the compressor.

The operating cost of PSA is shown in Eq. (46), where e_{PSA} (0.0032 USD·m⁻³) denotes the operating cost coefficient of PSA and f_p^{in} is the inlet flowrate of PSA.

$$OC_{\text{PSA}} = \sum_{p \in P} e_{\text{PSA}} \cdot f_p^{\text{in}} \cdot t \quad (46)$$

The operating costs of light hydrocarbon recovery mainly include raw material costs and energy costs, as shown in Eq. (47).

$$OC_{\text{LHR}} = (ar \cdot f_r^{\text{in}} + br \cdot e_{\text{Power}} \cdot f_r^{\text{in}}) \cdot t \quad (47)$$

where ar (0.021 USD·kg⁻¹) is the raw material cost parameter of light hydrocarbon recovery, and br (6.51×10^3 kJ·kg⁻¹) is the energy parameter of the light hydrocarbon recovery unit. In addition, the gas discharged into the fuel system will generate part of the economic benefits OC_{fuel} . The gas in the fuel system is mainly methane and hydrogen; thus, OC_{fuel} can be estimated using Eq. (48).

$$OC_{\text{fuel}} = e_{\text{heat}} \cdot t \cdot f_{\text{fuel}} \left[\Delta H_{\text{c,CH}_4}^0 \cdot y_{\text{fuel,CH}_4} + \Delta H_{\text{c,H}_2}^0 \cdot y_{\text{fuel,H}_2} \right] \quad (48)$$

where, e_{heat} represents the benefit generated by per unit fuel gas (2.5 USD·(MMbtu)⁻¹, 1 MMbtu=1.055 GJ), f_{fuel} represents the total flowrate of the fuel system. According to the NIST Chemistry Web-Book, $\Delta H_{\text{c,CH}_4}^0$ represents the enthalpy of natural gas combustion under standard conditions, which is 890.35 kJ·mol⁻¹; $\Delta H_{\text{c,H}_2}^0$ represents the enthalpy of hydrogen combustion under standard conditions, which is 285.830 kJ·mol⁻¹.

3.3.2. Investment costs

Investment costs mainly include compressor costs, pipeline costs, and light hydrocarbon recovery costs. The investment cost of the compressor can be estimated by Eq. (49). a_n (220.8×10^3 USD) and b_n (3.6672×10^3 USD·kW⁻¹) represent the investment cost coefficients of compressor n , respectively.

$$C_n = a_n + b_n \cdot \text{Power}_n \quad (49)$$

The pipeline investment estimation formulation is shown in Eq. (50), where a_{pipe} and b_{pipe} are the correlation coefficients [14], which are 3.2 and 11.42, respectively. D is the diameter of the new pipeline, which can be calculated from Eq. (51), and l is the length of the pipeline.

$$C_{\text{pipe}} = (a_{\text{pipe}} + b_{\text{pipe}} \cdot D^2) \cdot l \quad (50)$$

$$D^2 = \frac{4 \cdot F \cdot \rho_0}{\pi \cdot v \cdot \rho} \quad (51)$$

where v represents the gas velocity in the pipeline (usually 15–30 m·s⁻¹), ρ and ρ_0 respectively represent the gas density under design conditions and standard conditions. F represents the gas flowrate in the pipeline.

The investment cost of the light hydrocarbon recovery unit can be estimated by Eq. (52). C_{RO} (1090×10^3 USD) and F_{RO} (8000 m³) represent the cost correlation coefficient of light hydrocarbon

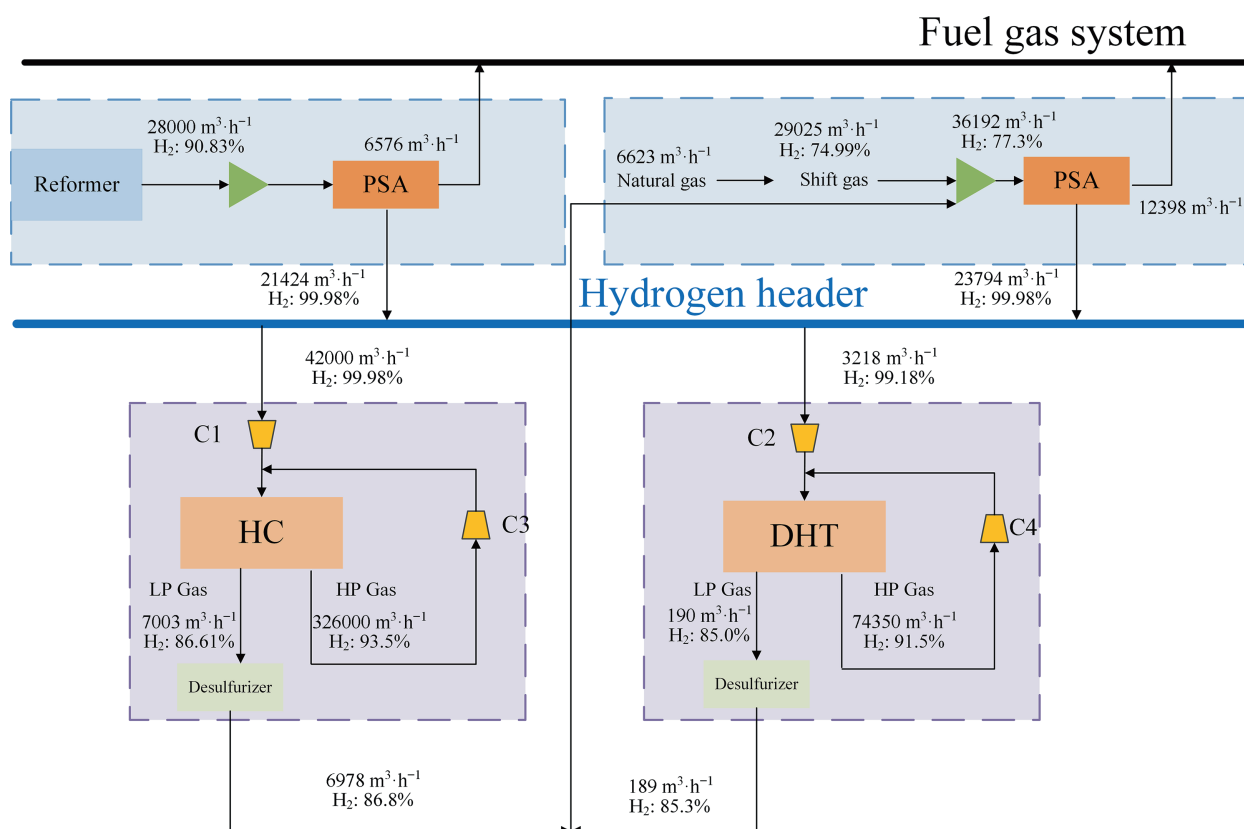


Fig. 2. Preliminary design scheme of refinery hydrogen system.

recovery, and f_R^{in} represents the inlet flowrate of the light hydrocarbon recovery unit, n is the correlation index that equals 0.8.

$$C_{\text{LHR}} = C_{\text{RO}} \cdot \left(\frac{f_R^{\text{in}}}{F_{\text{RO}}} \right)^n \quad (52)$$

3.4. Objective function

The objective function is to maximize the satisfaction λ of the hydrogen system that meets the requirements of fuzzy constraints, which is expressed by Eq. (53). The optimal value of the overall satisfaction λ of the hydrogen network can be explained as follows: For any given process, the parameters can define a fuzzy interval, ranging from optimistic (or risk) values to conservative (or low risk) values. For example, in Scenario 1b, the outlet flowrate constraint range of CCR is 27000–35000 $\text{m}^3 \cdot \text{h}^{-1}$. The upper bound of this range corresponds to conservative assumptions, while the

Table 2
Data for Compressors and PSAs

Compressor	Inlet Pressure/ MPa	Outlet Pressure/ MPa	Maximum Capacity/ $\text{m}^3 \cdot \text{h}^{-1}$
C1	2.2	16.6	46,000
C2	2.2	8.2	6800
C3	14.2	16.6	350,000
C4	7.0	8.2	80,000
PSA	Pressure/MPa	Hydrogen recovery ratio	Product hydrogen concentration (mole fraction)
PSA in CCR	2.5	0.85	0.9998
PSA in HPlant	2.5	0.85	0.9998

lower bound means riskier since the reduction flowrate of the external hydrogen source may cause the inlet flowrate of the hydrogenation unit to fail to meet the requirements.

Table 1
Data of hydrogen sources and sinks

	Streams	Flowrate/ $\text{m}^3 \cdot \text{h}^{-1}$	Pressure/MPa	Mole fraction			
				H_2	H_2S	C_{2+}	CH_4
Hydrogen sources	CCR	28000	2.2	0.908	0	0.048	0.053
	Shift gas	29025	2.2	0.750	0	0	0.250
	HP gas of HC	328100	14.3	0.935	0.001	0.017	0.047
	HP gas of DHT	74350	7.0	0.915	0.001	0.001	0.083
	LP gas of HC	7003	2.4	0.865	0.005	0.059	0.071
	LP gas of DHT	190	2.5	0.850	0.005	0.024	0.122
Hydrogen sinks	HC	368000	6.6	0.940	0.001	–	–
	DHT	77568	8.2	0.920	0.001	–	–

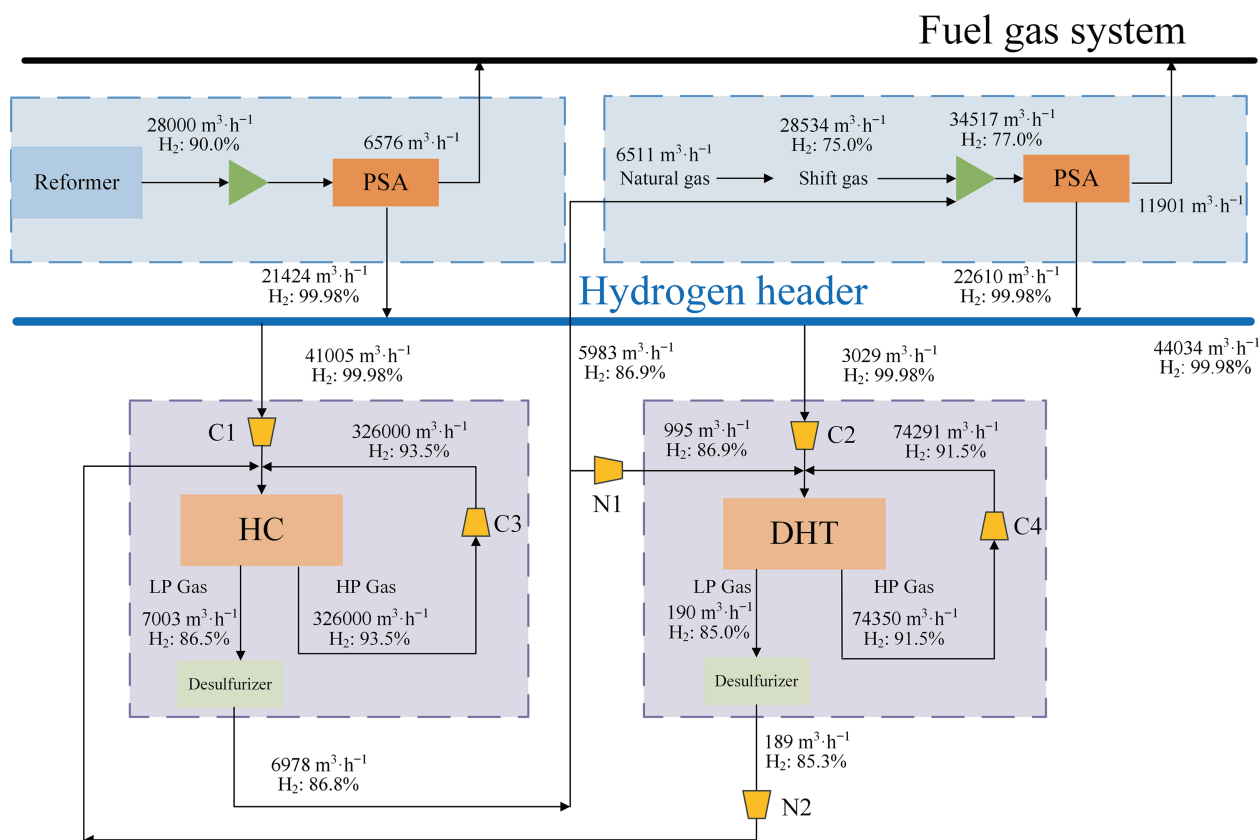


Fig. 3. Optimal design of hydrogen network (Scenario 1a).

Table 3
Investment cost of new compressors (Scenario 1a)

Equipment	Compressor N1	Compressor N2
Inlet pressure/MPa	2.4	2.4
Outlet pressure/MPa	8.2	16.6
Flowrate/m ³ ·h ⁻¹	995	189
Power/kW	101.77	33.98
Compressor cost × 10 ⁻³ /USD	593.29	345.17

Table 4
Fuzzy flowrate intervals of hydrogen sources and sinks

	Gas streams	Flowrate interval/ m ³ ·h ⁻¹
Hydrogen Sources	Hydrogen-rich stream in CCR	(27000, 35000)
	Shift gas in HPlant	(25000, 35500)
	HP gas from HC	(300000, 350000)
	HP gas from DHT	(70000, 80000)
	LP gas from HC	(6500, 7500)
Hydrogen Sinks	LP gas from DHT	(100, 300)
	Hydrocracking (HC)	(325000, 380000)
	Diesel hydrogenation (DHT)	(76000, 82000)

Table 5
Cost comparison of Scenario 1

	Preliminary design	Scenario 1a (non-fuzzy optimization)	Scenario 1b (fuzzy optimization)
Natural gas/m ³ ·h ⁻¹	6623	6511	6599
Hydrogen utility/m ³ ·h ⁻¹	23794	22610	22847
Fuel system/m ³ ·h ⁻¹	18973	18477	19086
Compressor investment cost × 10 ⁻³ /USD	–	938.47	884.31
Pipeline investment cost × 10 ⁻³ /USD	–	106.41	94.64
Total investment cost × 10 ⁻³ /USD	–	1044.88	978.95
Hydrogen production cost × 10 ⁻³ /USD	26595.02	25271.82	25536.24
Electricity cost × 10 ⁻³ / USD	6871	6866.16	6841.47
PSA operating cost × 10 ⁻³ / USD	1643.3	1599.86	1664.04
Fuel benefit × 10 ⁻³ /USD	2005.63	1953.16	2037.33
Total operating cost × 10 ⁻³ /USD	33103.69	31784.71	32004.43
TAC × 10 ⁻³ /USD	33103.69	32026.07	32230.13
Payback period/a		0.79	0.89

$$\max \lambda \quad (53)$$

The minimum total annualized cost (TAC) is taken as the objective function expressed by Eq. (54).

$$\min \zeta_{TAC} = OC_{H_2} + OC_{Power} + OC_{PSA} + OC_{LHR} - OE_{Fuel} - OE_{LHR} + Af(\sum_{new} C_n + \sum_{new} C_{pipe} + \sum_{new} C_{LHR}) \quad (54)$$

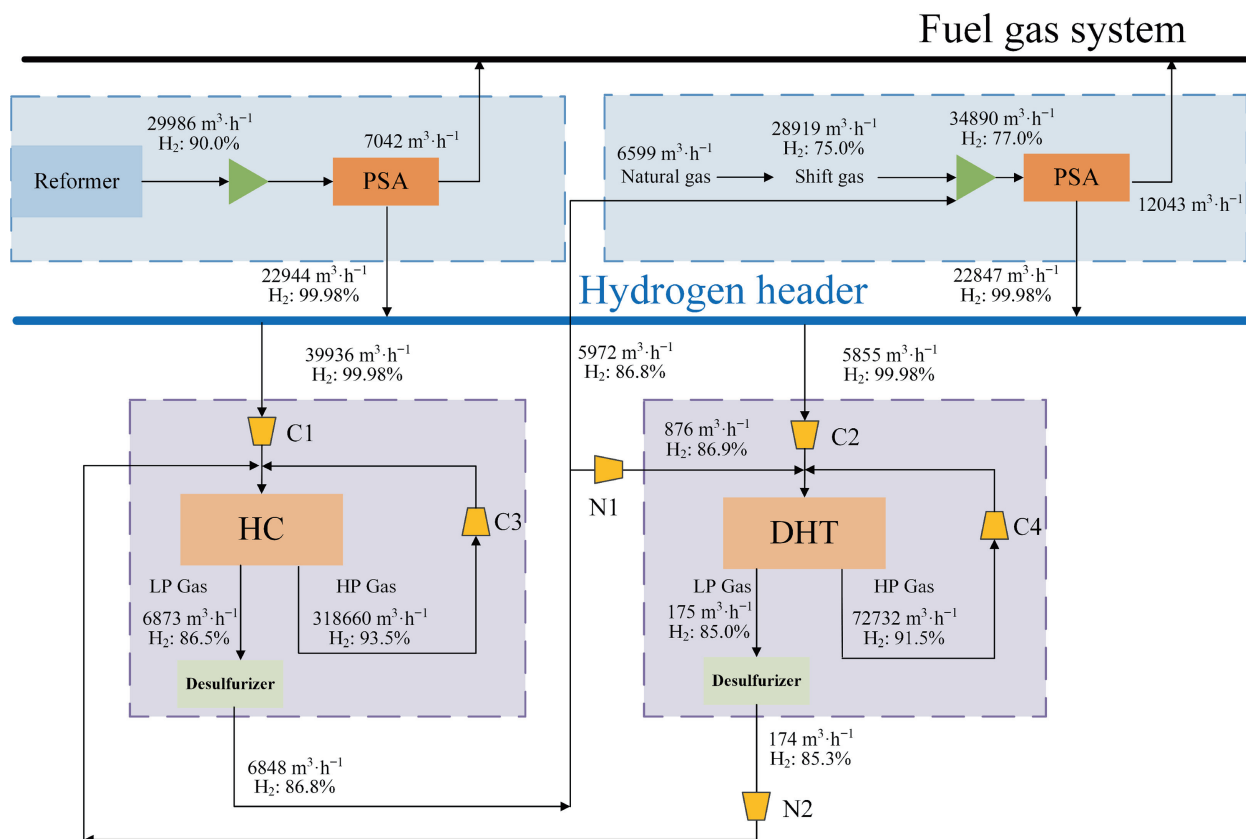


Fig. 4. Fuzzy optimized hydrogen network design (Scenario 1b).

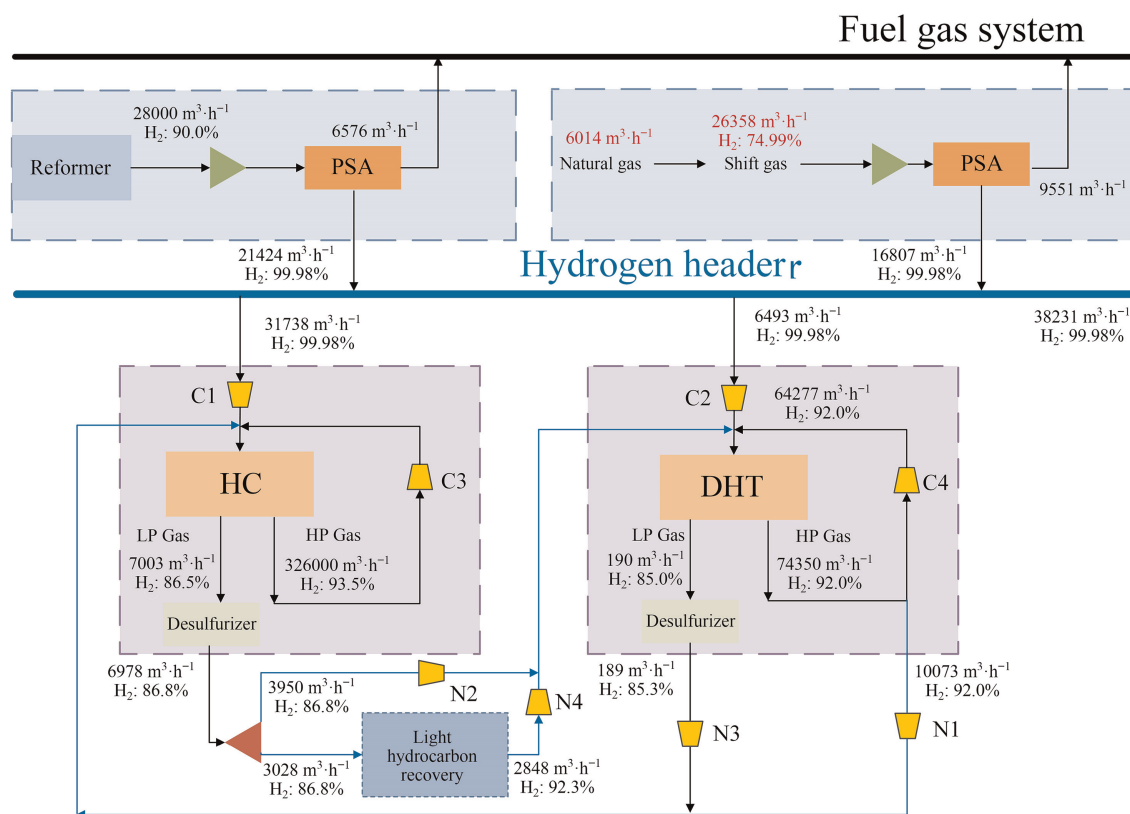


Fig. 5. Optimized design of hydrogen network with integrated light hydrocarbon recovery (Scenario 2a).

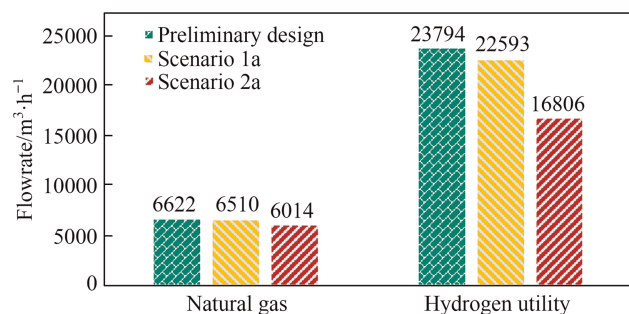


Fig. 6. Flowrate comparison of Scenario 1a and Scenario 2a.

Table 6

Compressors investment cost (Scenario 2a)

Equipment	N1	N2	N3	N4
Inlet pressure/MPa	7.0	2.4	2.4	1.05
Outlet pressure/MPa	16.6	8.2	16.6	8.2
Flowrate/m ³ ·h ⁻¹	10073	3950	189	2848
Power/kW	931.68	399.37	33.98	437.21
Compressor cost × 10 ⁻³ /USD	3628.94	1682.50	345.18	1820.96

3.5. Model summary

P1-Minimum TAC: The objective function is expressed by Eq. (54), which is a mixed integer nonlinear programming problem (MINLP).

Constraint Eqs: (1)–(34), (39)–(51).

Table 7

Compressors investment cost (Scenario 2b)

Equipment	N1	N2	N3
Inlet pressure/MPa	7.0	2.4	2.4
Outlet pressure/MPa	16.6	8.2	2.5
Flowrate/m ³ ·h ⁻¹	8047	5961	642
Power/kW	662.91	915.19	34.72
Compressor cost × 10 ⁻³ /USD	2647.04	3570.40	347.88

P2- Maximum overall hydrogen system satisfaction λ : the objective function is represented by Eq. (53), which is modeled as a nonlinear programming problem (NLP).

Constraint Eqs: (1)–(15), (18)–(22), (24)–(52).

The model runs on a 64-bit Windows 10, Intel® Core™ i7-8700 3.7 GHz, 16.00 GB memory system environment and GAMS 24.1.3 software environment. The MILP, NLP, and MINLP problems are solved by CPLEX, IPOPTH, and DICOPT, respectively. The absolute error of the solver's optimal value is set to 10⁻⁶.

4. Case Description

To illustrate the applicability of the proposed mathematical model, this paper optimizes the simplified hydrogen network case from a refinery in Northeast China. In the preliminary hydrogen network design as shown in Fig. 2, the hydrogenation unit includes hydrocracking (HC) and diesel hydrogenation (DHT). The HP gas is recycled directly without desulfurization as the H₂S concentration of HP gas meets the inlet requirements of the hydrogenation unit, and the desulfurized LP gas is sent to the PSA in hydrogen production plant. The product gas streams from PSAs in the CCR unit and

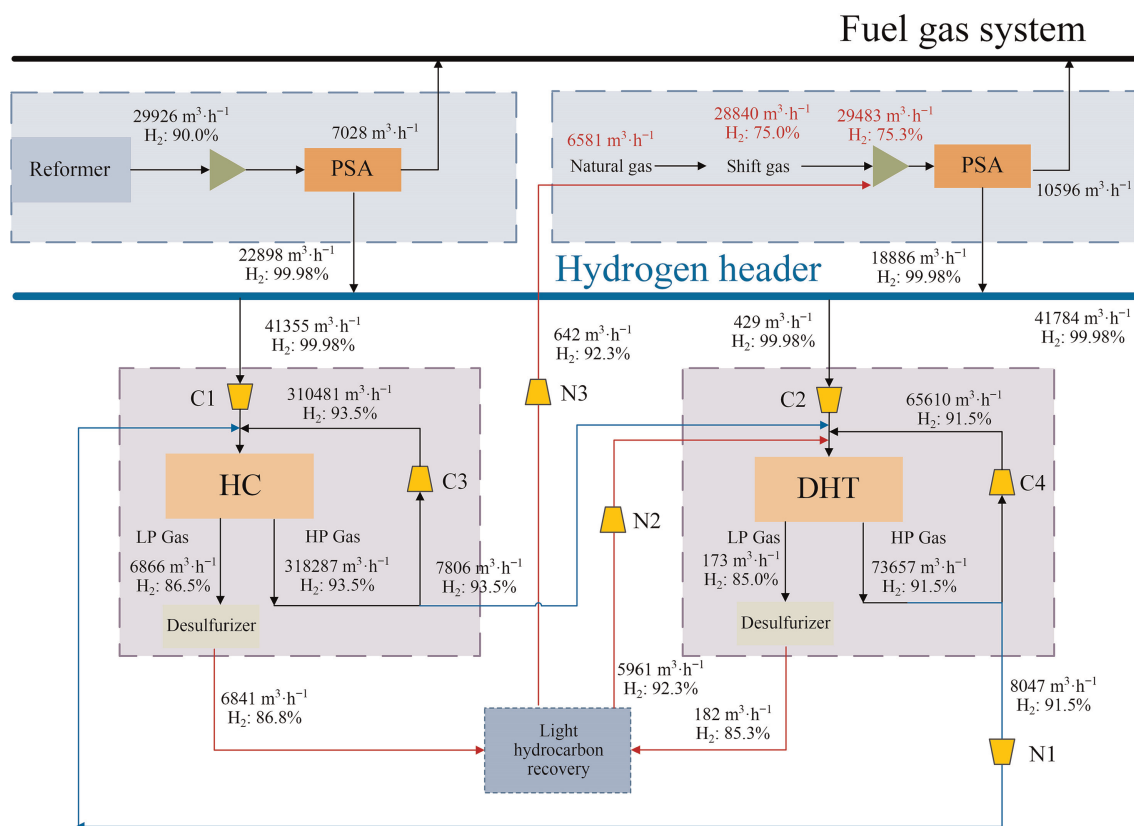


Fig. 7. Fuzzy optimization design of the hydrogen network with integrated light hydrocarbon recovery (Scenario 2b).

Table 8
Cost comparison of Scenario 2

	Preliminary design	Scenario 2a (non-fuzzy optimization)	Scenario 2b (fuzzy optimization)
Natural gas/ $\text{m}^3\cdot\text{h}^{-1}$	6623	6014	6581
Hydrogen utility/ $\text{m}^3\cdot\text{h}^{-1}$	23794	16807	18886
Fuel system/ $\text{m}^3\cdot\text{h}^{-1}$	18973	16127	17624
Compressor investment $\times 10^{-3}/\text{USD}$	–	7495.50	6565.33
Pipeline investment cost $\times 10^{-3}/\text{USD}$	–	889.56	998.59
LHR unit investment cost $\times 10^{-3}/\text{USD}$	–	1518.49	2974.09
Total investment cost $\times 10^{-3}/\text{USD}$	–	9903.54	10538.00
Hydrogen production cost $\times 10^{-3}/\text{USD}$	26595.02	18784.95	21109.61
Electricity cost $\times 10^{-3}/\text{USD}$	6871	6901.30	7216.88
PSA operating cost $\times 10^{-3}/\text{USD}$	1643.3	1391.56	1520.86
Fuel benefit $\times 10^{-3}/\text{USD}$	2005.63	1696.66	2037.33
Operation costs of LHR $\times 10^{-3}/\text{USD}$	–	498.93	1141.09
Benefit of LHR $\times 10^{-3}/\text{USD}$	–	1021.44	2336.16
Total operating cost $\times 10^{-3}/\text{USD}$	33103.69	24858.64	26614.96
TAC $\times 10^{-3}/\text{USD}$	33103.69	27146.36	29049.24
Payback period/a		1.07	1.19

hydrogen production plant are sent to the hydrogen header and then allocated to hydrogenation units. The flowrates of hydrogen utility and shift gas in hydrogen production plant are $23794 \text{ m}^3\cdot\text{h}^{-1}$ and $29025 \text{ m}^3\cdot\text{h}^{-1}$, and the consumed natural gas as material for hydrogen production is $6623 \text{ m}^3\cdot\text{h}^{-1}$.

The process data for the preliminary design of the refinery hydrogen system is listed in Table 1, and the data for the compressor and purifier (PSA) are shown in Table 2.

5. Results and Discussion

In this paper, the optimal utilization of HP gas and LP gas are considered. For model P1, the data is taken from the average value of the preliminary design of the refinery plant. For model P2, the upper and lower bounds of the flowrate are estimated by the preliminary design of the refinery plant.

Scenario 1 optimal design of hydrogen network with direct reuse of desulfurized LP gas stream

Scenario 1a Optimization design of hydrogen network

In Scenario 1, the desulfurized LP gas is utilized directly by other hydrogenation units while the HP gas is fully recycled. The minimization of TAC is taken as the objective function, and the hydrogen network can be obtained from the calculation results, as shown in Fig. 3.

The optimal results in Scenario 1a show that the flowrate of hydrogen utility is $22610 \text{ m}^3\cdot\text{h}^{-1}$, a decrease of $1183.85 \text{ m}^3\cdot\text{h}^{-1}$ is reached when it is compared with the preliminary design. The amount of natural gas for hydrogen production is $6511 \text{ m}^3\cdot\text{h}^{-1}$, which is $111.84 \text{ m}^3\cdot\text{h}^{-1}$ less than that for the preliminary design, and the reduction ratio is 1.69%. The desulfurized LP gas stream is directly reused in Scenario 1a. As a result, the flowrates of natural gas is reduced.

However, to satisfy the pressure level, two new compressors are needed. The investment cost of the new compressors is shown in Table 3. In addition, the relevant pipeline investment costs are estimated to be $106.41 \times 10^3 \text{ USD}$. The TAC is calculated as $32026.07 \times 10^3 \text{ USD}$, including $31784.71 \times 10^3 \text{ USD}$ for operation cost and $1044.88 \times 10^3 \text{ USD}$ for investment, $1077.62 \times 10^3 \text{ USD}$ less than TAC of preliminary design. The investment recovery period is 0.79a.

Scenario 1b Fuzzy optimization design of hydrogen network

In this scenario, the fuzzy design of the current hydrogen system is developed. In the actual operation process of the hydrogen system in the refinery, each stream is in variation. The concentration of each component is still the value in Table 1, and the flowrate variation is considered. To ensure that the optimal design of the hydrogen system can be applied to the flowrate variation in

actual operating conditions, the fuzzy interval of hydrogen sources and sinks estimated by the preliminary design of the refinery plant are listed in Table 4.

The model P2 is solved with the overall satisfaction of the hydrogen network as the objective function. Fig. 4 shows the hydrogen network with fuzzy optimization design. As shown in Fig. 4, the flowrate of natural gas is $6599 \text{ m}^3\cdot\text{h}^{-1}$, and the corresponding level of satisfaction of the fuzzy limits is determined as $\lambda = 0.6268$. When λ is at the optimal value, the optimal flowrate of the hydrogen-rich stream from CCR is deduced to be $29986 = (35000 + 0.6268 \times (27000 - 35000)) \text{ m}^3\cdot\text{h}^{-1}$, and this result is risky. The inlet flowrate of HC is $359474 \text{ m}^3\cdot\text{h}^{-1} = (325000 + 0.6268 \times (380000 - 325000)) \text{ m}^3\cdot\text{h}^{-1}$, and the HP gas from HC is $318660 \text{ m}^3\cdot\text{h}^{-1} = (350000 + 0.6268 \times (300000 - 350000)) \text{ m}^3\cdot\text{h}^{-1}$, and the flowrate to fuel system is $19083 \text{ m}^3\cdot\text{h}^{-1}$. The cost comparison of Scenario 1 is shown in Table 5. Corresponding to the non-fuzzy optimization result in Scenario 1a, the consumption of natural gas for hydrogen production is $6511.08 \text{ m}^3\cdot\text{h}^{-1}$. The hydrogen network connection of Scenario 1b is the same as non-fuzzy optimization Scenario 1a, with a total investment of $978.95 \times 10^3 \text{ USD}$ for the new equipment, which is $65.93 \times 10^3 \text{ USD}$ less than the result in Scenario 1a. However, the operating cost has slightly increased. Overall, the TAC is $32230.13 \times 10^3 \text{ USD}$, which is $873.13 \times 10^3 \text{ USD}$ less than that of the preliminary design. The payback period of investment costs is 0.89 years.

Among the preliminary design schemes and Scenario 1a and 1b, the amount for natural gas in preliminary design is the largest. And the consumption of natural gas is reduced in Scenario 1a and 1b with the direct reuse of desulfurized LP gas. However, to meet the distribution and pressure requirements of the optimal hydrogen network, the investment costs need to be increased. Due to the penalty brought by the fuzzy interval constraint, the amount of natural gas and the TAC in Scenario 1b are greater than those in Scenario 1a, but still less than those in the preliminary design.

Scenario 2 optimal design of hydrogen network integrated light hydrocarbon recovery with direct reuse of desulfurized HP and LP gas streams

Scenario 2a Optimal design of hydrogen network

In Scenario 2, the minimization of the TAC is taken as the objective function, considering the direct utilization of LP gas and HP gas. Generally, the light hydrocarbon components are the excellent raw material for the high-value chemical products. For example, ethane and propane are the raw material of ethylene and propylene, which can generate economic benefits. If the light hydrocarbon is burned as fuel, it may lead to a waste of resources. As shown in Table 1, the concentration of C_{2+} in the LP gas produced by the HC and DHT is 5.91% and 2.36%, respectively, and it indicates

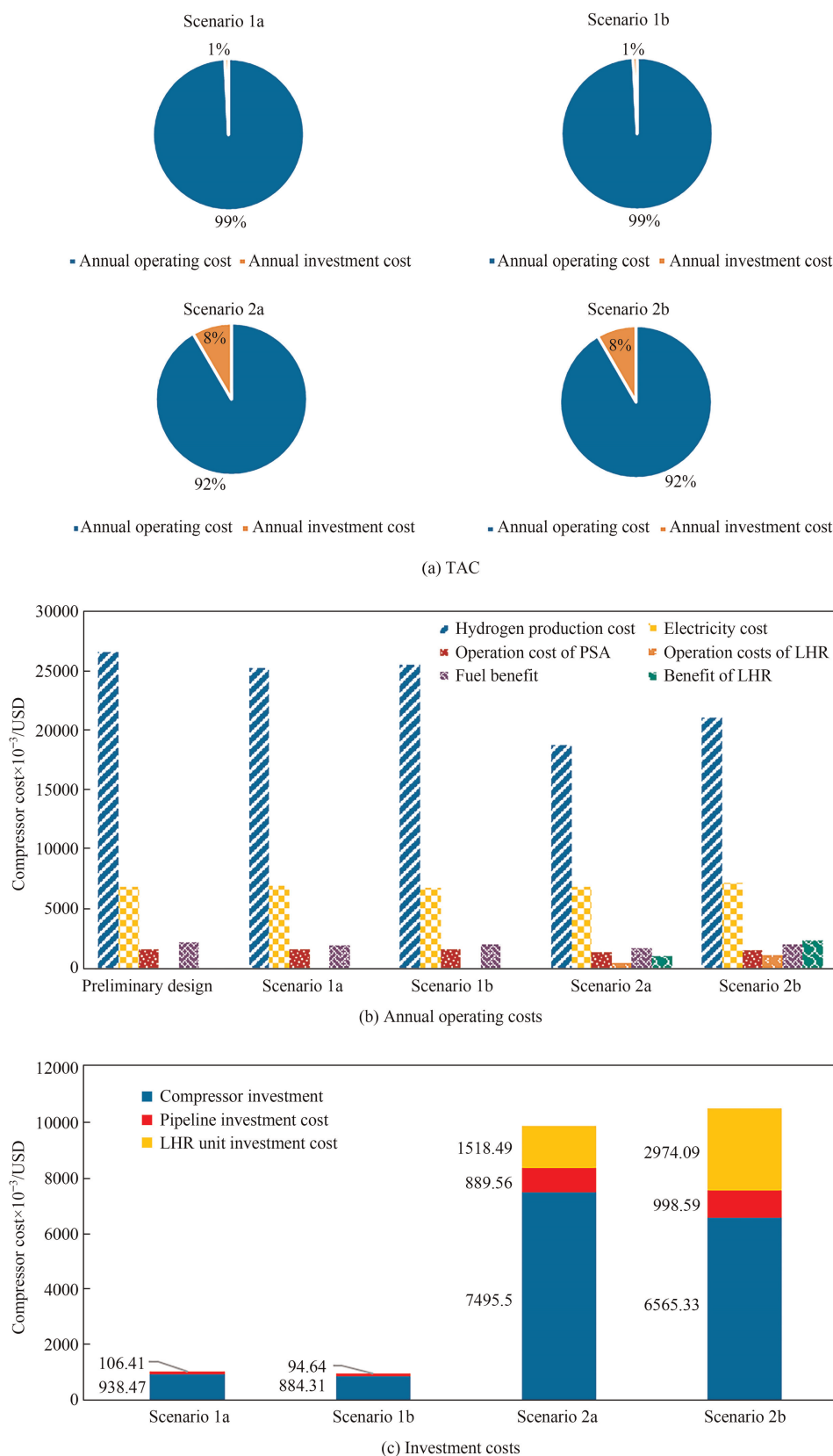


Fig. 8. Cost comparison: (a) TAC; (b) Annual operating costs; (c) Investment costs.

the potential for light hydrocarbon recovery. Thus, the two streams can be considered to recover the light hydrocarbon.

It can be seen from the hydrogen network shown in Fig. 5, all the HP gas from HC is directly recycled, while the HP gas from

DHT is partially recycled and partly treated as a process hydrogen source to HC. The flowrate of desulfurized LP gas from HC sent to the light hydrocarbon recovery unit is $3028 \text{ m}^3 \cdot \text{h}^{-1}$. The desulfurized LP gas from DHT is direct to HC, while the desulfurized LP

gas from HC is sent to the LHR unit to recover light hydrocarbon, then to DHT. And the mass flowrate of recovered light hydrocarbon is determined as $480 \text{ kg} \cdot \text{h}^{-1}$. In the optimal design Scenario 2a, the flowrate of hydrogen utility is $16807 \text{ m}^3 \cdot \text{h}^{-1}$, a reduction of 29.36% compared with the preliminary design. The corresponding consumption of natural gas is $6014 \text{ m}^3 \cdot \text{h}^{-1}$, 9.18% less than that in the preliminary design, and 7.62% less than in Scenario 1a. The direct reuse of HP gas and desulfurized LP gas and the recovery of light hydrocarbons are considered in Scenario 2a, so the flowrates of natural gas and discharged to the fuel system are reduced.

It can be seen from Fig. 6 that the direct reuse of HP and desulfurized LP gas can effectively reduce the flowrate of natural gas and hydrogen utility. The optimal design of Scenario 2a not only considers the direct reuse of HP gas and desulfurized LP gas but also recovers the light hydrocarbon components of the LP gas from DHT. The operating pressure of the new light hydrocarbon recovery unit is 1.05 MPa, the investment cost is 1518.49×10^3 USD, and the benefit of recovered light hydrocarbons is 1021.44×10^3 USD per year. At the same time, to meet the pressure constraints, four new compressors are needed. The investment cost of the compressors is shown in Table 6, and the related pipeline investment costs must also be considered. The TAC is reduced by 5957.33×10^3 USD compared with the preliminary design, which is 27146.36×10^3 USD, and the investment payback period is 1.07 years.

Scenario 2b Fuzzy optimization design of hydrogen network

In Scenario 2b, a fuzzy optimization design of the hydrogen network integrated with light hydrocarbon recovery is developed. And the fuzzy interval of the stream is the same as that in Table 4 in Scenario 1b. The result of hydrogen network with fuzzy optimization is shown in Fig. 7.

In this Scenario, the HP gases from HC and DHT are both divided into two streams, one of which is directly recycled, and the other stream is sent to other hydrogenation units as a process hydrogen source. The desulfurized LP gas is sent to the light hydrocarbon recovery unit. Then, the outlet hydrogen-rich stream of the light hydrocarbon recovery unit is divided into two parts, one part is sent to the PSA, and the other part is allocated to the DHT. The investment cost of new compressors is shown in Table 7.

The cost comparison of Scenario 2 is shown in Table 8. The overall satisfaction degree that satisfies the fuzzy constraints is $\lambda = 0.6343$, then the optimal flowrate for hydrogen-rich stream in CCR can be determined as $29926 \text{ m}^3 \cdot \text{h}^{-1} = (35000 + 0.6343 \times (27000 - 35000)) \text{ m}^3 \cdot \text{h}^{-1}$, and this result is risky. When the satisfaction λ is at the optimal value, the inlet flowrates of HC and DHT can be calculated to be $359884 \text{ m}^3 \cdot \text{h}^{-1}$ and $79806 \text{ m}^3 \cdot \text{h}^{-1}$ respectively, the results of the hydrogenation unit are more conservative (or low risk). The flowrate of natural gas is determined as $6581 \text{ m}^3 \cdot \text{h}^{-1}$ more than that in Scenario 2a. The TAC is estimated to be 29049×10^3 USD, which is 4054.45×10^3 USD less than the preliminary design, and the investment cost recovery period is 1.19a (Table 8).

In the two design schemes of Scenario 2, the total investment costs and the payback period are greater than those in Scenario 1. However, the reduced operating cost of the hydrogen system and the economic benefits of recovering light hydrocarbons result in a reduction of TAC.

It can be seen from Fig. 8 that the operating cost is much higher than the annualized investment cost, and the operating cost of the hydrogen production process accounts for the largest proportion of the operating cost, while the compressor accounts for the largest proportion of the investment cost. The TAC of four scenarios (Scenario 1a, 1b, 2a, 2b) is less than the preliminary design. In Scenario 2, the investment cost of the hydrogen network increases, but the TAC is reduced. Compared with non-fuzzy optimal design, the TAC of fuzzy optimal design increases due to the penalty caused by

fuzzy interval constraints. However, the fuzzy optimization design may be more suitable for the actual operating, and the TAC is still lower than that of the preliminary design.

6. Conclusions

In this paper, a multi-component hydrogen network superstructure integrated light hydrocarbon recovery is proposed. Based on the superstructure, two mathematical models (non-fuzzy and fuzzy optimization model) are developed with minimal TAC and maximum overall satisfaction hydrogen network as objective functions, respectively. Four components (*i.e.*, H_2 , H_2S , CH_4 , and C_{2+}), desulfurization and light hydrocarbon recovery units are included in the models. A simplified industrial case analysis to verify the proposed mathematical model. Four scenarios are discussed to explore the impact of different utilization of HP gas and LP gas streams on the hydrogen system structure and performance (flowrate of hydrogen utility and natural gas, economic, fuzzy level of satisfaction). The results show that the optimal design that includes the direct reuse of HP gas and desulfurized LP gas and integration light hydrocarbon recovery can maximize the economic benefits. Moreover, the fuzzy optimization model is applicable to the flowrate variation in the actual operation of the hydrogen system. It can be used to guide the optimal design of hydrogen systems and achieve a balance of hydrogen savings and the risks associated with flowrate variation. In the future work, the source-sink correlation model and surrogate-based models of separation units would be included in the mathematical model.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Nomenclature

Sets

C	set of components
F	set of fuel systems
H	set of hydrogen header
I	set of process hydrogen sources
K	set of hydrogen sinks
P	set of purifier
R	set of Light Hydrocarbon Recovery
U	external hydrogen sources

Parameters

A_f	annualized factor of equipment investment cost
a_n	investment cost coefficients of compressor n
a_{pipe}	correlation coefficients
ar	raw material cost parameter of light hydrocarbon recovery unit
b_n	investment cost coefficients of compressor n
b_{pipe}	correlation coefficients
br	energy parameter of the light hydrocarbon recovery unit
C_{RO}	cost correlation coefficient of light hydrocarbon recovery unit
e_{H_2}	the unit hydrogen production cost

e_{heat}	the benefit generated by fuel gas
e_{ng}	price of natural gas
e_{power}	the unit price of power
e_{PSA}	operating cost coefficient of PSA
F_i^L	lower limit of the stream flowrate of process hydrogen source
F_i^U	upper limit of the stream flowrate of process hydrogen source
F_k^L	lower limit of the stream flowrate of the process hydrogen sink
F_k^U	upper limit of the stream flowrate of the process hydrogen sink
F_{RO}	cost correlation coefficient of light hydrocarbon recovery unit
F_u^L	lower limit of the stream flowrate of the external hydrogen sources
F_u^U	upper limit of the stream flowrate of the external hydrogen sources
ny	the number of years of depreciation
P_a	the outlet pressure levels of supplier a
P_b	the outlet pressure levels of receiver b
$R_{k,c}^{\text{DS}}$	desulfurization rate
$R_{p,c}$	hydrogen recovery ratio of PSA unit
t	annual operating time
$\gamma_{k,c}^{\text{min}}$	the minimum concentration requirement of component c
$\gamma_{k,c}^{\text{max}}$	the maximum concentration requirement of component c
$\Delta H_{c,\text{CH}_4}^0$	the enthalpy of natural gas combustion under standard conditions
$\Delta H_{c,\text{H}_2}^0$	the enthalpy of hydrogen combustion under standard conditions
β	conversion coefficient

Variables

C_{LHR}	investment cost of the light hydrocarbon recovery unit
C_n	investment cost of the compressor
C_{pipe}	investment cost of the pipeline
F_k^{out}	outlet flowrate of the hydrogenation unit
f_{Fuel}	inlet flowrate of the fuel system
f_h^{in}	inlet flowrate of the hydrogen header
f_h^{out}	outlet flowrate of the hydrogen header
$f_{hk,h,k}$	flowrate from hydrogen header to hydrogenation unit
f_i	flowrate of process hydrogen source
$f_{if,i,f}$	flowrate of the process hydrogen source to the fuel system
$f_{ik,i,k}$	flowrate of the process hydrogen source to the hydrogenation unit
$f_{ip,i,p}$	flowrate of the process hydrogen source to the purifier
f_k^{in}	inlet flowrate for the hydrogenation unit
f_p^{in}	inlet flowrate of the purifier p
f_p^{prod}	product flowrate of PSA
f_p^{resd}	residual flow rate of the PSA
$f_{pf,p,f}$	the flow rate of PSA discharged to the fuel gas system
$f_{ph,p,h}$	the flow rate of PSA to the hydrogen network
f_r^{out}	outlet flowrate of the light hydrocarbon recovery unit
$f_{rf,r,f}$	flowrate of the light hydrocarbon recovery unit sent to the fuel system
$f_{rk,r,k}$	flowrate of the light hydrocarbon recovery unit sent to other hydrogenation units
$f_{rp,r,p}$	flowrate of the light hydrocarbon recovery unit sent to the purifier
f_u	flowrate of shift gas from the hydrogen production plant
$f_{up,u,p}$	flowrate of shift gas allocated to purifier p
f_p^{in}	inlet flowrate of the purifier p
$f_{ip,i,p}$	flowrate from process hydrogen sources i to purifier p

$f_{rp,r,p}$	flowrate from outlet streams of Light Hydrocarbon Recovery r to purifier p
$f_{u,ng}$	flowrate of natural gas for hydrogen production
$f_{up,u,p}$	flowrate of shift gas allocated to purifier p
OC_{fuel}	operating costs of the fuel system
OC_{H_2}	operating costs of the hydrogen utility
OC_{LHR}	operating costs of the light hydrocarbon recovery unit
OC_{Power}	operating costs of the compressor
OC_{PSA}	operating cost of the PSA
Power_n	compression work
$y_{h,c}$	concentration of component c of the hydrogen header
$y_{i,c}$	concentration of component c of the process hydrogen source
$y_{k,c}^{\text{in}}$	inlet concentration of component c of the hydrogenation unit
$y_{p,c}^{\text{in}}$	inlet concentration of component c of PSA
$y_{r,c}^{\text{out}}$	outlet concentration of component c of the light hydrocarbon recovery unit
$y_{p,c}^{\text{prod}}$	the product flowrate of PSA
$y_{p,c}^{\text{resd}}$	the residual flow rate of the PSA
$z_{a,b}$	connection variable from supplier a and the receiver b
$z_{a,b}^{\text{comp}}$	connection variable for compressors from supplier a and the receiver b
$z_{a,b}^{\Delta P}$	connection variable for pressure difference from supplier a and the receiver b
ζ_{TAC}	total annualized cost of the hydrogen system

Subscripts and Superscripts

c	index for component
DS	desulfurization
f	index for fuel system
h	index for hydrogen header
i	index for process hydrogen source
in	inlet
k	index for hydrogen sink
LB	lower bound
out	outlet
p	index for purifier
prod	product gas
r	index for Light Hydrocarbon Recovery
resd	residual gas
UB	upper bound
u	index for external hydrogen sources

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