Journal of Cleaner Production 18 (2010) 889–899

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Modelling and optimisation for design of hydrogen networks for multi-period operation

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ARTICLE INFO

Article history:
Received 13 August 2009
Received in revised form
4 January 2010
Accepted 5 January 2010
Available online 13 January 2010

Keywords:
Hydrogen management
Multi-period
Hydrogen network
Design
Optimisation

ABSTRACT

Hydrogen management is a problem of increasing importance: hydrogen consumption of refineries is rising sharply with additional capacities of hydrocracking and hydrotreating units in order to comply with cleaner fuel specifications. Product Specifications for transportation fuels are becoming increasingly stringent to ensure production of environmentally more benign fuels. Hydrogen management techniques currently do not account for varying operating conditions of hydrogen consuming processes and assume constant operating conditions. A novel approach is developed for the design of flexible hydrogen networks that can remain optimally operable under multiple periods of operation. The proposed methodology for multi-period design of hydrogen networks can take into account pressure differences, maximum capacity of existing equipment, and optimal placement of new equipment such as compressors.

1. Introduction

Petroleum refineries include many processes involving hydrogen production or consumption. Hydrogen management aims to achieve the optimal allocation of hydrogen resources in order to satisfy the demands of refinery processes. Hydrogen management is of interest to refiners as the cleaner fuel specifications require refineries to increase hydrogen production capacity and to increase the severity of operating conditions of hydrotreating and hydrocracking units in order to produce environmentally more benign fuels such as ultra low sulphur diesel. The product specifications for gasoline require reduction in the aromatics content of gasoline, resulting in lesser hydrogen being available from the catalytic reforming unit which has traditionally been the most cost efficient source of hydrogen (Alves and Towler, 2002). The capital costs of hydrogen production units may be equivalent to more than one-third of the capital costs of hydrocarbon conversion processes for upgrading heavy petroleum fractions (Peramanu et al., 1999). Therefore, hydrogen management has become essential to assure economically optimal performance of refineries.

The operating conditions of refinery processes are changed periodically to compensate for catalyst deactivation. This periodic change in operating conditions, in order to maintain satisfactory performance of a process while meeting the product specifications and market demands, is termed as multi-period operation. Multi-period operation needs to be taken into account in the design of hydrogen networks because refinery processes that consume hydrogen are operated under multiple periods of operation. This paper presents a novel approach for the design of hydrogen networks that can remain optimally operable under multiple periods of operation. The design approach is underpinned by mathematical models of hydrogen network and its components, and an optimisation framework that account for the dominant contributions to total annualised costs of a hydrogen network.

A hydrogen network may be described as a system of refinery processes that interact with each other through distribution of hydrogen. A typical refinery hydrogen network is shown in Fig. 1.

These refinery processes may be classified into two categories, i.e. hydrogen producers and hydrogen consumers, based on their contribution to the hydrogen network.

Hydrogen producers are units that supply hydrogen to the hydrogen distribution system, such as the hydrogen plant and catalytic reforming unit. The catalytic reforming process produces hydrogen as a by-product of cyclisation and dehydrogenation reactions of hydrocarbon molecules to increase the aromatic content and the octane number of naphtha products (Meyers, 1997). Hydrogen plants may employ steam reforming or partial oxidation for the production of high purity hydrogen through conversion of light hydrocarbon fractions such as refinery off-gases and light naphtha.

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0959-6526/$ – see front matter
doi:10.1016/j.jclepro.2010.01.003

Nomenclature

a	capital cost coefficient
AF	annualisation factor
b	capital cost coefficient
c	coefficient to calculate compression power
Cost	capital or operating cost
F	feed flow rate for distillation column, also used to represent flow rate of hydrogen stream (MMscfd or t/h)
F^*	constant flow rate (MMscfd or t/h)
Fy	variable representing a bilinear term i.e. product of flow rate and purity
HV	lower heating value of combustion
I	capital cost
If	integer variable to identify the existence of equipment or piping
Length	length of the pipeline between a source and a sink
n	number of years
oy	annual operating hours
P	pressure (MPa)
PI	price per unit quantity
Power	compressor power (kW)
u	a lower bound. Can be a small positive number
Up	upper bound. Can be a large positive number
y	hydrogen purity (volume or mass basis)
y^*	constant hydrogen purity (volume or mass basis)

Abbreviations:

AH	aromatic hydrogenation
CCR	continuous catalytic reformer
CNHT	cracked naphtha hydrotreater
CPROC	combined process sink
CRU	catalytic reformer unit
DHT	diesel hydrotreater
HCU	hydrocracker unit
HDA	hydrodealkylation unit
ISOM	isomerisation process
KHT	kerosene hydrotreater
NHT	naphtha hydrotreater
NLP	non-linear programming
TAC	total annualised cost

Indices:

comp	a compressor between a source and a sink
Compv	a compressor source
Compw	a compressor sink
p	period of operation
pipe	a pipeline between a source and a sink
Sk	a sink decomposed from a hydrogen consumer
SkFixed	a sink with fixed pressure
SkVaried	a sink with variable pressure
Sr	a source decomposed from a hydrogen consumer
SrFixed	a source with fixed pressure
SrVaried	a source with variable pressure
v	a source in the hydrogen network
w	a sink in the hydrogen network

Subscripts:

COMP	a compressor which can be decomposed into a sink *Compw* and a source *Compv*
fuel	fuel gas system
H_2U	hydrogen utility
HV	lower heating value of combustion
M	make-up gas
P	purge gas from a hydrogen consumer
PIPE	pipe
R	recycle gas for a hydrogen consumer

Superscripts:

*	constant flow rate or purity
dp	pressure difference between source and sink is greater than zero
Low	lower bound
Max	maximum value
Up	upper bound

Hydrogen consumers are conversion processes, such as the hydrocracking process for upgrading heavy hydrocarbon fractions, hydrotreating processes to satisfy cleaner fuel specifications, lubricant plants, the isomerisation process, and the hydro dealkylation units. These processes employ hydrogen as a reactant to upgrade the quality of refinery products. Amongst all the hydrogen consuming processes, hydrocracking and hydrotreating processes are the major hydrogen consumers (Liu, 2002).

Apart from the above mentioned refinery processes, compressors and purification units also form part of a refinery hydrogen network. Purification processes help recover hydrogen from refinery off-gases, i.e. the purge streams from hydrogen consuming processes. Compressors do not influence a hydrogen network in terms of the demand and supply of hydrogen. However, they are needed to ensure that pressure requirements of refinery processes and of the hydrogen network are met.

2. Review of previous research on hydrogen management

Hydrogen management is of increasing importance because of the contribution of hydroprocessing operations to the overall performance of modern refineries. The need for hydrogen management strategies for the design and operation of refinery processes was first acknowledged by Simpson (1984). This section reviews the methodologies developed previously for refinery hydrogen management.

2.1. Cost and value composite curves for hydrogen management

The first systematic approach for the assessment of hydrogen resources of a hydrogen network was developed by Towler et al.

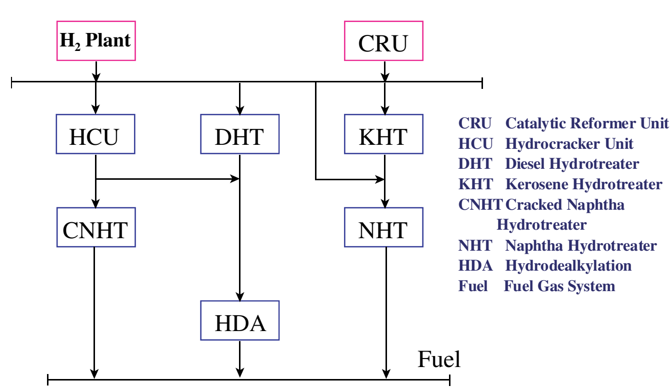

Fig. 1. A typical hydrogen network (Ahmad, 2009).

(1996). In this approach, cost and value composite curves are generated for refinery processes that produce hydrogen or consume hydrogen. The cost of hydrogen available from the hydrogen plant is dependant on the price of the feedstock of hydrogen plant. The value added to refinery products in hydrogen consuming processes is the value of the products of a refinery process less the cost of its feed. The cost of hydrogen recovery from a hydrogen source stream is calculated using the cost of purification. Using this concept of cost and value the driving force for hydrogen transfer may be defined as the difference between the cost of hydrogen available and the value added to refinery products (Towler et al., 1996).

The cost and value composite curves may be used for the economic analysis of a refinery hydrogen network. However, this approach does not provide a systematic method for the retrofit or design of hydrogen networks. The analysis relies on the availability of economic data, such as the value added to refinery products per unit hydrogen consumption which may not always be available, especially for design scenarios. Another major drawback of this approach is that it does not provide a systematic framework for screening design options such as the choice of streams to purify, the degree of purification, the selection of purification processes, and their placement within the hydrogen network.

2.2. Hydrogen pinch analysis and design of hydrogen networks

Hydrogen pinch analysis was developed by Alves (1999) using an analogy to pinch analysis for heat exchanger networks (Linnhoff et al., 1979). Hydrogen pinch analysis is a tool for the estimation of the minimum hydrogen requirement of a hydrogen network before the system design.

Hydrogen pinch analysis is developed using the following assumptions (Alves, 1999; Alves and Towler, 2002):

- Constant operating conditions of the refinery processes constituting the hydrogen network.
- Hydrogen streams in the hydrogen network are binary mixtures of hydrogen and methane.
- Any hydrogen source may supply any hydrogen sink if the hydrogen purity of the source is higher than that of the sink. Pressure differences are ignored.

Hydrogen pinch analysis requires the flow rate and purity constraints of the network, i.e. the overall mass balance and the hydrogen mass balance, to be satisfied for a given hydrogen network.

Alves (1999) defined the concepts of sink and source to classify components of a hydrogen network. A sink is a stream that consumes hydrogen from the hydrogen network while a source is defined as a stream supplying hydrogen to the system. The hydrogen producing processes, with their given purity and pressure levels, form the sources in a hydrogen network. The hydrogen consuming processes are represented in the framework of sinks and sources as shown in Fig. 2.

The net hydrogen-rich gas fed to the hydrogen consuming process, i.e. the mixture of the recycle stream from the process and the hydrogen make-up stream, is represented as the sink. The stream that is partly recycled and partly purged, in order to prevent the build-up of hydrocarbons in the recycle stream, is represented as the source. The flow rate and purity of the process source and sink are calculated using the data available for the process purge, recycle and make-up streams.

Apart from hydrogen producing and consuming processes, other components of a hydrogen network are also classified in terms of sources and sinks. Purification units consist of one hydrogen sink, i.e. the feed stream, and two hydrogen sources, the product and residue streams. Compressors consist of one sink and one source, i.e. the inlet and outlet streams.

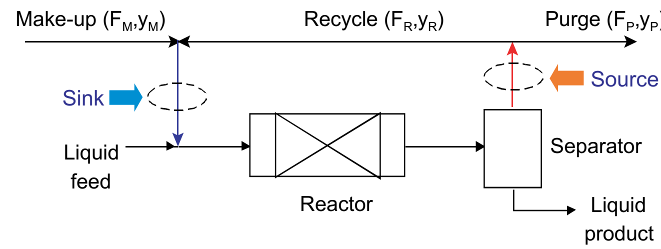

Fig. 2. Simplified representation of a hydrogen consuming process in hydrogen pinch analysis (Alves, 1999).

The hydrogen pinch represents the minimum amount of hydrogen required for the feasible operation of a hydrogen network. The hydrogen pinch also represents a bottleneck to the savings that can be made by reducing the hydrogen utility. One of the debottlenecking options discussed by Alves (1999) is the utilisation of purification processes to increase the purity of one or more hydrogen sources. Alves (1999) discussed the three possible options for placement of a purification unit for a given hydrogen network using hydrogen surplus diagram for graphical representation. Alves (1999) demonstrated that purification across the hydrogen pinch provides the highest reduction in the required amount of hydrogen utility.

The approach proposed by Alves (1999) for the design of hydrogen networks applies the overall mass balance and hydrogen balance to each hydrogen source and sink. The objective function to be minimised is the total cost of the design of a given hydrogen network, i.e. cost of hydrogen from the sources, distribution cost and the value of the gas sent to the fuel gas system (Alves, 1999). The design problem is formulated as a linear programming problem using linear cost correlations. This approach, based on the concept of hydrogen surplus and linear programming, has been successfully applied to address the problem of hydrogen distribution at the Porto Refinery of the GALP ENERGIA network by Fonseca et al. (2008).

Hydrogen pinch analysis with its graphical representation of the sources and sinks provides insights to a hydrogen network, and allows the minimum hydrogen utility required for feasible operation of the system to be estimated. In hydrogen pinch analysis gas streams are treated as binary mixtures of hydrogen and methane. This simplification of representing all impurities in a hydrogen stream, e.g. light hydrocarbons, sulphur and nitrogen compounds, by methane may lead to discrepancies in calculations (Zhang et al., 2008). Another limitation of hydrogen pinch analysis is that pressure differences between source and sink streams are ignored. Hydrogen pinch analysis assumes that any source may supply hydrogen to any sink if the purity of the source is higher than that of the sink. The estimated minimum amount of hydrogen utility required to satisfy the demands of a hydrogen network using this assumption may only be achieved at the expense of extra compression (Hallale and Liu, 2001).

2.3. Automated design of hydrogen networks

Hallale and Liu (2001) developed a superstructure based optimisation approach for the design of hydrogen networks addressing the limitations of hydrogen pinch analysis. For example, their approach incorporates pressure constraints and takes into account existing compressors for retrofit scenarios. The automated design approach (Liu, 2002; Liu and Zhang, 2004) also provides a strategy for selection of purification processes and their integration in hydrogen networks. The design problem is formulated as mixed integer non-linear programming and handles various objective functions, such as minimising the hydrogen utility, operating costs or the total annualised costs of the hydrogen network.

2.3.1. Superstructure based hydrogen network optimisation

In the approach of Hallale and Liu (2001), the components of a hydrogen network are represented as sources and sinks. A superstructure may be set up to allow connections between all the sources and all the sinks. In order to account for pressure constraints this superstructure is reduced by eliminating those connections where the source pressure is less than the sink pressure (Liu, 2002). Existing compressors are incorporated in the superstructure consisting of one sink, i.e. the compressor inlet and one source, i.e. the compressor outlet. The inlet and outlet pressures of existing compressors are fixed at their design values. New compressors may be included in the superstructure similarly. However, the inlet and outlet pressures of new compressors are not known in advance and become optimisation variables. The objective function is chosen to be the total annualised cost, i.e. the sum of annualised capital cost and annual operating cost, for optimum design of hydrogen networks. The approach of Hallale and Liu (2001) has been adapted by Khajehpour et al. (2009) for efficient hydrogen management of Tehran North Refinery, Iran. Khajehpour et al. (2009) proposed heuristic rules to reduce the superstructure of a given hydrogen network, and employed a genetic algorithm for optimisation.

2.3.2. Integration of purification processes in hydrogen network

Purification processes provide economical means of recovering hydrogen from refinery off-gases (Peramanu et al., 1999). Liu and Zhang (2004) developed shortcut models for pressure swing adsorption, and membrane separation processes for recovering hydrogen from refinery off-gases. A detailed account of the purification processes employed in refinery hydrogen networks can be found elsewhere (Miller and Stoecker, 1989; Spillmann, 1989; Winston and Sirkar, 1992; Ruthven et al., 1994). Liu and Zhang (2004) proposed a systematic methodology for selection of appropriate purification processes and their integration in design and optimisation of hydrogen networks.

A similar approach, for incorporating purification processes in hydrogen networks, has been proposed by Liao et al. (2010) successfully demonstrated the application of superstructure based approach for retrofit design of an existing refinery near Shanghai, China.

The automated design approach of Liu and Zhang (2004) addresses some of the limitations of hydrogen pinch analysis and design approach by taking into account practical constraints such as pressure differences, compression and piping costs. Their approach incorporates purification processes to improve the scope of reduction of hydrogen utility. However, there are two limitations of the automated design approach. The first limitation arises due to the simplification of treating hydrogen streams as binary mixtures. The impact of changes in composition of hydrocarbon compounds of recycle and purge streams from hydrogen consuming processes, on process performance parameters such as hydrogen to oil ratio cannot be captured if hydrogen streams are treated as binary mixtures of hydrogen and methane (Zhang et al., 2008). The second limitation of the automated design approach is the assumption of constant operating conditions for all refinery processes constituting a hydrogen network. Because of this assumption, the automated design approach (Hallale and Liu, 2001; Liu, 2002; Liu and Zhang, 2004) may help reduce the hydrogen utility demand but does not provide a framework for analysing the optimal utilisation of the possible savings of hydrogen surplus (Hallale et al., 2002).

A new methodology for design of hydrogen networks is developed in this work to account for changing operating conditions of refinery processes and to consider the impact of such changes on the operation and performance of the hydrogen network.

3. Multi-period design of hydrogen networks

Changing operating conditions of refinery processes may influence the performance of a hydrogen network. For example, changes in composition of feedstock, operating temperature, and pressure result in varying conversion of reactions. Consequently, the hydrogen consumption, flow rate, and purity of recycle and purge streams of hydrogen consuming processes may also change. Hydrogen networks designed using data for one set of process conditions, i.e. a single-period data, may not satisfy the requirements of hydrogen consuming processes when the operating conditions of one or more refinery processes are changed.

The methodology developed in this work for multi-period design of hydrogen networks is an extension of the automated design approach of Hallale and Liu (2001) and Liu and Zhang (2004) for multiple periods of operation. The new methodology is limited by the underlying assumption of the automated design approach that hydrogen streams are represented by a binary mixture of hydrogen and methane. The methodology developed for multi-period hydrogen management is applicable to retrofit and new design of flexible hydrogen networks, i.e. hydrogen networks that can operate under different operating conditions, with minimum total annualised cost. This paper presents the mathematical formulation and the optimisation framework developed for the multi-period design of hydrogen networks.

3.1. Process constraints

The design of hydrogen networks for multiple periods of operation requires process constraints to be satisfied for all the sources and sinks of a system for all the operating periods under consideration. Process constraints include the overall material and hydrogen balance which ensure that each hydrogen sink is supplied by a hydrogen-rich gas of sufficient flow rate and hydrogen purity in each operating period:

$$\sum_{\mathrm{v}} F_{\mathrm{v,Sk,p}} = F^{*}_{\mathrm{Sk,p}} \tag{1}$$

$$\sum_{\mathrm{v}} F_{\mathrm{v,Sk,p}} \cdot y_{\mathrm{v,p}} = F^{*}_{\mathrm{Sk,p}} \cdot y_{\mathrm{Sk,s}} \tag{2}$$

Similarly, the amount of gas supplied to all the sinks, including the fuel system, must be equal to the amount available from each hydrogen source:

$$\sum_{\mathrm{w}} F_{\mathrm{Sr,w,p}} = F^{*}_{\mathrm{Sr,p}} \tag{3}$$

where F represents the flow rate of a stream, y the hydrogen purity of a stream in weight fraction, the subscripts p, Sk, Sr, v, and w represent a period of operation, sink of a hydrogen consumer, source from a hydrogen consumer, a source in a given hydrogen network, and a sink in a given hydrogen network, respectively. The superscript * represents the total flow rate of a source or sink in a given hydrogen network.

3.2. Compressors

For multi-period design of hydrogen networks, the mathematical formulation for compressors is similar to that in the approach of Hallale and Liu (2001).

A compressor is incorporated in the superstructure as a sink and a source to represent the compressor inlet and outlet. The flow rate of the inlet stream of a compressor must be equal to the flow rate of the outlet stream for each operating period:

$$\sum_{w} F_{\mathrm{Compv,w,p}} = \sum_{v} F_{\mathrm{v,Compw,p}} \tag{4}$$

where the subscripts Compv and Compw represent a compressor source and a compressor sink respectively. Similarly, the amount of pure hydrogen entering a compressor must be equal to the amount of pure hydrogen leaving the compressor in each operating period:

$$\sum_{w} F_{\mathrm{Compv,w,p}} \cdot y_{\mathrm{Compv,p}} = \sum_{v} F_{\mathrm{v,Compw,p}} \cdot y_{\mathrm{v,p}} \tag{5}$$

The above formulation may also be used to represent new compressors with inlet and outlet pressures as variables. The inlet pressure of a new compressor may be assigned the lowest pressure over all the sources that feed the compressor, while the outlet pressure may be assigned the highest pressure over all the sinks that the compressor will feed (Hallale and Liu, 2001).

An additional constraint may be required for existing compressors as these machines would be designed for a maximum stream flow rate:

$$\sum_{v} F_{\mathrm{v,Compw,p}} \le F_{\mathrm{Compw}}^{\mathrm{Max}} \tag{6}$$

The compressor model used in this work is a shortcut model developed by Liu (2002) to predict the power demand of a compressor. This model for compressors is developed using the power and cost equations from Peters and Timmerhaus (1991).

The power required by a compressor is estimated using the following equations. It should be noted here that new compressors are represented with sources and sinks with unknown pressure while all other components of a hydrogen network are represented as sources and sinks with known pressure (Liu, 2002):

From a source with known pressure to a sink with known pressure:

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrFixed,SkFixed}}^{\mathrm{COMP}} - 1\right) \ge \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkFixed,p}}}{P_{\mathrm{SrFixed,p}}}\right) F_{\mathrm{SrFixed,SkFixed,p}} \tag{7}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrFixed,SkFixed}}^{\mathrm{COMP}} - 1\right) \le \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkFixed,p}}}{P_{\mathrm{SrFixed,p}}}\right) F_{\mathrm{SrFixed,SkFixed,p}} \tag{8}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}} \cdot \mathrm{If}_{\mathrm{SrFixed,SkFixed}}^{\mathrm{COMP}} \le 0 \tag{9}$$

From a source with known pressure to a sink with unknown pressure:

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrFixed,SkVaried}}^{\mathrm{COMP}} - 1\right) \ge \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkVaried,p}}}{P_{\mathrm{SrFixed,p}}}\right) F_{\mathrm{SrFixed,SkVaried,p}} \tag{10}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrFixed,SkVaried}}^{\mathrm{COMP}} - 1\right) \le \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkVaried,p}}}{P_{\mathrm{SrFixed,p}}}\right) F_{\mathrm{SrFixed,SkVaried,p}} \tag{11}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}} \cdot \mathrm{If}_{\mathrm{SrFixed,SkVaried}}^{\mathrm{COMP}} \le 0 \tag{12}$$

From a source with unknown pressure to a sink with known pressure:

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrVaried,SkFixed}}^{\mathrm{COMP}} - 1\right) \ge \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkFixed,p}}}{P_{\mathrm{SrVaried,p}}}\right) F_{\mathrm{SrVaried,SkFixed,p}} \tag{13}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(If_{\mathrm{SrVaried,SkFixed}}^{\mathrm{COMP}} - 1\right) \le \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkFixed,p}}}{P_{\mathrm{SrVaried,p}}}\right) F_{\mathrm{SrVaried,SkFixed,p}} \tag{14}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}} \cdot \mathrm{If}_{\mathrm{SrVaried,SkFixed}}^{\mathrm{COMP}} \le 0 \tag{15}$$

From a source with unknown pressure to a sink with unknown pressure:

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrVaried,SkVaried}}^{\mathrm{COMP}} - 1\right) \ge \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkVaried,p}}}{P_{\mathrm{SrVaried,p}}}\right) F_{\mathrm{SrVaried,SkVaried,p}} \tag{16}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}}\left(\mathrm{If}_{\mathrm{SrVaried,SkVaried}}^{\mathrm{COMP}} - 1\right) \le \left(c_{\mathrm{COMP}} + d_{\mathrm{COMP}} \cdot \frac{P_{\mathrm{SkVaried,p}}}{P_{\mathrm{SrVaried,p}}}\right) F_{\mathrm{SrVaried,SkVaried,p}} \tag{17}$$

$$\mathrm{Power}_{\mathrm{COMP,p}} - U_{\mathrm{COMP}} \cdot \mathrm{If}_{\mathrm{SrVaried,SkVaried}}^{\mathrm{COMP}} \le 0 \tag{18}$$

where Power represents the power demand of a compressor COMP, U is the upper bound for the power required by a compressor, $\mathrm{If}_{\mathrm{COMP}}$ is the binary variable indicating the existence of a compressor, P represents pressure at a source or a sink in hydrogen network, c and d represent the coefficients of the correlation for compression power (Liu, 2002), the subscripts SkFixed, SkVaried, SrFixed, and SrVaried represent sinks of known pressure, sinks of unknown pressures, e.g. new compressors, sources of known pressure, and sources of unknown pressure in a given hydrogen network respectively.

The capital cost of a new compressor is calculated using the linear correlation:

$$I_{\mathrm{COMP,p}}[\mathrm{k\$US}] = a_{\mathrm{COMP}} \cdot \mathrm{If}_{\mathrm{COMP}} + b_{\mathrm{COMP}} \cdot \mathrm{Power}_{\mathrm{COMP,p}}[\mathrm{kW}] \tag{19}$$

where $\mathrm{I}_{\mathrm{COMP}}$ is the capital cost of a compressor COMP, a and b are the cost coefficients for estimation of capital cost of a compressor (Liu, 2002).

3.3. Piping system

The sources and sinks of a hydrogen network are connected to each other through the piping system. The existence of a piping connection depends on the flow of gas from a source to a sink and is indicated by the binary variable $\mathrm{If}^{\mathrm{PIPE}}$. The following inequality constraints describe the piping connections between sources and sinks of a hydrogen network:

$$F_{v,w,p} - If^{PIPE}_{v,w} \cdot U_{flow} \le 0 \tag{20}$$

$$F_{v,w,p} - If^{PIPE}_{v,w} \cdot u_{flow} \ge 0 \tag{21}$$

The capital cost of the piping system for hydrogen network is estimated using a multi-period formulation of the model developed by Liu (2002):

$$I^{PIPE}_{SrFixed,w,p}[\$US] = \left(a^{PIPE}_{SrFixed,w} \cdot If^{PIPE}_{SrFixed,w} + b^{PIPE}_{SrFixed,w} \cdot \frac{0.02352 F_{SrFixed,w,p}[MMscfd]}{P_{SrFixed,p}[MPa]} \right) \cdot L^{PIPE}_{SrFixed,w}[m] \tag{22}$$

$$I^{PIPE}_{SrVaried,w,p}[\$US] = \left(a^{PIPE}_{SrVaried,w} \cdot If^{PIPE}_{SrVaried,w} + b^{PIPE}_{SrVaried,w} \cdot \frac{0.02352 F_{SrVaried,w,p}[MMscfd]}{P_{SrVaried,p}[MPa]} \right) \cdot L^{PIPE}_{SrVaried,w}[m] \tag{23}$$

where I^{PIPE} is the capital cost of a piping connection, and L^{PIPE} is the length of the piping connection between a source and a sink in hydrogen network. The length of piping connections may be specified for an existing hydrogen network or for a new design using the information about the plant layout (Liu, 2002).

3.4. Pressure constraints

Pressure constraints need to be incorporated in the design of hydrogen networks to account for the capital and operating costs of compressors required to meet the pressure differences between sources and sinks of a hydrogen network. Pressure constraints dictate the choice of installation of new compressors in the design of hydrogen networks. The need to install a new compressor depends on the conditions; that there is a flow of gas between a source and a sink, and that the pressure of sink is higher than the source, i.e. the pressure difference is less than zero (Liu, 2002):

$$If^{PIPE}_{v,w} = 1 \text{ And } If^{dp}_{v,w} = 1 \rightarrow If^{COMP}_{v,w} = 1 \tag{24}$$

where If^{dp} represents the binary variable to indicate the requirement to install a compressor because of pressure difference between a source and sink in hydrogen network. This condition is expressed as:

$$P_{w,p} - P_{v,p} - If^{dp}_{v,w} \cdot U_{dp} \le 0 \tag{25}$$

$$P_{w,p} - P_{v,p} + \left(1 - If^{dp}_{v,w}\right) \cdot U_{dp} \ge u_{dp} \tag{26}$$

The logical constraints for installation of a new compressor may be expressed as (Liu, 2002):

$$If^{COMP}_{v,w} - If^{dp}_{v,w} \le 0 \tag{27}$$

$$If^{COMP}_{v,w} - If^{PIPE}_{v,w} \le 0 \tag{28}$$

$$If^{dp}_{v,w} + If^{PIPE}_{v,w} - If^{COMP}_{v,w} \le 1 \tag{29}$$

3.5. Objective function

The objective function for the design and optimisation of hydrogen networks for multiple periods of operation is chosen to be the total annualised cost. The choice of total annualised cost as the objective function gives the advantage of expressing both the capital and operating costs of the hydrogen network on a common basis (Liu and Zhang, 2004). The total annualised cost (TAC), taking into account all the operating periods under consideration, is given by:

$$TAC = Cost_{H_2} - Credit_{Fuel} + Cost_{Power} + Af \cdot \left(\sum_{COMP} I_{COMP} + \sum_{PIPE} I_{PIPE} \right) \tag{30}$$

where Af is the annualisation factor, and may be given by Smith (2005):

$$Af = \frac{i \cdot (1+i)^n}{(1+i)^n - 1} \tag{31}$$

where i is the fractional interest rate per year, and n is the number of years.

For a multi-period design the capital and operating costs may vary for different operating periods as the operating conditions of refinery processes may have to be modified in order to satisfy the product demands. For example, in the operation of hydrotreating processes, operating conditions, such as reactor inlet temperature, is varied to compensate catalyst deactivation. Changing market demands may also influence the operation of individual refinery processes. The objective function, i.e. the total annualised cost therefore needs to be the sum of the capital and operating costs taking into account the maximum loads, and capacities on individual units from all the operating periods under consideration.

The cost of hydrogen utility, i.e. the cost of producing hydrogen for a hydrogen network, or the cost of hydrogen imported to a hydrogen network is taken as a weighted average of the cost for all the operating periods under consideration. The cost of hydrogen utility for each operating period is given by:

$$Cost_{H_2,p} = oy \cdot \sum_{w} F_{H_2U,w,p} \cdot PI_{H_2U} \tag{32}$$

where PI_{H_2U} represents the price per unit of hydrogen, and oy represents the annual number of hours of operation. The credit for gas sent to the fuel system is calculated using the heating value of the components of the gas stream, i.e. hydrogen and methane:

$$Credit_{Fuel,p} = oy \cdot \left(F_{H_2U,Fuel,p} \cdot HV_{H_2} + F_{CH_4,Fuel,p} \cdot HV_{CH_4} \right) \cdot PI_{HV} \tag{33}$$

where HV represents the heating value of a component. The operating cost of compressors consisting of the power consumed is given as:

$$\mathrm{Cost}_{\mathrm{Power}} \geq \mathrm{Cost}_{\mathrm{Power,p}} = \mathrm{oy} \cdot \sum_{\mathrm{COMP}} \mathrm{Power}_{\mathrm{COMP,p}} \cdot \mathrm{PI}_{\mathrm{Power}} \quad (34)$$

The capital cost of compressors and piping system is also handled in a similar fashion. The constraints presented here take into account the capital and operating costs corresponding to the maximum capacities and loads on individual units over all the operating periods under consideration.

3.6. Optimisation framework for multi-period design of hydrogen networks

The modelling equations described in Sections 3.2, and 3.3 include some equations with non-linear terms (model for compressors, Equations (5), (10), (11), (13), (14), (16), (17); model for piping system, Equation (23)). The presence of discrete variables along with non-linear terms makes design and optimisation of multi-period hydrogen networks a mixed integer non-linear programming problem. This section presents the optimisation framework adopted in this work for multi-period design of hydrogen networks.

3.6.1. Linear relaxation of non-linear terms

For mixed integer non-linear programming optimisation problems a global optimum may not be guaranteed if the problem is non-convex (Beigler et al., 1997). However, a suitable initialisation may lead the program to a good local optimum. All the non-linear terms in the hydrogen network model are either in bilinear form or can be converted to a bilinear form (Liu, 2002). Quesada and Grossmann (1995) proposed a methodology for optimisation of problems with networks consisting of splitters, mixers etc. involving multi-component streams, using linear relaxation of bilinear terms in order to improve the robustness of optimisation and help locate good local optimum. According to this approach, a bilinear term may be transformed into four linear inequalities using the upper and lower bounds for the two variables constituting the bilinear term (Quesada and Grossmann, 1995). Liu and Zhang (2004) illustrated successful application of this strategy for automated design of hydrogen networks. Therefore, this approach is employed in this work for design and optimisation of hydrogen networks for multiple periods of operation. All the non-linear terms of the multi-period hydrogen network model are linearised using this method and the original mixed integer non-linear programming problem is transformed into a mixed integer linear programming problem for which a global optimum may be obtained. The upper and lower bounds for variables may be specified easily using physical insight of a given system.

3.6.2. Design and optimisation algorithm

The mixed integer linear programming formulation is solved, and the solution is used to initialise the original mixed integer non-linear programming formulation. In this way convergence to a feasible solution is facilitated and the probability of obtaining a good local optimum solution is improved.

This design and optimisation strategy for multi-period hydrogen management is shown in Fig. 3. This approach assumes that there is a compressor between every source and sink because of the unknown pressures initially. This assumption leads to a high number of compressors with some of them providing only small compression duties (Liu, 2002). In order to avoid a high number of compressors, the design obtained by the optimisation procedure may be evolved by merging compressors, i.e. use of a single compressor with a capacity equivalent to the combined power demand of the smaller compressor units, to achieve simpler, and more practicable network designs (Liu, 2002).

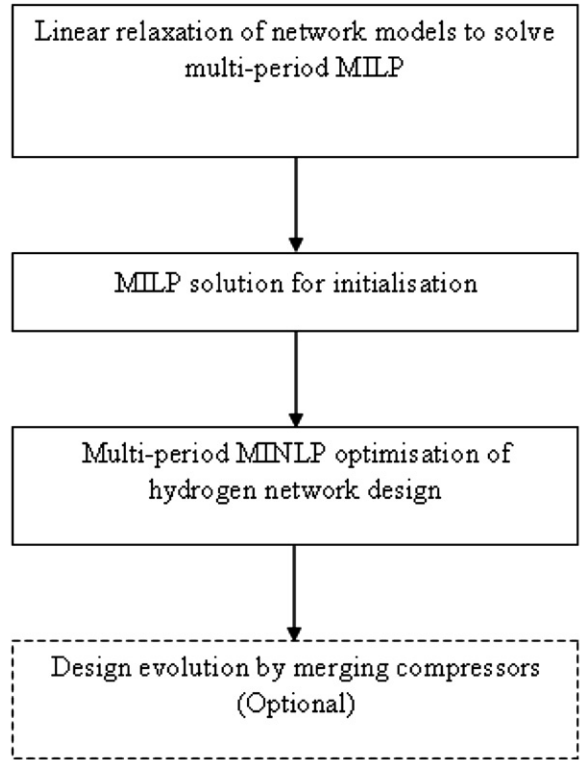

Fig. 3. Design and optimisation procedure for multi-period hydrogen network management.

The design and optimisation strategy of the approach developed in this work is different from the single-period automated design approach as the proposed formulation consists of continuous variables, e.g. flow rates and purities of streams, with an additional dimension to account for multiple periods of operation. Both the mixed integer linear programming formulation, to obtain a suitable initialisation, and mixed integer non-linear programming formulation, to optimise the design of hydrogen networks, are multi-period models to provide the flexibility to satisfy the demands of hydrogen consuming processes in all the operating periods under consideration. The overall material and component balances (Section 3.1) as well other constraints, e.g. Section 3.2 for compressors, Section 3.3 for piping system, and Section 3.4 for pressure constraints need to be satisfied for all the operating periods under consideration.

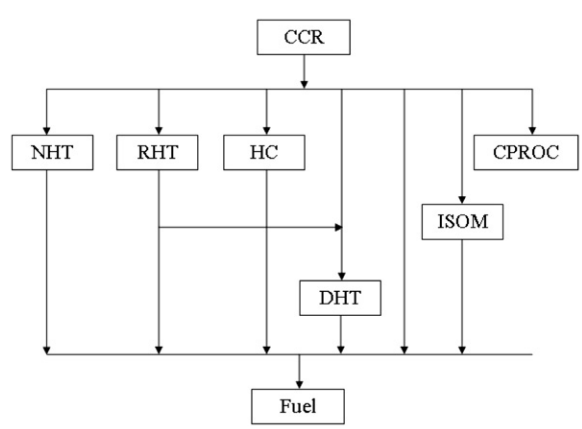

Fig. 4. A simplified refinery configuration for design of hydrogen network (Sun, 2004).

Table 1
Net hydrogen sink data of the hydrogen network under consideration.

Unit	Flow rate (t/h)	Purity (mass fraction)	Pure H_2 flow (t/h)
NHT	3.108	0.262	0.815
RHT	5.081	0.262	1.333
HC	3.251	0.262	0.853
DHT	25.798	0.202	5.198
ISOM	5.913	0.259	1.535
CPROC	2.535	0.262	0.665
CCR fuel	0.446	0.262	0.117

Finally it is important to note that the number of operating periods under consideration, duration of each operating period and its representation in terms of the source and sink data influences the optimisation of the design of a multi-period hydrogen network. The choice of number of operating periods reflects a trade-off between the performance of hydrogen network and the complexity of design calculations. The data used to represent each operating period in design of multi-period hydrogen networks needs to be a reflection of the changing operating conditions over the whole catalyst life of hydrogen consuming processes such as hydrotreaters.

4. Case study

In this section a case study is presented to illustrate the application and benefits of the methodology developed for multi-period design of hydrogen networks. The results of the case study will elucidate the importance and impact of changing operating conditions of refinery processes, such as increase in reactor operating temperature of hydrotreating processes, on the design of hydrogen networks. The objective of this case study is to design a hydrogen network with a lower total annualised cost, compared to conventional design approaches, i.e. single-period hydrogen network design and optimisation. The multi-period hydrogen network designed using the new methodology must satisfy the demands of hydrogen consuming processes, for all the periods of operation under consideration, with minimum total annualised cost, while the single-period design obtained using the approach of Liu (2002) leads to a design with sub-optimal overall cost.

The refinery configuration for which a hydrogen network needs to be designed is taken from Sun (2004). A simplified representation of the refinery configuration under consideration is shown in Fig. 4.

It should be noted here that some refinery processes with minor hydrogen consumptions have been represented as a combined process sink CPROC. The process data, of refinery processes for a single operating period, are classified as source and sink data and are shown in Table 1 and Table 2 (Sun, 2004).

Hydrogen pinch analysis is used to estimate the minimum amount of hydrogen required from the hydrogen producer, i.e. the catalytic reformer in this case, to satisfy the demands of hydrogen consuming processes. Fig. 5 shows the balanced hydrogen surplus diagram for the given hydrogen network.

Hydrogen pinch analysis indicates that a reduction of 3.42 t/h of hydrogen gas stream flow rate is possible in the hydrogen producer for the system under consideration. It may be observed from the

Table 2
Net hydrogen source data of the hydrogen network under consideration.

Unit	Flow rate (t/h)	Purity (mass fraction)	Pure H_2 flow (t/h)
CCR	16.73	0.262	4.39
NHT	2.24	0.286	0.64
RHT	4.95	0.244	1.21
HC	3.16	0.244	0.77
DHT	18.79	0.180	3.38
ISOM	4.58	0.259	1.18

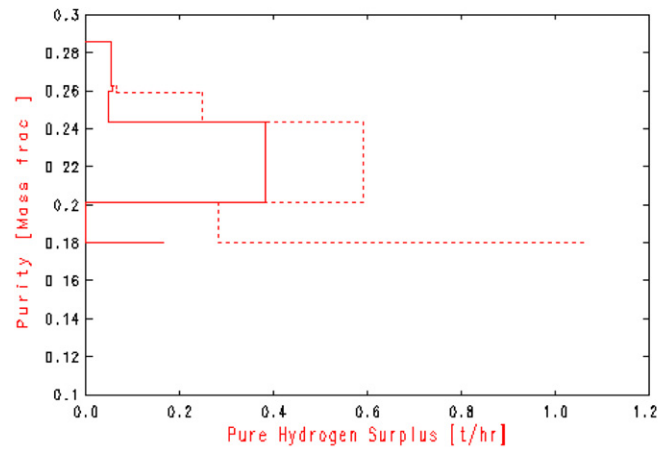

Fig. 5. Hydrogen surplus diagram of the hydrogen network under consideration.

hydrogen surplus diagram, shown in Fig. 5, that the hydrogen pinch occurs at a hydrogen mass fraction of 0.201. This purity corresponds to the requirements of the diesel hydrotreating unit, as can be seen in Table 1. The purity at which hydrogen pinch occurs, indicates that the diesel hydrotreating unit may cause a bottleneck for any expansion that may require additional hydrogen consumption.

The excess hydrogen available in the given system may be utilised in one of the refinery processes to increase the profit of the refinery. The hydrogen demand of the given system is met by the catalytic reformer unit, with a specified production, and not from a hydrogen production plant. Therefore, utilising the excess amount of hydrogen to improve the performance of refinery processes would be a logical choice (Sun, 2004).

Sun (2004) illustrated two different ways to improve the economic performance of the diesel hydrotreating unit through utilising the surplus hydrogen available in the hydrogen network under consideration:

- By increasing the throughput of the diesel hydrotreater.
- By blending light cycle oil into the diesel hydrotreater feed.

The option of blending light cycle oil with the diesel hydrotreater feed, i.e. straight-run gas oil in for the given case study, enables the upgrading of cheaper feedstock. However, the higher sulphur and aromatic content of light cycle oil require higher hydrogen consumption and shorten the catalyst life for the process (Sun, 2004). Sun (2004) illustrated that the available surplus hydrogen may be utilised for blending 5 volume % of light cycle oil with the diesel hydrotreater feed. Blending 5 volume % of light cycle oil with the diesel hydrotreater feed increases the net hydrogen

Table 3
Economic data for case study (Liu, 2002).

Operating costs	
Hydrogen cost	\$2000/MMscfd
Electricity cost	\$0.03/kWh
Capital costs	
Compressors	$\mathrm{Cost[k\$US]} = 764.86 \cdot \mathrm{If}^{\mathrm{COMP}} + 1.7596 \cdot \mathrm{Power_{COMP}[kW]}$
Piping system	$\mathrm{Cost}^{\mathrm{PIPE}}_{\mathrm{SrFixed},w}[\$\mathrm{US}] = \left(420.74 \cdot \mathrm{If}^{\mathrm{PIPE}}_{\mathrm{SrFixed},w} + 1484.76 \cdot \frac{0.02352 F_{\mathrm{SrFixed},w}[\mathrm{MMscfd}]}{P_{\mathrm{SrFixed}}[\mathrm{MPa}]}\right) \cdot L^{\mathrm{PIPE}}_{\mathrm{SrFixed},w}[\mathrm{m}]$
	$\mathrm{Cost}^{\mathrm{PIPE}}_{\mathrm{SrVaried},w}[\$\mathrm{US}] = \left(420.74 \cdot \mathrm{If}^{\mathrm{PIPE}}_{\mathrm{SrVaried},w} + 1484.76 \cdot \frac{0.02352 F_{\mathrm{SrVaried},w}[\mathrm{MMscfd}]}{P_{\mathrm{SrVaried}}[\mathrm{MPa}]}\right) \cdot L^{\mathrm{PIPE}}_{\mathrm{SrVaried},w}[\mathrm{m}]$

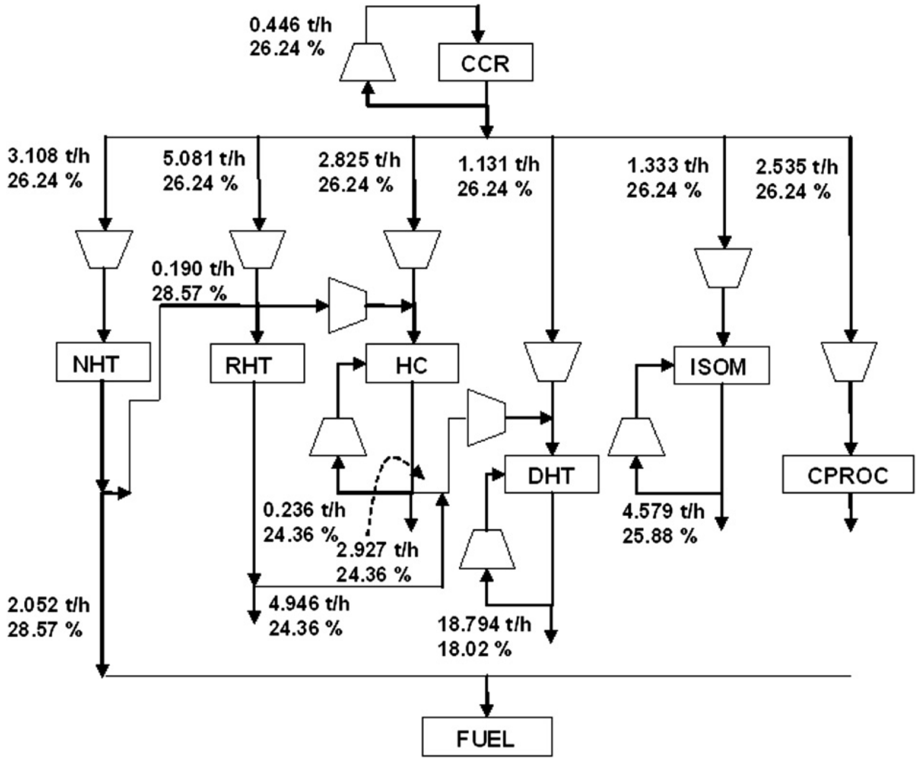

Fig. 6. Hydrogen network design using single-period automated design approach (gas flow rates and hydrogen purity (mass basis) are shown for all streams).

sink of the diesel hydrotreating process to 27.8 t/h, at a purity of 0.2015 mass fraction, and consumes the surplus hydrogen that was originally available in the given hydrogen network.

In this case study first a hydrogen network is designed for the new scenario with increased hydrogen consumption of diesel hydrotreating process using the automated design approach of Liu (2002). The automated design approach of Liu (2002) can only handle single-period operating conditions. In this case study the changing operating conditions of the diesel hydrotreating process that result in changing hydrogen consumption of the diesel hydrotreater are taken into account. A multi-period hydrogen network is designed for the same refinery configuration using the new design methodology. The resulting hydrogen network is compared with the network designed using the automated design approach of Liu (2002) to elucidate the advantages of the proposed approach for design of hydrogen networks for multiple periods of operation. The comparison also demonstrates the ability of the proposed approach to account for changing operating conditions of refinery processes.

The economic data for this case study, i.e. the capital and operating cost data for compressors, and piping system are taken from Liu (2002) and are shown in Table 3. The capital costs are annualised over 5 years, with 5% interest rate.

A simplified block diagram of the hydrogen network designed using the automated design approach of Liu (2002) is shown in Fig. 6.

The cost breakdown for the hydrogen network designed using the design approach of Liu (2002) is shown in Table 4.

The total annualised cost of the hydrogen network designed using the approach of Liu (2002) is 147.3 MM USD/yr. The capital cost consists of the investment for installation of compressors and the piping system. The operating cost of the hydrogen network consists of the power tariff, and cost of hydrogen from the hydrogen producer.

The automated design approach of Liu (2002), as well as hydrogen pinch analysis and the design approach of Alves (1999), assume constant operating conditions of refinery processes. These approaches therefore do not provide a framework for analysis of the influence of changing operating conditions on the performance of a hydrogen network. For example, the operating conditions e.g. reactor temperature of diesel hydrotreating process, which is the largest hydrogen consumer in the given hydrogen network, will change periodically in order to compensate for the catalyst deactivation due to coke formation. The changing operating conditions of the diesel hydrotreating process result in a change in the hydrogen consumption and therefore, in the net source flow rate from the diesel hydrotreating unit.

Table 4
Cost breakdown for single-period design obtained using the design approach of Liu (2002).

Operating cost (MM USD/yr)	133.508
Hydrogen cost	130.309
Electricity cost	3.198
Capital cost (MM USD)	49.901
Compressors	43.535
Piping system	6.365
Total annualised cost (MM USD/yr)	147.305

Table 5
Process data for diesel hydrotreating unit using molecular diesel hydrotreating reaction model.

Operating period	Catalyst activity (%)	Reactor temperature (K)	Hydrogen consumption (t/h)
1	84	618.7	0.681
2	57	625.4	0.601
3	32	634.9	0.494

Table 6
Net source data of diesel hydrotreating unit for the three operating periods under consideration.

Operating period	Flow rate (t/h)	Purity (Mass fraction)
1	18.795	0.1802
2	19.239	0.1802
3	19.833	0.1802

The impact of catalyst deactivation on the process conditions and net source flow rate of the diesel hydrotreating unit is predicted using a molecular pathways level model of the diesel hydrotreater developed by Sun (2004). Table 5 shows the process data for the diesel hydrotreating unit obtained using the molecular pathways level model of diesel hydrotreater.

The operating conditions of the diesel hydrotreating unit are varied in order to compensate for the catalyst deactivation while satisfying the product specifications (Ahmad, 2009). The operating conditions of diesel hydrotreating unit have been characterised in terms of three operating periods, using the expected catalyst life and rate of loss of catalyst activity (Ahmad et al., 2009). The percentage catalyst activity, reactor temperature and hydrogen consumption of the reactor as shown in Table 5 are the mean values for second and third operating period. The hydrogen consumption of the first operating period shown in Table 5 represents the maximum hydrogen consumption of the diesel hydrotreater with 100% active catalyst.

In this way the limiting condition of maximum hydrogen consumption, along with the changing hydrogen consumption over the whole catalyst life, of the diesel hydrotreating process is taken into account. These values are determined by simulation of the diesel hydrotreater operation over the whole catalyst life using the molecular pathways level model of diesel hydrotreater. The changing hydrogen consumption of the diesel hydrotreating unit may be attributed to the decrease in conversion of aromatic hydrogenation reactions with increase in reaction temperature, resulting in different net sources flows, i.e. the recycle and purge flows available from the diesel hydrotreating unit, for each operating period. Table 6 shows the net source data of the diesel hydrotreating unit for the three operating periods under consideration.

It can be seen in Table 6 that the net source flow rate available from the diesel hydrotreating process increases with time. This change results in additional available hydrogen of up to 1.038 t/h. The additional hydrogen available from the diesel hydrotreating unit may be utilised to improve allocation of hydrogen in the second and third operating periods. The single-period approaches e.g. (Liu, 2002; Alves, 1999) for design of hydrogen networks do not include such considerations. The hydrogen networks designed using single-period approaches would typically send excess hydrogen to the fuel system or to be flared eventually.

The proposed methodology takes into consideration the information available from all the operating periods of refinery processes, e.g. the diesel hydrotreating unit in this case. Fig. 7 shows a simplified block diagram of the hydrogen network designed using the proposed methodology. The flow rates of gas streams shown in Fig. 7 represent the maximum flow rates over the three periods of operation under consideration.

The design obtained using the proposed methodology is similar to the single-period design, shown in Fig. 6, obtained using the design approach of Liu (2002) in terms of structural features. However, additional compression and piping connections of the multi-period design provide flexibility for better utilisation of hydrogen in all three operating periods. A comparison of the cost breakdown for the multi-period design of hydrogen network with the single-period design obtained using the automated design approach of Liu (2002) is shown in Table 7.

A characteristic feature of multi-period designs may be observed from Table 7 that multi-period designs may require higher capital costs compared to single-period designs. The reason

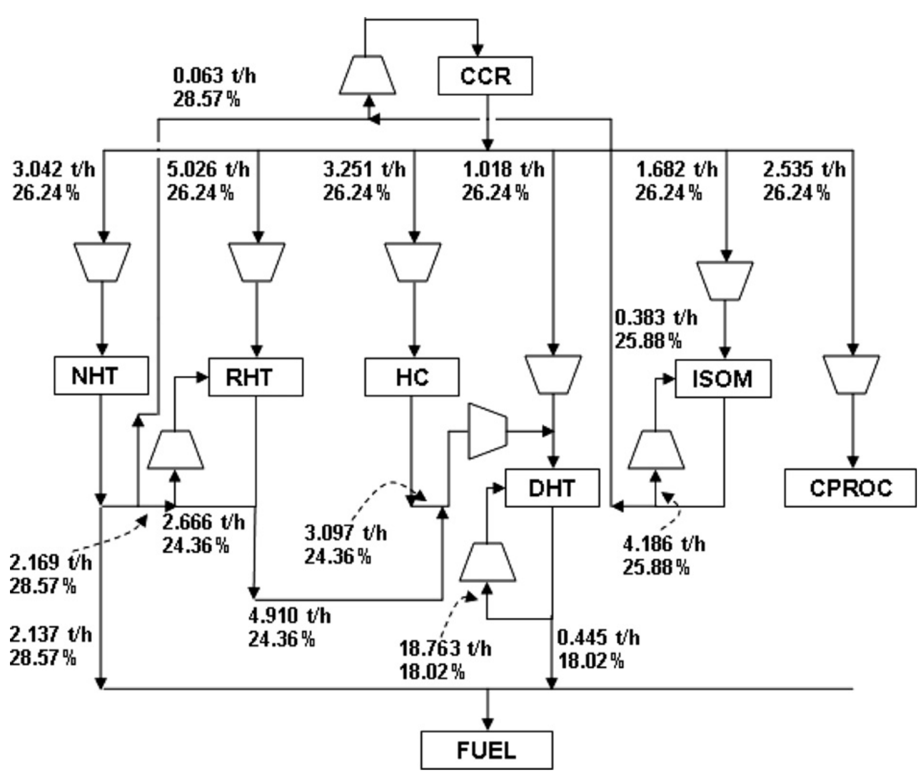

Fig. 7. Hydrogen network designed using the proposed methodology for multi-period hydrogen management.

Table 7
A comparison of cost breakdown between the hydrogen network designs.

	Single-period hydrogen network design	Multi-period hydrogen network design
Operating cost (MM USD/yr)	133.508	123.304
Hydrogen cost	130.309	118.551
Electricity	3.198	4.753
Capital cost (MM USD)	49.901	56.908
Compressors	43.535	47.897
Piping	6.365	9.010
TAC (MM USD/yr)	147.305	136.450

for the higher capital costs of multi-period designs is that different operating conditions may require different capacities of equipment, e.g. of compressors. In multi-period design the capital costs are calculated using the maximum capacity of all the equipment for a given system. This feature is evident in Table 7 which shows that the capital cost of compressors and piping system is higher for the multi-period design compared to the single-period design. However, the hydrogen network designed using the proposed methodology provides improved hydrogen allocation resulting in lower operating cost i.e. the hydrogen cost.

The hydrogen network designed using the multi-period formulation has a total annualised cost of 136.5 MM USD/yr and results in a saving of 10.9 MM USD/yr corresponding to a reduction of 7.4% in the total annualised cost compared to the hydrogen network designed using the automated design approach of Liu (2002).

5. Conclusions

Hydrogen management is essential for efficient and economic operation of a refinery. The major drawbacks of the existing techniques for hydrogen management are that hydrogen streams are treated as binary mixtures of hydrogen and methane, and the operating conditions of refinery processes are assumed to remain constant. A multi-period approach for the design of hydrogen networks is developed in this work to address this simplifying assumption of constant operating conditions of refinery processes. The proposed methodology accounts for the changing operating conditions of refinery processes. The hydrogen networks designed using the proposed methodology for multi-period hydrogen management can operate under multiple periods of operation with lower total annualised cost compared to hydrogen networks designed using single-period design approaches.

A limitation of the proposed methodology is that hydrogen streams are treated as binary mixtures of hydrogen and methane. A multi-component approach may be needed for further development of multi-period hydrogen management.

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