Design of Oil Refineries Hydrogen Network Using Process Integration Principles

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ABSTRACT: *This paper describes the application of process integration principles to the design of oil refineries hydrogen network. In this regard, a design hierarchy as well as heuristics and required guidelines are proposed. The recommended rules compensate lack of procedure to the design and make the design process easier. The guiding principles of the design are based upon pinch technology and extending the heat integration concepts to mass integration. This research makes a designer able to maximise the amount of hydrogen recovered across the site during the design. Besides, it provides an opportunity for refineries to make most efficient use of hydrogen. The study is illustrated with an industrial case study. The work stages such as targeting, simulation etc. are performed in the REFOPT software environment. It is finally shown that this approach can design a network, which saves the total cost by $ 6.3459 million per year.*

KEY WORDS: *Design, Hydrogen network, Oil refineries, Process integration.*

INTRODUCTION

In the last decade, the worldwide refining industry has been impacted by several trends that have increased hydrogen demand significantly. Environmental legislation is increasing demand for hydrotreating, while reducing hydrogen production from catalytic reforming. The declining value of heavy fuel oil and shift to heavier crude oils also create increased demand for hydrogen. These factors have led to higher hydrogen consumption for upgrading crude oil into light transportation fuels and removing sulphur and nitrogen compounds [1, 2].

Hydrogen is an important and expensive utility in oil refining and petrochemicals processing. It is required for

many operations such as hydrotreating (where it is used to remove impurities such as sulphur from streams and to hydrogenate aromatics and ole-fins) and hydrocracking (where it breaks down large hydrocarbons into smaller, higher-value molecules).

The main sinks are the refinery processes, which consume hydrogen such as hydrotreaters and hydro-crackers. Fig. 1 shows a typical hydrogen consumer.

A liquid feed stream is mixed with a gas rich in hydrogen, heated and fed to the hydrotreating or hydrocracking reactor. Some of the hydrogen is consumed in the reactor and light hydrocarbons and other gases are formed. The reactor effluent is cooled and sent to a gas-liquid separator. The gas from the separator may be treated in an amine unit to remove hydrogen sulfide and part of it is re-compressed and recycled to the reactor inlet. The remainder of the gas stream is purged in order to prevent build-up of contaminants in the recycle loop. This purge stream may be re-used elsewhere, but is often sent to the fuel system, where it is burned for its heating value, or flared. Sometimes, the liquid product may be sent to a low-pressure separator where an off-gas stream is taken and typically sent to the fuel gas system. Fuel systems and flares are therefore also potential sinks.

There are several possible sources of hydrogen. Typically, most of the make-up hydrogen in refineries is supplied from catalytic reforming. Catalytic reforming produces aromatic compounds from the cyclization and dehydrogenation of hydrocarbon molecules and is used to increase the octane number of naphtha. At the same time, large amounts of hydrogen are produced as a byproduct. If the hydrogen from catalytic reforming is insufficient, additional hydrogen may be supplied by building a hydrogen plant that produces the gas by either steam reforming or partial oxidation of hydrocarbons. Alternatively, hydrogen may be imported via a pipeline. Hydrogen that is obtained from a hydrogen plant or import is termed a utility. Finally, the off-gases from hydrogen consumers are also sources because they can be re-used in other consumers. Sources of hydrogen can be intercepted to make them more acceptable by using compressors (which upgrade their pressure) and or purifiers (which upgrade their hydrogen purity).

The purge streams from the hydrogen consumers may contain a reasonably high percentage of hydrogen and it is often beneficial to recover and re-use these streams
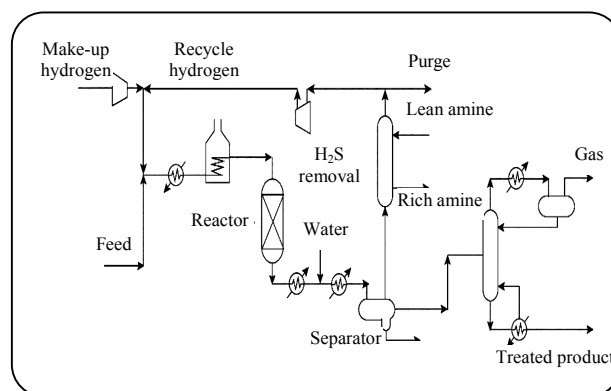

Fig. 1: Schematic diagram of a typical hydrogen consumer.

rather than send them to the fuel gas system or flare. Any hydrogen that can be recovered will reduce the size and capital cost of a new hydrogen plant. Alternatively, it will reduce the operating costs of existing hydrogen plants (feedstock and fuel) or imports.

The published literature focuses mainly on improve-ments to individual consumer units. For example, discuss modifications within hydro-crackers in order to reduce the hydrogen usage and improve the unit flexibility. *Baade et al.* discuss ways of expanding steam reformers while *Kramer et al.* address the problem of designing flexible hydrogen plants. While these works are all important, it is arguably more important to consider all the hydrogen consumers as part of an integrated refinery in order to exploit the interactions and synergies, which may exist [3].

Another topic, which has been extensively addressed in the literature, is hydrogen purification. The most common unit operations used for purifying hydrogen are pressure-swing adsorption (PSA) and membranes. Other methods do exist, for example, cryogenic separation or liquid absorption, but these are not as widely used. Most of the published literature appears once again to focus on the individual process rather than the overall refinery context. For example, *Ratan* gives an excellent description on each of the commonly used purification processes and discusses the operational advantages and disadvantages of each of them. *Spillman* discussed the use of membranes in refinery applications while *Tomlinson* and *Finn* compared various purification processes. *Peramanu et al.* carried out an economic evaluation of different purification technologies for hydrotreater and hydrocracker purge gases [3].

These authors examined four specific industrial case studies in order to determine the best choice of purifier for purge gases of different purities and pressures.

However, each case study only involved a single consumer (hydrotreater or hydrocracker) and was therefore not a refinery-wide or site-wide one. In order to gain the maximum benefit from purification, the purifier should be correctly integrated in an overall context. In other words, it is necessary to determine the best stream (s) to intercept as well as the destination (s) of the purified hydrogen product. In many cases, the best option is to purify the purge from one consumer so that it may be re-used as part of the make-up for a totally different consumer. *Towler et al.* proposed analysing the hydrogen distribution system by comparing the cost of recovering hydrogen and the value added by hydrogen in refinery processes. This work gave insight into the economics of the refinery, but did not account for the physical constraints that influence the distribution network [3].

Graphical targeting approach, which is one of the most promising and industrially exploited approaches to date, was developed by *Alves* and is based upon pinch technology. This work exploits an analogy with heat exchanger network synthesis. The method identifies sources, sinks of hydrogen, which are analogous to hot and cold streams in heat exchanger networks. The major limitation of Alves's approach is that the targets consider only the flowrate and purity requirements and ignore pressure. The targeting method assumes that any stream containing hydrogen can be sent to any consumer, regardless of the stream pressure. In reality, this will only be true if the stream is at a sufficiently high pressure. Thus, the targets generated may be too optimistic and unachievable in a real design [3].

Hallale et al. developed an improved method for hydrogen network retrofit. The methodology is based upon mathematical optimisation of a superstructure and maximises the amount of hydrogen recovered across a site. The techniques account fully for pressure constraints as well as the existing equipment and are suited for revamping real, industrial systems. The optimum placement of new equipment such as compressors and purification units is also considered. The method is not limited to minimising hydrogen, but can account for all the dominant trade-offs in order to perform cost optimization [3].

Although the Hallale's work presents a proper methodology up to now, but it does not introduce a procedure for hydrogen networks design or retrofit. By the way, lack of guidelines and heuristics, which make the design process easier, is felt. In this regard, a designer may be faced with the following questions,
- Where the design must be started?
- What are the design criteria and tools?
- How is the design procedure and trend?
- How the network must be analyzed and which parameters are more important?
- etc.

This paper develops the Hallale's work by proposing a new hierarchy in addition to guidelines and heuristics to the design of hydrogen networks. The study is elaborated with a case study, in which a hydrogen network is designed through the hydrogen integration principles.

GRAPHICAL TARGETING APPROACH
Hydrogen composite curves and surplus diagram

The material balance on total gas for each of the hydrogen sinks is conveniently represented in a two-dimensional plot with flowrate of total gas on the horizontal axis and purity on the vertical axis, as shown in Fig. 2a. This purity profile contains the hydrogen sinks plotted in order of decreasing purity as a composite sink profile. The same procedure is used to plot the hydrogen sources in order of decreasing purity as a composite source profile. The position in the vertical axis and the length of the first step in the sink curve corresponds to the purity and flowrate of the hydrogen sink with highest purity. The next step corresponds to the hydrogen sink with the second highest purity, etc. The construction of the curve continues until the lowest-purity hydrogen sink is represented. The source curve also starts at zero flowrate. Its construction follows the same procedure used for the sink curve, representing the highest-purity hydrogen source first and the lowest-purity source last. The purity profile diagram does not require any information on connections within the hydrogen network and is based solely upon the source and sink data. In the purity profile diagram, the projection of both curves onto the horizontal axis represents the material balance on total gas.

The first necessary condition for the feasibility of the hydrogen distribution system is that the amount of gas available from the sources must equal or exceed the
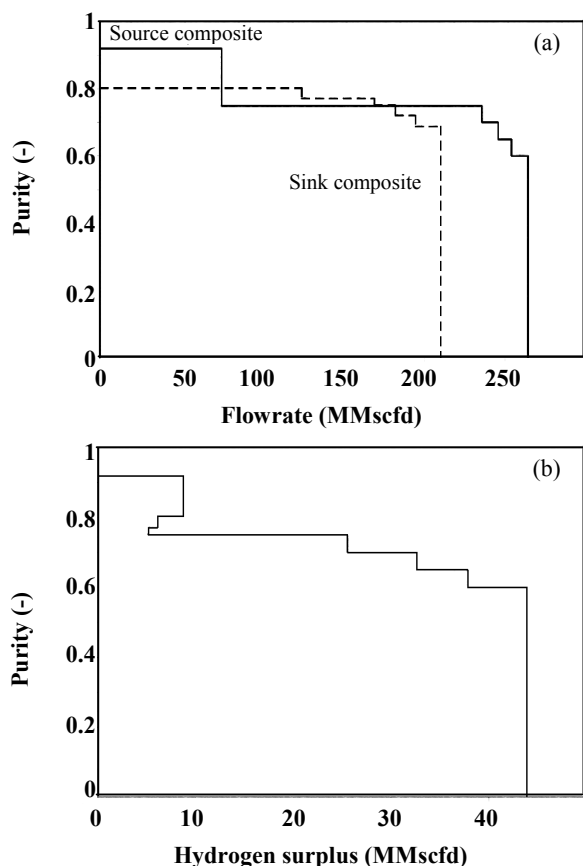
Fig. 2: a) The hydrogen composite curves, b) the hydrogen surplus diagram.

amount of gas required by the sinks. The amount of gas supplied in excess of the demand is flared or sent to the fuel gas system. If the source curve is shorter than the sink curve, then the material balance on total gas is violated for at least one of the sinks, rendering the system unfeasible.

The solution is to add more gas to the system or to operate one or more hydrogen-consuming processes with reduced liquid throughput (which reduces the amount of gas required by the corresponding hydrogen sink). The supply available from the sources exceeding the demand from the sinks is not a sufficient condition for feasibility of the hydrogen distribution problem. The supply also must comply with the constraints on the purity of hydrogen imposed by each sink. A new variable, hydrogen surplus, is introduced to describe the availability of hydrogen at different purity levels. The hydrogen surplus is developed from the information contained in the hydrogen purity profiles, although it can be calculated independently of

the profiles. It can be represented graphically in a hydrogen surplus diagram, as shown in Fig. 2b.

The hydrogen surplus can be calculated from the purity profile. The area underneath each segment of the sink curve is equal to the flowrate of pure hydrogen required by the hydrogen sink represented by that segment. The area underneath the whole sink curve is the flowrate of pure hydrogen that the system must provide to all of the sinks. The area underneath the source curve is the total amount of pure hydrogen available from the sources. When both curves are put together to build the purity profile, there are regions where the source curve lies above the sink curve and regions where it lies below the sink curve, as illustrated in Fig. 2a. If the source curve is above the sink curve for a given range of hydrogen purity, then in this range of purity, the sources provide more hydrogen than is required by the sinks. Here, the purity profile has an excess of hydrogen that amounts to the area of the space between the curves. Because this amount of hydrogen is in excess to that needed in this purity range, it constitutes a surplus and can be used to compensate for a deficit in hydrogen supply at a lower purity. If the source curve is below the sink curve, then in this region, the sources do not provide enough hydrogen to the sinks; here, the purity profile has a deficit of hydrogen.

The amount of deficit of hydrogen is equal to the area between the curves. This deficit can be compensated by use of surplus hydrogen of a higher purity, but not by use of hydrogen of lower purity. The physical meaning of a deficit region is that the sink process requires hydrogen of greater purity than that available from the corresponding source. If surplus hydrogen is available at a higher purity, then it can be mixed with the lower-purity source to raise its purity until the sink purity requirement is satisfied. Overall, the purity profile is divided into several regions with alternating excess and deficit of hydrogen.

If the hydrogen surplus is negative at any value of flowrate between zero and total flowrate available from the sources, then the system is not receiving sufficient hydrogen at adequate purity. At least one of the constraints on hydrogen flowrate imposed by the sinks cannot be satisfied. More hydrogen or higher-purity hydrogen is required to make the system feasible. The second necessary condition for feasibility of the hydrogen distribution system can then be stated as hydrogen surplus

function can be represented as a hydrogen surplus diagram, as shown in Fig. 2b.

The curve in the diagram is generated by plotting whichever is the lower (or higher) of the source and the sink curve purities, versus hydrogen surplus for each value of flowrate of total gas between zero and total flowrate available from the sources. It is necessary to use the lower of the source and the sink curve purities, as the surplus or deficit of hydrogen changes depending on whether the source or the sink curve is lowermost. If the sink curve is lowermost, then there is an excess of hydrogen, and hydrogen surplus will increase. In this case, the surplus is plotted at a purity of the sink curve purity. If the source curve is lowermost, then there is a deficit of hydrogen. In this case, hydrogen surplus will decrease and is plotted at a purity of the source curve purity. If the whole hydrogen surplus curve lies at or above zero hydrogen surplus, then the second necessary condition for feasibility is satisfied. If both the first and the second necessary conditions are satisfied, then the network design problem has at least one feasible solution [3, 4].

Hydrogen pinch

The hydrogen supply target is defined when the system is constrained on hydrogen. This occurs if there is at least one place in the hydrogen surplus diagram where the hydrogen surplus is zero and any reduction in the supply creates a negative hydrogen surplus, making the distribution problem unfeasible. When the constraint is just satisfied, then the hydrogen surplus diagram appears to be pinched where the surplus is zero, as shown in Fig. 3. The hydrogen pinch thus sets a target for the minimum hydrogen consumption.

The pinch occurs at the end of a range in the purity profile where the hydrogen is in deficit. It corresponds to a discontinuity in the sink line where a hydrogen sink that is above the source line ends and another sink, below the source line, starts. Because of the discontinuity in the sink profile, a vertical segment at zero hydrogen surplus follows the pinch. This segment is located between the values of purity of the hydrogen source at the pinch and sink after the pinch. It represents the fact that the pinch divides the overall distribution system into a subsystem with net zero hydrogen surplus (above the pinch) and a subsystem with net hydrogen surplus (below the pinch).
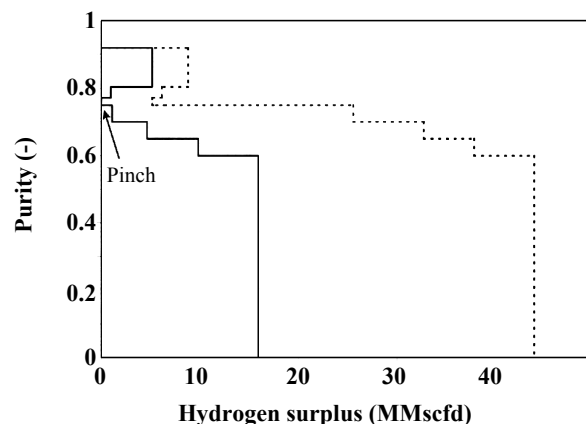

Fig. 3: The minimum utility target is found when the surplus diagram exhibits a pinch.

The pinch purity is the purity associated with the deficit that causes the pinch, i.e., the purity of the hydrogen source at the pinch.

The subsystem above the pinch includes the pinch recycle, which is the portion of the hydrogen source at the pinch purity that belongs to the region above the pinch. The pinch recycle flowrate tells us how much gas from the source at the pinch must be reused by the sinks above the pinch to meet the hydrogen supply target. If an additional amount of gas from below the pinch is reused above the pinch, then an identical amount of gas originally above the pinch, at higher purity, must be sent to the sinks below the pinch to maintain material balance. This reduces the hydrogen surplus above the pinch, and a penalty in the supply must be paid to keep the system feasible. Thus, a simple rule for designing a hydrogen system at the minimum supply is that gas should never be exchanged across the pinch. This is the main design guideline provided by the hydrogen targeting method. It corresponds closely to the pinch design rule used in the analysis of heat exchanger networks [3, 4]. In other words, the recovery of hydrogen from off gases for reuse in refinery processes is a problem analogous to the recovery of process heat [5].

The procedure for calculating the supply target requires that the flowrate of gas supplied to the system be varied until a hydrogen pinch is found. Few sources of hydrogen in the refinery have variable flowrates. The sources from hydrogen-consuming processes or from processes that generate hydrogen as a secondary product, such as catalytic reformers and other dehydrogenation
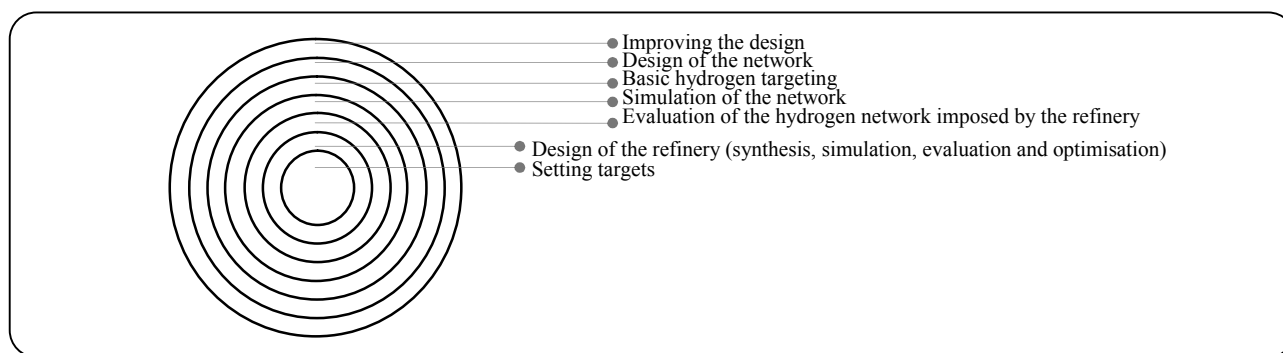
Fig. 4: The hierarchy proposed to the design.

plants, have flowrates that will be determined by the normal operation of the processes. These flowrates will be determined by optimization of the refinery and can be assumed to be fixed for the hydrogen network design problem. Purification units usually have low operating costs, and once installed, they should be operated at maximum capacity. Their outlet gas streams are therefore also fixed sources of hydrogen. The hydrogen sources that are flexible with respect to flowrate are thus imports from external suppliers and processes that generate hydrogen as the main product (steam reformers or partial oxidation units). These sources act as hydrogen utilities. The targeting procedure is the same for a feasible system initially operating at low efficiency or for an initially unfeasible set of sinks and sources: start with maximum flowrate for all of the utilities, and minimize the most costly one. If this utility is eliminated before the system is at the minimum supply, then the minimization proceeds with the next utility in decreasing order of cost [4].

HYDROGEN NETWORK DESIGN
The design hierarchy

It is helpful when developing a methodology have a clearer picture of the nature of the problem [6]. Design of a hydrogen network is feasibly dependent on the refinery flow diagrams. Hence, a hydrogen network should be designed after the refinery design. As a result, some parameters such as inlet and outlet flowrates, purities and pressures of process units are imposed to the design. These parameters are usually fixed during the design. Design of chemical processes has four major steps, which are synthesis, simulation, evaluation and optimization, respectively. In this paper in order to design hydrogen networks, a design hierarchy has been proposed, as shown in Fig. 4.

The diagram shows that before the refinery design, the targets must be initially identified. The refinery is subsequently designed. The refinery design introduces a number of process constraints to the hydrogen network design. The hydrogen network design is started whenever the refinery design is completed. In this regard, the network imposed by the refinery is initially evaluated. In order to understand the network mass balance, both hydrogen composite curves and surplus diagrams must be plotted. After that, the hydrogen network is simulated to specify the design objectives and the network costs. To identify the hydrogen recovery potential as well as pinch point, the hydrogen in the network must be targeted. This stage specifies that whether both the first and the second necessary conditions for feasibility of hydrogen distribution system can be fulfilled or not. In other words, is the system balanced? At the end, the network is designed through the data acquired in addition to the design tools.

The design procedure for maximum hydrogen recovery

So far, the basic concepts of hydrogen pinch analysis has been described in sufficient detail to demonstrate that, for complex processes, this approach should be an important part of a hydrogen-saving study or process design procedure.

The pinch point divides the problem in two separate systems: one above, and one below the pinch. The system above the pinch requires a hydrogen input and is therefore a net hydrogen sink. Below the pinch, the system rejects hydrogen and so is a net hydrogen source. This paper proposes the guiding principles of the design in order to achieve the minimum utility target for a network, as:

- Hydrogen must not be transferred across the pinch (with the exception of using a purifier).

- No hydrogen utility must be used below the pinch.

- The site fuel system must be located below the pinch.

- The true location of a purification system is across the pinch. In other words, the purifiers must be located across the pinch.

- The hydrogen sources located above the pinch can be connected together to boost the purity of hydrogen supplied.

- Send the outlet stream of hydrogen-consuming processes to a purifier unit instead of sending directly to the site fuel system.

- Send the product stream of purifier to above the pinch and the residue stream to the site fuel system.

- The necessary condition of direct connection between hydrogen source and sink is defined as: the pressure of the source must be equal or greater than the pressure of the sink.

- The sufficient condition of direct connection between hydrogen source and sink is defined as: the purity of the source must be equal or greater than the purity of the sink.

- Do not mix streams at different purities. Such mixing may involve cross-pinch mass (hydrogen) transfer, and should not become a fixed feature of the design. For example, if the pinch is located at the purity of 70 %, mixing a stream at 90 % with a stream at 50 % creates a cross-pinch, and will increase the utility targets.

- Do not include utility streams (generally the hydrogen stream generated in hydrogen plant or imported form outside of the refinery) in the process data unless they are involved directly in the process or they cannot be replaced. One of the goals of using pinch analysis is to reduce the usage of utilities. Therefore, if utility streams are extracted in a similar way to process streams, they will be considered as fixed requirements and no opportunities of reduction in utility use will be identified. In some cases, utility streams can be included because it is not practical to replace them by any form of hydrogen recovery.

- Do not consider the existing plant layout. When selecting the inlet and outlet parameters for a process stream, existing equipment and plant topology should not be taken into account at first. True utility targets should be set regardless of the existing plant layout. Current plant hydrogen consumption can then be compared with minimum hydrogen targets. In retrofit of existing facilities, once these targets have been determined, plant layout

(existing compressors and piping, distances, etc.) needs to be taken into account in order to identify practical and cost-effective projects to reach or approach these targets.

- Identify hard and soft constraints on purity levels. For example, a hard constraint would be the inlet purity or pressure of a unit that cannot be changed in any way, while a soft constraint would be the discharge pressure of a new compressor, which is flexible. It is sometimes possible to change the potential for hydrogen recovery by changing some process parameters (purity, flowrate and pressure) at the data extraction phase.

The key to success is to use these rules as an initial framework, which provides guidelines and general directions. Breaking most of these rules will lead to cross-pinch mass (hydrogen) transfer, which would result in an increase in the hydrogen utility requirement beyond the targets. In other words, the difference between current hydrogen utility use and the targets is the sum of all cross-pinch inefficiencies. The targets can therefore be achieved by eliminating cross-pinch mass (hydrogen) transfer.

HYDROGEN NETWORK ECONOMICS

The objective function generally considered to design is to minimize total network cost. In this regard, both capital and operating costs need to be annualized to bring them to the same basis. The total cost is summation of the capital and the operating costs.

The capital cost is made up of the costs of new compressors, new purifiers and piping. The capital cost of a compressor is related to its power consumption:

$$C_{Comp} = \alpha_{Comp} + \beta_{Comp} \times Power \qquad (1)$$

Where, α_{Comp} and β_{Comp} are constants. The most common unit operations used for purifying hydrogen are pressure-swing adsorption (PSA) and membranes. Other methods do exist, for example, cryogenic separation or liquid absorption, but these are not as widely used. The cost of a PSA unit has been correlated as a simple linear function of the feed flowrate:

$$C_{PSA} = \alpha_{PSA} + \beta_{PSA} \times F_{in,PSA} \qquad (2)$$

Membrane costs are more complex to estimate as those that depend on product purity as well as the membrane pressure drop. The capital cost of a pipe is often expressed as a function of its length as well as the
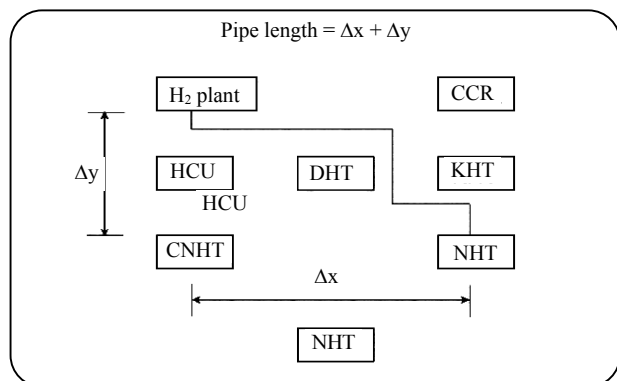
Fig. 5: Estimating piping lengths if no other information is available.

square of its diameter. The diameter squared, D^2, of a pipe is determined from the flowrate through it:

$$D^2 = \frac{4 \times F \times \rho_o}{\pi \times u \times \rho} \tag{3}$$

Where, u is the superficial gas velocity (usually 15-30 m/s). If no detailed information is available, then the length of piping, L, is estimated as shown in Fig. 5.

The cost per unit length of a pipe is a linear function of the cross-sectional area [3, 7], so that the actual cost is:

$$C_{Pipe} = \left(\alpha_{Pipe} + \beta_{Pipe} \times D^2\right) \times L \tag{4}$$

Different cost laws, i.e. different values of α and β can be used in different parts of the network to account for different conditions, such as temperature, pressure and different construction materials.

The operating cost of a hydrogen system is made up of the cost of the hydrogen utility (produced by hydrogen plant or imported from outside) plus the operating cost of purifiers and the cost of electricity used in compressors minus the value of the gas sent to site fuel system (burning gas has a value for a refinery). The operating cost of a hydrogen utility is assumed to be directly proportional to the utility flowrate. The compression power can be estimated from the stream properties, the inlet and outlet pressures and the flowrate being compressed, by the following equation:

$$Power = \frac{C_p \times T}{\eta}\left(\left(\frac{P_{out}}{P_{in}}\right)^{\gamma-1/\gamma} - 1\right) \times \frac{\rho_o}{\rho} \times F \tag{5}$$

- Where, C_P is the heat capacity of the stream at constant pressure;

-T is the inlet temperature;
- η is the compressor efficiency;
- P_{in} and P_{out} are the compressor inlet and discharge pressures, respectively;
- γ is the ratio of heat capacity at constant pressure to that at constant volume;
- ρ is the density of the gas at the design conditions and ρ_o is the density at standard conditions;
- F is the flowrate being compressed.

The value of the gas sent to the fuel system is calculated from the heating value of the gas. If the gas is assumed for simplicity to be a mixture of hydrogen and methane, the following equation can be used to estimate the fuel value (FV) per unit flowrate of gas:

$$FV = \left(y \times \Delta H^o_{C,H_2} + (1-y) \times \Delta H^o_{C,CH_4}\right) \tag{6}$$

Where, ΔH°_c is the standard heat of combustion and C_{Heat} is the cost per unit of heat energy [3].

CASE STUDY
The evaluation phase

The hydrogen network of an oil refinery has been shown in Fig. 6. Remember that the network is forced by the refinery to the design. Catalytic reforming and hydrogen plant are two main sources of hydrogen in the network, which supply hydrogen. Six hydrogen-consuming units exist in the network, which are diesel hydrotreater, cracked naphtha hydrotreater, jet fuel hydrotreater, naphtha hydrotreater, isomerisation plant and hydrocracker. All of the consuming units, except the isomerisation plant, have internal recycle compressors. Two compressors compress the outlet stream of hydrogen plant and catalytic reformer. These compressors supply the pressure required in the network. The streams, which their purity is low, are sent to the site fuel system. Hydrogen plant produces hydrogen by 44.5 MMscfd. Streams flowrate and pressure data are supplied in tables 1 and 2. In addition, network data are shown in table 3. Nowadays, hydrogen demand is increasing both to satisfy increased hydrotreating requirements and to process lighter fuel products from cracking processes in oil refineries [8].

To understand the distribution of hydrogen in the network, both the sources and the sinks of hydrogen are statistically analyzed. The sources and the sinks data are demonstrated through separated clustered column charts
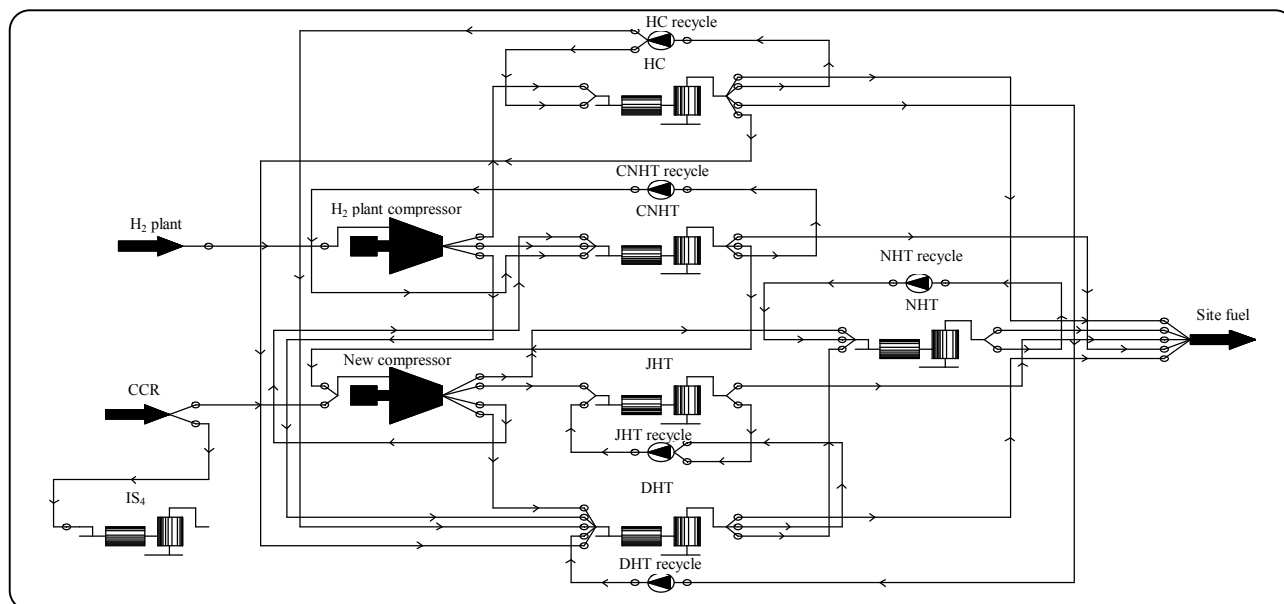
***Fig. 6: The hydrogen distribution network imposed by the refinery
(the network has been modeled in the REPOPT software environment).***

Table 1: Stream data for the hydrogen sources.

No.	Source units	Purity (Vol. %)	Flowrate (MMscfd)
1	H_2 plant	92.0000	44.5419
2	CRU	75.0000	23.4997
3	DHT	70.0000	10.1700
4	CNHT	75.0000	40.2200
5	JHT	65.0000	7.92000
6	NHT	59.9999	10.1400
7	HC	75.0000	96.9899

Table 2: Stream data for the hydrogen sinks.

No.	Sink units	Purity (Vol. %)	Flowrate (MMscfd)
1	DHT	75.2478	12.8700
2	CNHT	77.1074	44.9600
3	JHT	72.0612	12.2500
4	NHT	68.8201	15.6800
5	IS4	75.0000	0.04000
6	HC	80.2961	124.480

Table 3: Process data for the hydrogen network.

Surplus flow to fuel (MMscfd)	23.2017
Fuel purity (Vol. %)	68.5678
No. pinches	1
Pinch purity (Vol. %)	75.0000

in Figs. 7 and 8. Three columns have been allocated to all units that describe the state of a unit. The first column shows the purity of the hydrogen produced or consumed in a unit. The second column displays the total flowrate of each unit. Finally, the third column presents the total flowrate of the hydrogen produced or consumed in a unit. Total hydrogen flowrate is calculated by means of multiplying hydrogen purity by total flowrate. In other words, the third column is numerically equal to multiplication of the first column by the second column.

Fig. 7 shows that the hydrogen plant produces hydrogen at the maximum purity level and the NHT produces hydrogen at the minimum purity level. The first columns, which represent the purity of the units, affect the design. The source, which has the maximum purity, can supply all the sinks. The sink, which has the minimum purity, must be sent to the site fuel system. Purification is the way, which lets designer return the stream to the network. Hence, the purity levels must be considered well. The third columns can be regarded as suitable criteria to the design and especially in selection of purifiers. The maximum amount of hydrogen is usually produced by the HC and the maximum purity belongs to the hydrogen plant. Both the hydrogen purity and the total flowrate influence the total hydrogen flowrate. The hydrogen sinks data are represented in Fig. 8.
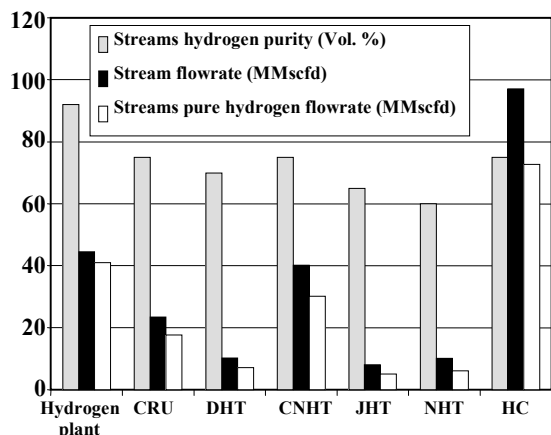
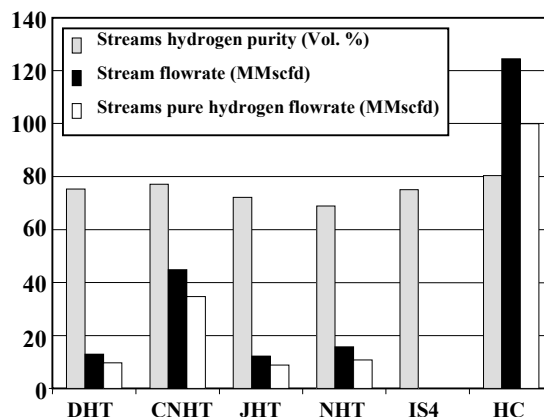*Fig. 7: The statistical analysis of the hydrogen sources.*

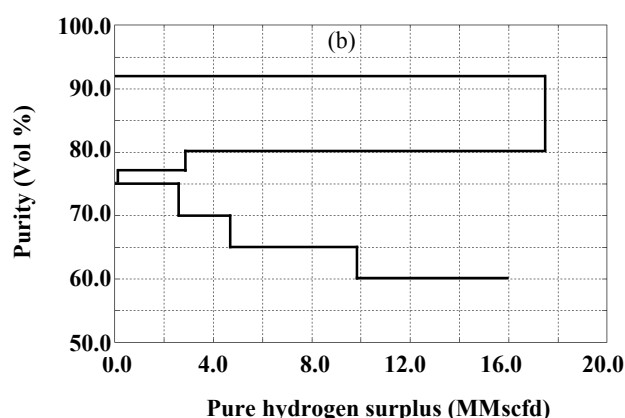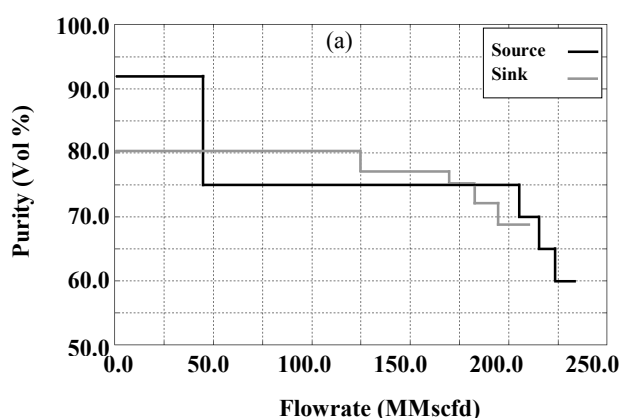*Fig. 8: The statistical analysis of the hydrogen sinks.*

Fig. 9: a) The hydrogen composite curves, b) the hydrogen surplus diagram of the network.

To allocate hydrogen sources to sinks properly, purity levels of hydrogen sinks must be considered well by designer. As shown in Fig. 8, all the purity levels of the sinks are between 60 to 80 volume percentages. This implies that the fluctuation of the purity levels is limited. Generally, any reduction in fluctuations makes the design easier. The third columns show that the maximum amount of the hydrogen consumption belongs to the HC and the minimum consumption of hydrogen belongs to the IS_4 that the amount of its consumption is near zero. After the HC, the CNHT is the most significant hydrogen-consumer in the network. Both the maximum purity and total flowrate of the hydrogen consumed belong to the HC.

The material balance on total gas for each of the hydrogen sinks is conveniently represented in a two-dimensional plot with flowrate of total gas on the horizontal axis and purity on the vertical axis [4], as

shown in Fig. 9a. The second necessary condition for feasibility of the hydrogen distribution system can then be stated as hydrogen surplus function that can be represented as a hydrogen surplus diagram [4], as shown in Fig. 9b.

The simulation phase

To specify the costs of the network, the network is simulated in the REFOPT[(1)] software environment. The main results of the simulation are shown in table 4. In addition to the items tabulated, total flowrate of the units, the cost of distribution and detailed data of the sinks are determined during the simulation phase. By the way, the objective function of the design is to minimize total cost.

The targeting phase

Fig. 9 shows that there is zero hydrogen surplus at the pinch purity (75 %) as well as at the point of the highest

Table 4: Cost data for the hydrogen network.

Total producer flowrate	68.0417	(MMscfd)
Total production cost	32101.8	(k$/y)
Total compress operating cost	1.10627	(k$/y)
Total compress investment cost	35000.0	(k$/y)
Total pipework cost	767.629	(k$/y)
Total site fuel value	9518.97	(k$/y)
Total network cost	58351.6	(k$/y)

(a)

(b)
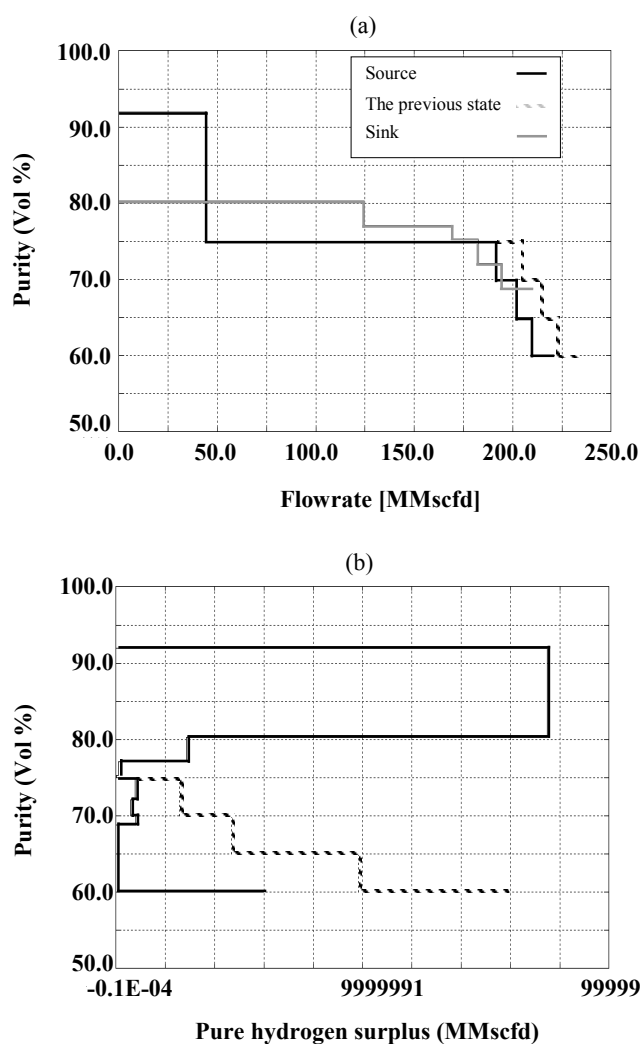
Fig. 10: a) The balanced hydrogen composite curves, b) the balanced hydrogen surplus diagram of the network (after modifying the CRU).

purity. This point at the highest purity is the value of hydrogen surplus at zero flowrate, which is always zero. Because this point can never have negative hydrogen surplus, regardless of the hydrogen supply, it is not a pinch; however, a pinch point might occur at the top of the network. In this case, the hydrogen surplus diagram shows a segment at zero hydrogen surplus from the purity of the first source to the purity of the sink underneath.

Fig. 10 illustrates the reduction in the utility flowrate required by the system until a pinch appears in the hydrogen surplus diagram. The reduction in the utility flowrate moves the remainder of the source curve toward the vertical axis. Consequently, there is a change in the overlap of the purity profiles. All of the areas of hydrogen excess are reduced, and all of the areas of hydrogen deficit are increased. The utility target is found when the reduction in the hydrogen surplus creates a pinch in the hydrogen surplus diagram. If the system is not feasible with all of the utilities at maximum capacity, then there are three debottlenecking options available at the network level to make it feasible: add more hydrogen production, increase (or start using) hydrogen imports, or add purification capacity. On the hydrogen-consuming process side, reducing the throughput in at least one of the processes above the unfeasible region is a costly short-term option that will also make the system feasible [4].

In this section, in order to specify targets, both the hydrogen plant and the CRU should be modified. Because the hydrogen surplus diagram shows an initial pinch point, it is impossible to reduce the flowrate of the hydrogen plant. Remember that the hydrogen plant is the first utility unit. If the flowrate of the hydrogen plant is reduced, the hydrogen surplus will become negative and the system will be infeasible. Hence, the CRU, the second utility unit, is modified to balance the hydrogen surplus into pinched condition. In this regard, its flowrate is reduced until a new (second) pinch point appears. The pinch point shows the minimum hydrogen utility. Fig. 10 shows the balanced hydrogen composite curves and surplus diagram. Generally, the information of source and sink composites and the information of accumulated hydrogen surplus are plotted on a Flowrate (X)-Purity (Y) graph.

Table 5 shows that the flowrate of the CRU has reduced from the initial value of 23.4997 to 10.2471 MMscfd. Because of Reduction in the flowrate of the CRU, the amount of surplus flow to fuel decreased from

Table 5: Stream data for the hydrogen sources (after targeting).

No.	Source unit	Purity (Vol. %)	Flowrate (MMscfd)
1	H$_2$ plant	92.0000	44.5419
2	CRU	75.0000	10.2471
3	DHT	70.0000	10.1700
4	CNHT	75.0000	40.2200
5	JHT	65.0000	7.92000
6	NHT	59.9999	10.1400
7	HC	75.0000	96.9899

Table 6: Process data for the hydrogen network.

Details	Before targeting	After targeting
Surplus flow to fuel (MMscfd)	23.2017	9.94908
Fuel purity (Vol. %)	68.5678	59.9999
No. pinches	1	1
Pinch purity (Vol. %)	75.0000	75.0000

Table 7: Stream data for the hydrogen sources (after the design).

No.	Source units	Purity (Vol. %)	Flowrate (MMscfd)
1	H$_2$ plant	92.0000	53.3211
2	CRU	75.0000	23.4983
3	DHT	70.0000	10.1700
4	CNHT	75.0000	40.2200
5	JHT	65.0000	7.92000
6	NHT	59.9999	10.1400
7	HC	75.0000	96.9899

Table 8: Process data for the hydrogen network.

Details	Before the design	After the design
Surplus flow to fuel (MMscfd)	23.2017	31.9794
Fuel purity (Vol. %)	68.5678	75.0003
No. pinches	1	0
Pinch purity (Vol. %)	75.0000	-

initial value of 23.2017 to 9.94908 MMscfd (by 57.12 %). In addition, its purity reduced from 68.5678 to 59.9999 volume percentages (by 12.495 %). The pinch point and its purity did not change during the targeting phase. Table 6 compares the surplus flows to fuel, the fuel purities, the number of pinches and the pinch purity before and after targeting.

The design phase

Recovery of hydrogen reduces the amount of hydrogen generated and decreases operating costs consequently. In this regard, new purifiers and compressors must be added to hydrogen network, which increases capital cost. Hence, there is an important trade-off between operating costs and capital cost that it has to be considered well during the design. Besides, the maximum potential of hydrogen recovery must be identified. Therefore, designer has to understand the network mass balance and utilize the hydrogen generation potential in a network. Generally, process engineers should undertake network design [9].

The objective of the design is to minimize total cost defined into the software. Selected solution method of the software is to use linear then non-linear systems. This means that the software uses the LP solver first then it uses the result of LP solver as an initialization for the NLP solver [10]. The network is designed through the REFOPT software. Fig. 11 shows the designed network.

The results of the design are supplied in tables 7 and 8. Table 7 shows purity and flowrate of the sources after the design. The previous data has not changed with respect to the initial values for the sinks. Table 8 supplies the major data of the network before and after the design. The results of the simulation are shown in table 9. In order to understand the mass balance of the network after the design, the hydrogen composite curves and surplus diagram are plotted again, as shown in Fig. 12.

Table 7 shows that after designing the network, only the flowrate of the CRU and the hydrogen plat have changed with respect to its initial value. The flowrate and the purity of the other units have not changed. Table 8 shows that all data have changed with respect to the initial values after the design. In addition, the surplus flow to fuel has increased from 23.2017 to 31.9794 MMscfd. Besides, the fuel purity has increased from 68.5678 to 75.0003 volume percentage. Removing hydrogen from fuel gas is beneficial due to increased

Table 9: Cost data for the hydrogen network.

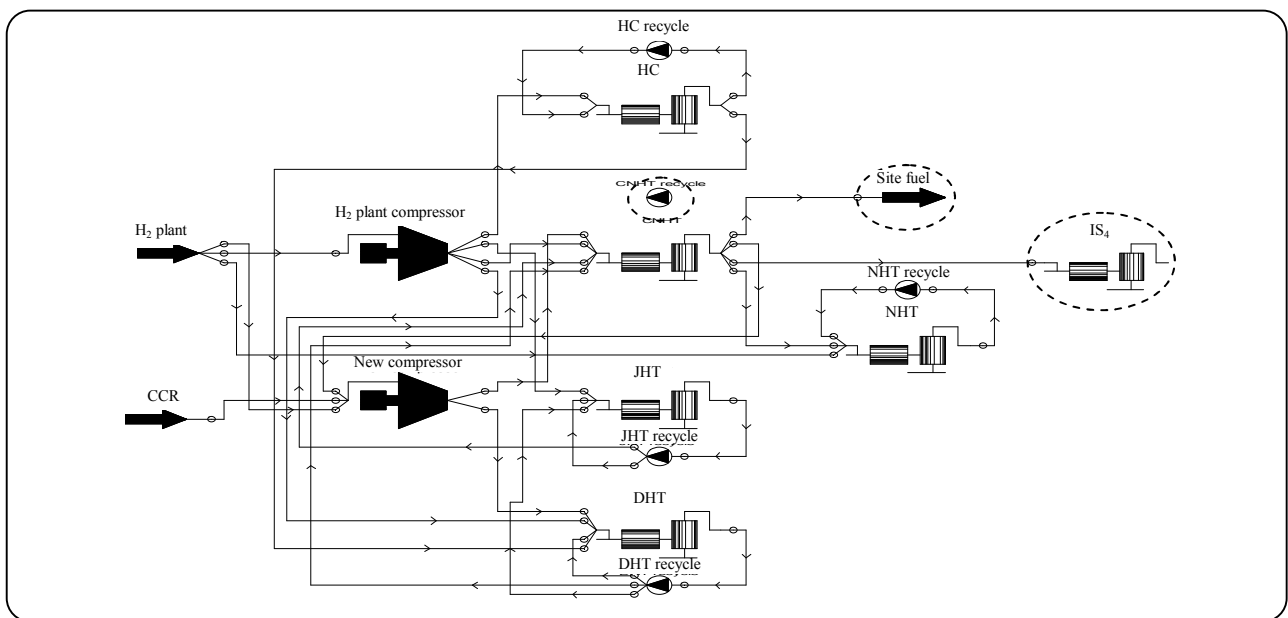| No.	Costs	Before the design	After the design
1	Total producer flowrate (MMscfd)	68.0417	76.8194
2	Total production cost (k$/y)	32101.8	38429.1
3	Total compress operating cost (k$/y)	1.10627	0.868806
4	Total compress investment cost (k$/y)	35000.0	30000.0
5	Total pipework cost (k$/y)	767.629	754.653
6	Total site fuel value (k$/y)	9518.97	12045.3
7	Total network cost (k$/y)	58351.6	57139.3

Fig. 11: The network designed.
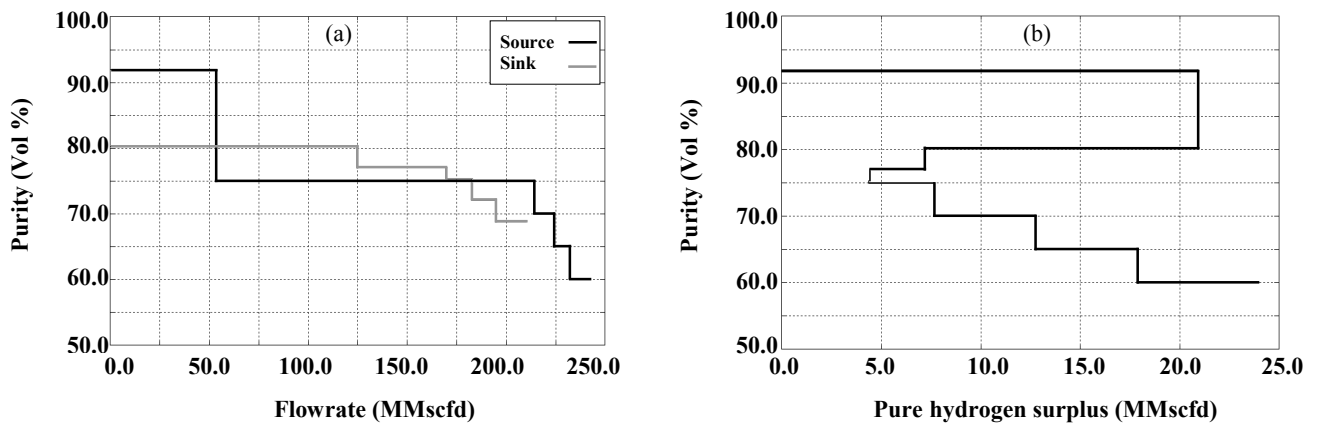
Fig. 12: a) The hydrogen composite curves, b) the hydrogen surplus diagram of the network designed.

calorific value, decreased NOx emissions, etc. [11]. The number of pinches in the network is zero. In other words, the network designed does not have any pinch point. Besides, it could be understood through the hydrogen surplus diagram after the design. In order to show the costs, the network is simulated again. To compare the results of the simulation before and after the design, all data have been supplied in table 9. The results are based upon the objective of the design, which is to minimize total cost.

Table 7 as well as the hydrogen surplus diagram imply that the flowrate of the hydrogen plant has increased from 44.5519 to 53.3211 MMscfd after the design, meaning that the network produces hydrogen more than its requirement by 8.78 MMscfd. In other words, the network is not fully balanced yet. Hence, in order to keep the design at the optimum point and to balance the network, only the flowrate of the hydrogen plant must be reduced. Remember that the outlet flowrate and purity of the CRU is fixed.

The design improvement phase

As mentioned above, the network produces hydrogen more than the amount required. The excess production of hydrogen decreases when the flowrate of the hydrogen plant is reduced. In addition, the total hydrogen production cost reduces consequently. In this regard, the current flowrate of the hydrogen plant reduced by 8.78 MMscfd which balanced the network. The hydrogen composite curves and surplus diagram of the network, after improving the design, are shown in Fig. 13. In order to understand the state of the network in the new conditions, the network's data have been compared before and after the design as well as after improving the design, shown in table 10.

As shown in table 10, the surplus flow to fuel has improved from the designed value, 31.9794, to 23.1994 MMscfd. Similarly, the fuel purity has reduced from 75 to 68.5666 percentages. In addition, a new pinch with 75 % purity has appeared in the network. This implies that the network is balanced now. In order to specify the costs of the network after improving the design, the network is simulated once more in the REFOPT software environment. The results of the simulation in different states are shown in table 11.

DISCUSSION

During the design of hydrogen networks, operating

Table 10: Process data for the hydrogen network.

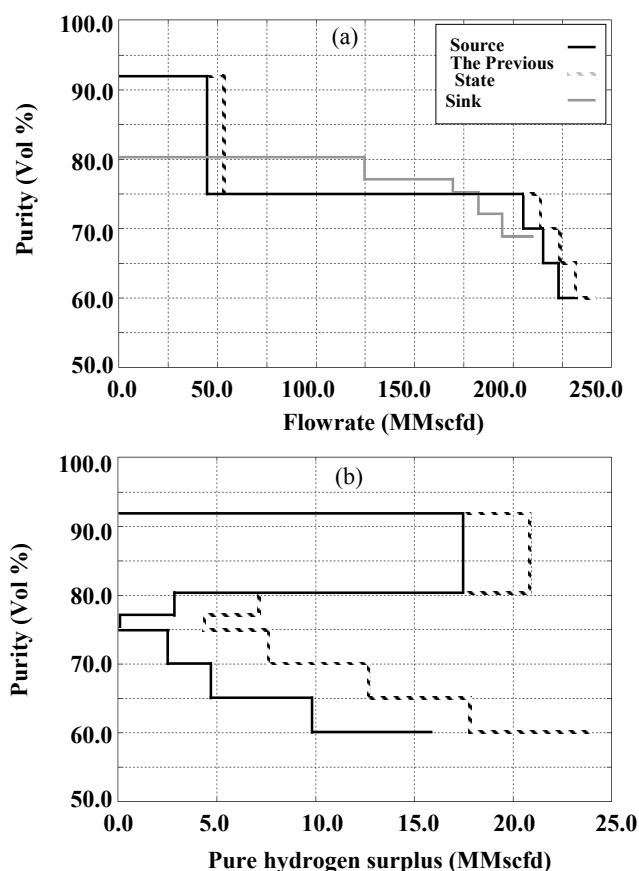| No.	Details	Before the design	After the design	After the design improvement
1	Surplus flow to fuel (MMscfd)	23.2017	31.9794	23.1994
2	Fuel purity (Vol. %)	68.5678	75.0003	68.5666
3	No. Pinches	1	0	1
4	Pinch purity (Vol. %)	75.0000	-	75.0000

Fig. 13: a) The hydrogen composite curves, b) the hydrogen surplus diagram (after improving the design).

units of the site have an effect on the design process. As a result, this is practically impossible to design the hydrogen networks independently. In this regard, the network imposed must be initially analyzed well. After the design, the work is not finished and the design results must be evaluated, which identifies hidden opportunities and reduces the network costs more. So the design improvement can be considered as final stage of the design hierarchy. In this study, the design was improved in the previous section which reduced the network costs remarkably.

Table 11: Cost data for the hydrogen network.

No.	Costs	Before the design	After the design	After the design improvement
1	Total producer flowrate (MMscfd)	68.0417	76.8194	68.0394
2	Total production cost (k$/y)	32101.8	38429.1	32101.2
3	Total compress operating cost (k$/y)	1.10627	0.868806	0.813874
4	Total compress investment cost (k$/y)	35000.0	30000.0	30000.0
5	Total pipework cost (k$/y)	767.629	754.653	736.632
6	Total site fuel value (k$/y)	9518.97	12045.3	12045.3
7	Total network cost (k$/y)	58351.6	57139.3	50793.4

CONCLUSIONS

In this paper, the application of process integration principles to the design of hydrogen networks was elaborated. Incidentally, a design hierarchy together with heuristics and guidelines were proposed. Thus, lack of strategy to the design was compensated. The design instructions are based upon heat integration concepts.

The major strength of this study is that the proposed criteria illuminate the way for engineers to design the hydrogen networks easily and maximise the amount of hydrogen recovered across the site during the design.

The main guiding principles of the design described in the work can be summarized as below:

1) Hydrogen must not be transferred across the pinch. 2) There must be no hydrogen utility used below the pinch. 3) Site fuel system must be located below the pinch. 4) The purifiers must be located across the pinch. 5) The pressure of the source must be equal or greater than the pressure of the sink. This is the necessary condition of connection between sources and sinks. 6) The purity of the source must be equal or greater than the purity of the sink. This is the sufficient condition of connectivity. 7) Do not mix streams at different purities. 8) Do not include utility streams in the process data unless they are involved directly in the process or they cannot be replaced. 9) Do not consider the existing plant layout. 10) Identify hard and soft constraints on purity levels.

The hydrogen network of the industrial case study was initially designed and then the design was improved. By the way, the proposed guiding rules ware applied all through the design process, which has the following effects: The hydrogen plant flowrate reduced by 8.78 MMscfd which decreased the total producer flowrate from designed value of 76.8194 to 68.0394 MMscfd. In addition, the total production cost reduced by $ 6.3279

million per year. The hydrogen plant compressor compressed one of the outlet streams of the hydrogen plant, so the reduction in the hydrogen plant flowrate reduced the total compress operating cost by $ 55 per year. Because the pipework cost laws relate the flowrate and length of the pipe to capital cost [10], hence the total pipework cost reduced by $ 18.021 thousands per year. For the reason that the network piping structure was not changed and one of the outlet streams of CNHT still goes to site fuel system individually, so the total site fuel value did not change during the design improvement. Generally, improving the design reduced the total network cost by $ 6.3459 million per year.

Nomenclatures

CNHT	Cracked Naphtha Hydrotreater
CCR	Catalytic Reformer
CRU	Catalytic Reforming Unit
DHT	Diesel Hydrotreater
HC	Hydrocracker
HP	Hydrogen Plant
HPC	Hydrogen Plant Compressor
IS_4	Isomerisation Plant
JHT	Jet fuel Hydrotreater
NC	New Compressor
NHT	Naphtha Hydrotreater
PSA	Pressure-Swing Adsorption
RC	Recycle Compressor

Symbols

C	Cost
C_P	Heat capacity at constant pressure
D	Pipe diameter
F	Flowrate

L	Pipe length
P	Pressure
T	Temperature
u	Superficial gas velocity through pipe
y	Hydrogen purity

Greek letters

γ	Ratio of heat capacity at constant pressure to that at constant volume
ΔH_c°	Heat of combustion
η	Compressor efficiency
ρ	Density

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