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# Hierarchical Targeting of Hydrogen Network System and Heat Integration in a Refinery

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Refineries are big consumers of energy and hydrogen. However, most current studies focus on the optimisation of H<sub>2</sub> Network or Heat Integration in refineries H<sub>2</sub> and heat are not considered together. The presented paper proposes a hierarchical targeting method for H<sub>2</sub> and heat using the Onion Model to solve this problem. H<sub>2</sub> networks belong to the inner layer of the onion, and heat recovery to the outer layer. The targets for H<sub>2</sub> recovery are obtained, followed by Heat Exchanger Network targeting. A case study of an oil refinery shows that both H<sub>2</sub> and energy can be significantly reduced. This study shows an approach to achieve a synergy of targeting H<sub>2</sub> and heat recovery. It has great significance to the energy-saving and emission reduction of an oil refinery.

### 1. Introduction

With the long-term rise of the energy demands, the requirements of energy-saving and emission reduction of petroleum refineries keep increasing. In the cost structure of the refining industry, energy consumption is a major item, next to crude oil purchase. It accounts for more than half of the cash operating expenses of an enterprise. Reducing energy consumption is of great significance for a refinery helping to reduce production costs, improve economic benefit and enhance competitiveness, promote environmental protection and socially sustainable development. The Pinch methodology (Klemeš, 2013) is based on thermodynamics to analyse energy distribution along with temperature in the process to find the "bottleneck" of an energy system and remove the "bottleneck". Pinch Analysis (Klemeš et al., 2018) is one of the bases of Heat Integration (HI) technology development. It is a method of targeting and prioritising the integration of process energy systems. The energy conservation methods of refineries have been developed from individual Process Integration (Chen et al., 2004) to the Total Site Heat Integration (Klemeš et al., 1997).

Hydrogen as an important raw material in refineries has been paid more and more attention. The H<sub>2</sub> demand for refineries keeps increasing rapidly, and the cost of H<sub>2</sub> is rising too. The problem of H<sub>2</sub> supply shortage is becoming increasingly prominent (Lou et al., 2019). How to use H<sub>2</sub> reasonably and make the best use of everything has become a new problem faced by refineries. It is necessary to analyse, optimise and control H<sub>2</sub> network to reduce production cost and improve H<sub>2</sub> utilisation. It has great significance to improve the economic benefit of the refinery. H<sub>2</sub> Pinch is an extension of Pinch Analysis for Heat Integration. H<sub>2</sub> Pinch analysis is an important method for H<sub>2</sub> network optimisation. Since it was put forward in the late 1990s (Alves, 1999), it has made a mature and steady development (Elsherif et al., 2015) in solving the bottleneck of H<sub>2</sub> networks. It has the advantages of being simple and intuitive, efficient and easy to understand. A variety of H<sub>2</sub> Pinch Analysis tools have been proposed, such as the H<sub>2</sub> Surplus Diagram (Alves, 1999), Material Recycle Pinch Diagram - MRPD (El-Halwagi et al., 2003), Average Pressure Profiles (Ding et al., 2011), Material Surplus Composite Curve (Saw et al., 2011) and H<sub>2</sub> network purification targeting (Zhang et al., 2016).

Following the foundational work on Total Site Heat Integration (Klemeš et al., 1997), there have been other studies, including algorithmic targeting (Liew et al., 2014) and extensions to trigeneration (Jamaluddin et al., 2019). Hydrogen Integration (Lou et al., 2019) is still considered separately. Few studies have considered both Heat Integration and H<sub>2</sub> optimisation in refineries. However, Heat Exchange Networks and H<sub>2</sub> Mass Networks

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coexist in most cases. Petroleum refineries are big consumers of both energy and hydrogen. It is necessary to consider both the Total Site Integration of heat and  $H_2$  simultaneously in refineries to minimise resource consumption and emissions.

This study aims to combine  $H_2$  and Heat Integration. It investigates the heat exchange network and mass network by applying the Onion Model (Linnhoff, 1994). A hierarchical targeting method of energy and  $H_2$  is proposed. A petroleum refinery is taken as a case study. The energy and hydrogen consumption are analysed and optimised by using the proposed method.

## 2. Methods

## 2.1 The Onion Model

Hydrogen in the refinery not only takes part in the hydrogenation reaction, but the related streams also need heating and cooling before/after the hydrogenation reaction. The H<sub>2</sub> Network (HN) and the Heat Exchanger Network (HEN) affect each other in refineries. In the current work, the heat recovery problem is formulated from the design of the H<sub>2</sub> network. The interface between the two integration phases is the Data Extraction which identifies and specifies the process streams for Heat Integration. At the stage of process design and targeting, the detailed information for building a model, which reflects the interactions between the reactors, the separators and the heat recovery subsystems, is not available. This makes the feedback link from the heat recovery subsystem to the Mass Integration subsystem indirect. Upon performing Heat Integration, the incentive for reduction of the energy demands can be evaluated, and core process changes can be initiated based on the energy analysis. The most commonly used hierarchy design for chemical processes is the Onion Model (Linnhoff et al., 1994), as shown in Figure 1. The Onion Model emphasises the ordered and hierarchical property of process development and design -- layer by layer design from the inside out.

In this study, the Onion Model is used for targeting the H<sub>2</sub> Network (HN) and HEN of a refinery. In a refinery, the H<sub>2</sub> Network includes H<sub>2</sub>-production units, H<sub>2</sub>-using units, H<sub>2</sub> purification units and H<sub>2</sub> pipe network systems. H<sub>2</sub>-production units mainly provide H<sub>2</sub> to the system, e.g. specialised H<sub>2</sub> production process, continuous reforming by-product H<sub>2</sub> production process. H<sub>2</sub>-using units include hydrotreating, hydrocracking, etc. H<sub>2</sub>-production units and H<sub>2</sub>-using units are mainly located in the "Reactor" layer. H<sub>2</sub> purification unit is a process to purify H<sub>2</sub> from low concentration to high concentration, such as Pressure Swing Adsorption (PSA), membrane separation, etc. The H<sub>2</sub> purification units are located in the separation and recycle layer. They decided on the H<sub>2</sub> distribution network, and the amount of H<sub>2</sub> needed of a refinery together. The H<sub>2</sub> Network spans two layers of "Reactor" and "Separation & recycle" (Figure 1). The object of process Heat Integration is the design of HEN and utility specifications. The Heat Integration in this paper mainly focuses on the Heat Exchange Network, which belongs to the "Heat Exchange Network" layer (Figure 1). The H<sub>2</sub> is in the inner layer of the onion, and the heat is in the outer layer of the H<sub>2</sub>. H<sub>2</sub> and heat can be hierarchy targeted. The H<sub>2</sub> network should be targeted first, and then HEN targeted.



Figure 1: The Onion Model (Linnhoff et al., 1994) of the production process

### 2.2 Integration of hydrogen network

H<sub>2</sub>-related units in a refinery can be divided into H<sub>2</sub>-producing units, H<sub>2</sub>-using units, H<sub>2</sub> purification units and H<sub>2</sub> pipe network systems. The development of H<sub>2</sub> Pinch provides an important theoretical and practical basis for H<sub>2</sub> Network optimisation. The H<sub>2</sub> Network targeting is to assign the maximum targets to improve the H<sub>2</sub> utilisation rate and reduce H<sub>2</sub> utility consumption. In this study, the Pinch Analysis was used to analyse a bottleneck of the H<sub>2</sub> Network and diagnose unreasonable H<sub>2</sub> use. The specific steps include: (i) the collection and extraction of H<sub>2</sub> source and H<sub>2</sub> sink streams data, (ii) H<sub>2</sub> Problem Table Algorithm, (iii) H<sub>2</sub> targeting and H<sub>2</sub> Pinch diagram. The H<sub>2</sub> Pinch diagram is drawn as follows: (a) All H<sub>2</sub> sources and H<sub>2</sub> sinks are sorted in descending order of H<sub>2</sub> concentration. (b) The flowrate and impurity load of all the H<sub>2</sub> sinks is plotted. The H<sub>2</sub> Sink Composite Curve

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(CC) is obtained by connecting them in order from small to large. (c) The H<sub>2</sub> Source Composite Curve can be obtained in the same way. (d) The H<sub>2</sub> Source CC is moved until it intersects the H<sub>2</sub> Sink CC. The H<sub>2</sub> Source CC is completely below the H<sub>2</sub> Sink CC in the overlap region. The intersection of the two CCs is the Pinch of the H<sub>2</sub> network.

The position of the H<sub>2</sub> Pinch, the minimum H<sub>2</sub> utility and the minimum waste H<sub>2</sub> discharge are determined by the impurity load -- flowrate diagram. To achieve the minimum fresh H<sub>2</sub>, the basic criterion to be followed in the H<sub>2</sub> network design is that the matching of H<sub>2</sub> source and H<sub>2</sub> sink cannot cross the Pinch. The matching of H<sub>2</sub> source and H<sub>2</sub> sink is carried out according to the purity, quantity, impurity content of H<sub>2</sub> source and the order of the same device and different devices. The method of Pinch Analysis is simple and intuitive. It shows and determines the minimum utility H<sub>2</sub> consumption and H<sub>2</sub> Pinch position of the H<sub>2</sub> Network.

#### 2.3 Heat Integration

In this paper, various refinery hydrotreating units are analysed for simultaneous reduction of the H<sub>2</sub> and thermal utilities. The Heat Integration is performed at two levels – starting with the processes. Total Site Profiles are then applied to analyse the refinery for Heat Integration between units. The specific steps include: (1) Complete Heat Integration within each unit. The Grand Composite Curve (GCC) is used to analyse the cold and hot streams in each process unit. The energy target of a single unit is obtained. The maximum reasonable energy matching within the unit is achieved. (2) Heat recovery can be realised through matching the remaining cold and hot utility demands between different processes. Those are represented by the Site Source and Sink Profiles (Total Site Profiles). (3) By plotting the steam-main levels against on the Total Site Profiles, the Total Site Composite Curves are obtained and used to analyse heat recovery.

### 3. Case study

A refinery is taken for a case study. Firstly, the  $H_2$  Network in the inner layer of the onion is targeted. The  $H_2$  Pinch is used to target the whole  $H_2$  Network. The  $H_2$  source and  $H_2$  sink streams data are shown in Table 1, based on an adaptation of Wang (2012). The stream SR1 with the highest purity of  $H_2$  is considered to be the  $H_2$  utility.

Stream type	Streams	H <sub>2</sub> concentration, vol%	Flowrate, kNm3/h	H <sub>2</sub> load, kNm <sup>3</sup> /h
H <sub>2</sub> source	SR1	99.9	22.75	22.73
	SR2	95.6	164.05	156.83
	SR3	92	137.05	126.09
	SR4	89	240	213.60
	SR5	88	5.07	4.46
	SR6	85	54.65	46.45
	SR7	84.3	5	4.22
H2 sink	SK1	99.9	10	9.99
	SK2	95.1	188.2	178.98
	SK3	93.5	11.82	11.05
	SK4	92	102.08	93.91
	SK5	90.3	280	252.84
	SK6	89.2	1.73	1.54
	SK7	88.2	4.08	3.60
	SK8	85.8	64.65	55.47

Table 1: The streams of an H<sub>2</sub> Network (HN) in a refinery

According to the method in Section 2.2, the Cascade Table is used for calculation, and the resulting H<sub>2</sub> Pinch diagram is shown in Figure 2. The targets for the H<sub>2</sub> Pinch, the required minimum utility H<sub>2</sub> and the minimum waste H<sub>2</sub> emission of the H<sub>2</sub> Network are obtained. The H<sub>2</sub> source Composite Curve (CC) and H<sub>2</sub> sink CC at the Pinch is shown in Figure 2a. According to the calculation, the H<sub>2</sub> Pinch concentration of the H<sub>2</sub> network is 89 %, as shown an enhanced plot segment (Figure 2b). At H<sub>2</sub> Pinch, the minimum utility H<sub>2</sub> required for the H<sub>2</sub> Network is 10.95 Nm<sup>3</sup>/h (Figure 2c). The minimum waste H<sub>2</sub> discharged is 32.13 Nm<sup>3</sup>/h (Figure 2d). It can provide guidance for the corresponding design and optimisation of the H<sub>2</sub> network. If the discharged waste H<sub>2</sub> can be recycled, it can further reduce the use of H<sub>2</sub> utilities.

The H<sub>2</sub> Pinch determines the target consumption and concentration of new and circulating H<sub>2</sub> in the different hydrotreating units. The H<sub>2</sub> streams need to be heated or cooled before or after hydrotreating. The H<sub>2</sub> Network Integration affects the optimisation of Heat Exchanger Network. Now comes HEN targeting. The hydrogen installations of a refinery are also big energy consumers. Hydrotreating (Robinson and Dolbear, 2006) unit is

mainly the hydrotreating of distillate oil, including gasoline, diesel and kerosene. The process is roughly the same. The raw oil is mixed with circulating  $H_2$  and heated to a certain temperature before entering the reactor for hydrotreating reaction. After the reaction product is separated from the circulating  $H_2$ , the product is removed from the unit by stripping or fractionation. The specific process flow is slightly different, but the process operating conditions are significantly different. The energy consumption structure of the unit varies greatly.



Figure 2:  $H_2$  Network Pinch diagram for (a) the  $H_2$  CC; (b)  $H_2$  Pinch; (c) minimum utility  $H_2$ ; (d) minimum waste  $H_2$  discharge

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Stream type	Stream name	T₅, °C	Tt, ℃	$\Delta H$ , MW
Hydrotreating l	Jnit A	-	-	
Hot stream	Hot A1	360	35	17.91
	Hot A2	165	40	5.23
Cold stream	Cold A1	35	168	4.73
	Cold A2 (H <sub>2</sub> )	70	125	0.59
	Cold A3	40	103	1.92
	Cold A4	103	220	7.26
Hydrotreating l	Jnit B			
Hot stream	Hot B1	272	207	10.3
	Hot B2	205	36	2.33
	Hot B3	197	40	15.04
	Hot B4	140	42	1.94
Cold stream	Cold B1	45	166	10.93
	Cold B2 (H <sub>2</sub> )	50	188	0.56
	Cold B3	164	243	15.54
	Cold B4	36	170	1.26
Hydrotreating l	Jnit C	-	-	-
Hot stream	Hot C1	395	220	19.12
	Hot C2	225	95	12.67
	Hot C3	220	50	3.31
	Hot C4	155	40	1.47
	Hot C5	220	50	0.4
	Hot C6 (H <sub>2</sub> )	395	220	8.53
Cold stream	Cold C1	120	370	26.1
	Cold C2 (H <sub>2</sub> )	73	370	20.26

Table 2: The streams of three different hydrotreating processes

In this study, the Heat Integration analysis and optimisation of three different hydrotreating process in a refinery is studied. The process streams of three different hydrogenation units are shown in Table 2 based on an adaptation from (Zhang et al., 2013). Streams Cold A2 (H<sub>2</sub>), Cold B2 (H<sub>2</sub>), Hot C6 (H<sub>2</sub>) and Cold C2 (H<sub>2</sub>) are H<sub>2</sub> extracted from the H<sub>2</sub> Network. Energy analysis is carried out in each unit according to the method described above. The GCCs of all units are given (Figure 3). It can be seen that unit A still had 8.25 MW excess heat source after adequate heat recovery in the unit (Figure 3a). Unit B had a 2.94 MW redundant heat sink and 4.26 MW redundant heat source (Figure 3b). Unit C also had a 3.96 MW redundant heat sink and a 3.10 MW redundant heat source (Figure 3c). The Heat Integration between units has great potential for energy saving.



Figure 3: The GCC of three different hydrogenation processes for (a) Unit A; (b) Unit B; (c) Unit C

Total Site Analysis is used to integrate the heat between the units. Total Site Profiles with utilities are shown in Figure 4. The solid red curve represents the Site Source Profile, and the solid blue curve represents the Site Sink Profile. The utility includes high-pressure (HP) steam (55 bar, 270 °C), medium-pressure (MP) steam (8 bar, 170 °C), and cooling water supplied at 20 °C and returned to the utility system at 30 °C. The blue dotted curve represents the site source CC, and the red dotted curve represents the Site Sink CC. It can be seen that the heat source can generate 4.41 MW HP steam and 1.57 MW MP steam. It also needs 9.64 MW of cooling water. The heat sink requires 5.09 MW HP steam, and 1.81MW MP steam.



Figure 4: The Total Site Composite Curves (TSCC) (a) Total Site Profiles with utilities; (b) Hot Utility Generation Composite Curve (HUGCC)

To identify the minimum cold and hot utilities for inter-unit integration, the Site Source CC and Site Sink CC were combined to obtain the Hot Utility Generation Composite Curve (Figure 4b). The minimum hot utility is 0.93 MW, and the cold utility is 9.4 MW. Before the Interplant integration, the minimum hot utility required for the three processes was 6.91 MW and the minimum cold utility required was 15.62 MW. This shows a significant utility saving potential – up to 86.5 % of hot utilities and 39.8 % of cold utilities. This would require further analysis and economy calculation (for extra Heat Exchanger (HE), piping, possible extra pressure drop, safety issues).

### 4. Conclusions

In this paper, the problem of combined  $H_2$  and Heat Integration for oil refineries has been considered, applying the Onion Model. This has resulted in a hierarchical targeting procedure. Firstly,  $H_2$  Pinch targets are obtained. They are followed by the identification of the Heat Integration problem and the HI targeting at two levels –

process and site. The Total Site Composite Curves are used to target the HI of the refinery. The case study results show that the H<sub>2</sub> network targeting can guide the optimisation of the H<sub>2</sub> Network, simultaneously reducing H<sub>2</sub> utility demands and the waste H<sub>2</sub> emissions. At the Total Site level, the targets show that the maximum reduction can be 86.5 % for hot utilities and 39.8 % for cold utilities. The unidirectional method, which combines the Onion Model and Pinch analysis, is simple to use and has the advantages of graphic visualisation.

It has been demonstrated that the combined targeting of H<sub>2</sub> and thermal utilities for a refinery can save significant amounts of both types of resources, leading to potential environmental and economic benefits. Future steps should consider pressure drops and investments. This should include the analysis of economic benefits, piping, pressure drop, and safety issues.

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