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Review article

State-of-the-art of hydrogen management in refinery and industrial process plants

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ABSTRACT

Hydrogen has been identified as the best raw material for hydro-treating conventional fuels such as gasoline, kerosene and diesel in oil refineries. Hydrogen is actually created and is in short supply. A new approach to minimize hydrogen waste in the industry that already consumes a large percentage of the world's existing hydrogen and to offset the cost of that hydrogen by recycling is an issue receiving much attention. This paper reviews the concepts and the available tools for hydrogen management in industrial processing plants and oil refineries. In addition, the paper emphasizes the challenges faced by refinery product quality regulations.

1. Introduction

The challenges of hydrogen management, product market changes and the strict regulations on fuel specifications are problems and concerns for both industry and researchers. Sulfur and aromatic compounds in gasoline and other traditional hydrogen sources, such as catalytic naphtha reformers, have made refineries encounter uncertainty with the rapidly changing market and deteriorating crude oil. On the other hand, hydrogen is considered as the next generation alternative renewable energy source, particularly for the freight sector. Because of the scarcity of hydrogen resources and growing concerns about environmental sustainability, better management, and utilization of hydrogen as well as reduction of hydrogen waste have become important issues in the process industry. Rising demand and shortage of hydrogen supply is a significant problem for refineries throughout the world.

The cost of hydrogen is rising; it is considered the largest cost after the cost of crude oil in refineries (Jiao et al., 2012b). In addition, research in oil refinery hydrogen integration has included wider areas than the determination of targets for minimum flowrate of fresh utility hydrogen. The scope has been extended to include hydrogen production strategies, design of hydrogen networks (Liu and Zhang, 2004; Jinsong and Zhaoxia, 2009) and optimal placement of purification units (Liao et al., 2010) along with process optimization for the scheduling and distribution of hydrogen (Van den Heever and Grossmann, 2003). The requirement of robust design, better optimization (Jiao et al., 2011), quantitative and qualitative analyses of hydrogen distribution systems have encouraged process engineers to identify possible options for the maximization of hydrogen reuse and the minimization of fresh hydrogen utilization. This review is concerned with research studies on optimizing hydrogen use in refineries and industrial plants. Although several traditional and hydroprocessing routes for the production of hydrogen have been practised commercially, there is still a need for improved and efficient management of hydrogen within the hydroprocessing systems. The focus of this review is threefold. First, the review covers the mathematical optimization approaches for hydrogen network management, for which a significant amount of research work has been performed. Second, the review covers papers that were published in available journals as well as important international conferences. Third, the review only covers hydrogen network management for continuous processes. In the concluding section, various suggestions for revamping the hydrogen network in oil refineries have been proposed as guidelines for further investigations.

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2. Scope of the review

The purpose of, this work is to review most of the accessible literature on hydrogen management in refineries and industrial processing plants and identify the current state-of-the-art of the technologies. The study starts by summarizing the main ideas in more than 60 published studies, classifying the publications into different groups and expanding their particular features independently in the subsequent sections.

Most of the research on refinery hydrogen management aims at improving the quality of the petroleum product. Recently, analysing the possibility of introducing hydrogen as a fuel in the passenger transportation sector by remodelling the existing fuelling stations has also been investigated. The information we have collected is mainly from publications in major journals (Table 1), conference proceedings (Table 2), books, guidelines, and standards. The number of published studies gradually rose over the years from 1982 until 2014 with the majority of publications focussing on developments in hydrogen system integration and management. As shown in Fig. 1, 80% of the research was published between 2002 and 2014. Most of this work concerns the development of mathematical optimization techniques, as shown in Fig. 1.

Many different methods have been developed for the integration of hydrogen systems not only in petroleum refineries but also in petrochemical complexes. Fig. 2 shows that the reviewed literature consists of three main research topics. However, other particular topics within each main research area may be expanded easily, as will be shown in the next sections.

It is useful, informative and interesting to specify the earliest contributions to the study of refinery hydrogen network management. This paper follows the continuous progress in hydrogen management over the years to the present, highlighting the techniques used in optimizing hydrogen as the most used element in the petroleum refining value chain. Pinch analysis is one of the most widely used optimization techniques for management and optimization of utility systems (Foo, 2009). Section 3 begins with an evaluation of the technologies used for hydrogen purification and recovery in processing plants and oil refineries. Section 4 provides a brief overview on the problems and issues of refinery hydrogen systems, while subsection 4.1 discusses the detailed issues and methodologies for hydrogen management and integration in oil refineries. The review includes a detailed discussion of various techniques developed for optimal hydrogen distribution in oil refineries. Finally, the future research directions in hydrogen management are outlined at the end of the review.

Table 1
Sources of most published papers.

Source	Number
Advances in Environmental Research	1
Advanced Control of Chemical Processes	1
Computer and information Science	1
Chemical Engineering Transactions	1
Chemical Engineering Journal	2
Chemical Engineering Research and Design	6
Chemical Engineering Science	5
Chinese Journal of Chemical Engineering	2
Computer Aided Chemical Engineering	4
Computers & Chemical Engineering	2
Control Engineering Practice	2
Energy	2
AIChE Journal	3
Applied Energy	2
Energy Conversion and Management	1
Energy Policy	1
Evolutionary Computation, IEEE Transactions on	1
Industrial and Engineering Chemistry Research	13
International Journal of Hydrogen Energy	7
International Journal of Engineering	1
International Journal of Modelling and Optimization	1
Iranian Journal of Chemistry & Chemical Engineering	1
Journal of Cleaner Production	3
Operations Research	1
Petroleum and Coal	1
Resource Conservation and Recycling	1

3. Hydrogen production, use and purification in process plants and oil refineries

Hydrogen is currently being used in many industries from chemical and refining to metallurgy glass and electronics. The enormous breadth and extent of these applications make hydrogen an important element in refineries and processing plants. The new fuel specifications as well as economic forces drive refineries to process more heavy crudes, at the same time demanding improved hydroprocessing and hydrotreating processes, where hydrogen plays a major role. Hydrogen has also been used as a reactant to remove heterogeneous atoms, such as sulfur and nitrogen, in petrochemical productions such as methanol production, plastic recycling, oil and fat hydrogenation, fertilizer production and also as an oxygen scavenger in float glass manufacture (Ramachandran and Menon, 1998). Recently, hydrogen has been proposed as a future fuel for automobiles in the transportation sector. For fuel cell vehicles, storage of hydrogen fuel is a key technology towards the hydrogen society (Mori and Hirose, 2009; Agnolucci, 2007).

Process innovation and factors that increase hydrogen production in refineries and process plants continue to be crucial and challenging. They are critical in the production of hydrocarbons, and in many instances, still a challenge in terms of attaining target levels of profitability. The extensive use of hydrogen in refineries and petrochemical plants has become an important research area for the design and utilization of efficient hydrogen networks. There have been rapid innovations and advances in many of the key issues of hydrogen production, use and recovery in process plants and oil refineries.

Hydrogen production from conventional and supplementary sources falls into three general practices, refinery catalytic naphtha reforming, steam reforming of natural gas or naphtha and partial oxidation. However, the measures taken for energy conservation in refineries frequently results in an excess of fuel gas which in consequence is a waste of valuable hydrogen content. These practices, along with the shortage of hydrogen production, the change in crude oil properties and product states from the design basis, can make it difficult for even a new refinery to run efficiently (Aitani, 1996).

The need to obtain cleaner burning fuels, the reconfiguration of refinery processing plants for better production of gasoline, kerosene and diesel with less sulfur and aromatic content and the increased demand for hydrogen in the refinery hydroprocessing and hydrotreating units are the major challenges for the proper balance to optimize utilization of hydrogen. The proper balance of hydrogen assets in an environment that currently demands cleaner burning fuels makes restricting processing plants the major challenge facing the industry today. Hydroprocessing and hydrotreating in the production of crude oil products take more and more of the available hydrogen, while increasing the costs of their operations.

The demand for hydrogen in the refinery hydroprocessing units has been continually growing to sustain the operation requirements. However, opportunities for better utilization and recovery of hydrogen can be made by the use of purification techniques, for example, pressure swing adsorption (PSA), membrane systems and cryogenic systems. These techniques have different mechanisms for hydrogen separation from hydrocarbons which summarized in Table 3.

Table 2
Conference proceedings.

Advanced control of chemical processes
2011 International symposium on Advanced Control of Industrial Processes (ADCONIP)
AIChE 2001 Spring meeting, April, Houston TX
European Symposium on Computer Aided Process Engineering
International Conference on Information and Automation 2009 ICIA'09
2005 IEEE International Symposium on Intelligent Control and 13th Mediterranean Conference on Control and Automation

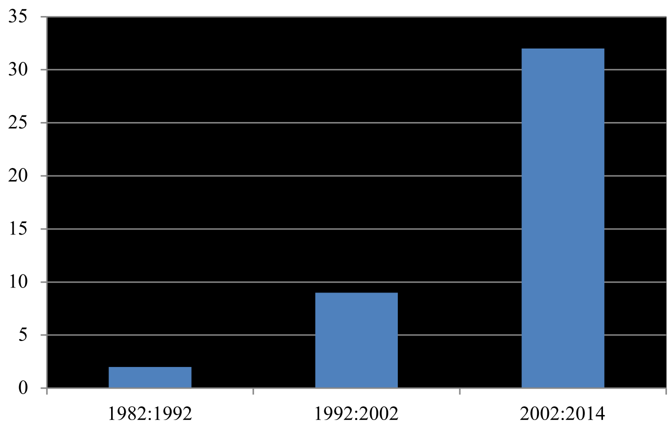

Fig. 1. Trend of related hydrogen management publications.

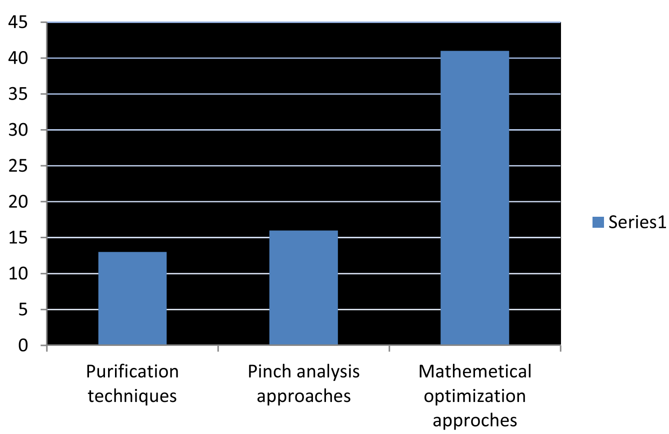

Fig. 2. Hydrogen management research topics.

Towler et al. (1996) illustrated the cost correlation of hydrogen recovery from refinery off-gas using purification units. The methodology proposed in their work facilitates the cost of hydrogen recovery by PSA and membrane permeation using a linear programming (LP) model to supplement the information in a graphical display. Work performed by Zhou et al. (2002) represents a novel PSA technology that is fitting to both the operating conditions of low pressure and avoiding the other limitations of PSA technologies, which they consider to be the purging ratio and separation performance at low operating pressure. Rabiei (2012) discussed the difference between the three most common purification and recovery units for hydrogen in the oil refinery, which are PSA, membrane processes and cryogenic processes. This work investigates the cost for process recovery selection and the capital investment of PSA and membrane processes in terms of hydrogen content and operating pressure of the refinery.

Hallale and Liu (2001) have produced a valuable process that interprets the work done by Alves (1999) by considering the purifiers as interception units that upgrade the purity of the hydrogen source taking into account the pressure constraints as well as existing compressors. In order to gain the highest recovery from purification, Liu and Zhang (2004) have proposed a hybrid plan and discussed a detailed strategy for purifier selection that includes both PSA and a membrane. The process deploys a mixed integer linear programming (MILP) model to integrate purification units with refinery hydrogen. The optimized superstructure allows engineers to define the available options for better purification of refinery off-gases and placement of purifiers. Liao et al. (2011b) presented a rigorous, systematic approach to the problem of the optimal placement of purifiers in a hydrogen system. The process identifies hydrogen network targets with one purifier, taking into account minimum hydrogen utility consumption and the purifier feed concentration. Liu et al. (2013a) have presented a conceptual method using a quantitative relationship diagram for a hydrogen conservation strategy with purification reuse/recycle, to identify the optimal Purification Feed Flow-rate (PFFR) and the corresponding maximum Hydrogen Utility Saving (HUS).

The growing demand for refinery hydrogen is being addressed by modification or development of new production and recovery routes. However, the hydrogen content and operating pressure of refinery off-gases have a large influence on both the choice of process recovery and capital investment. Foo and Manan (2006) have made an extensive study of gas resources allocation targets, evaluating both membrane and PSA as purifiers for a conventional hydrogen system in an oil refinery. However, in their approach using the Gas Cascade Analysis method, which is practical to

Table 3
Hydrogen purification technology comparison source (Sabram et al., 2001).

Features	Adsorption	Membranes	Cryogenics
H_2 Purity	99.9%+	90–98%	90–96%
H_2 Recovery	50–92%	85-95%	90–99%
Feed Pressure	150–600psig	300–2300psig	˃75–1100psig
Feed H_2 Product	˃40%	˃25–50%	˃10%
H_2 Product Pressure	Feed	˂1/3 Feed P	Feed/Low Pressure
H_2 Capacity	1-200 MM scfd	1-50 MM scfd	10-75 MM scfd
Pretreatment Requirement	None	Minimum	CO_2,H_2O Removal
Multiple Products	No	No	Liquid HC s
Power Requirements	None/Fuel	H_2/Feed	None/H_2/Refrigeration
Capital Cost	Medium	Low	Higher
Scale Economics	Moderate	Modular	Valid

determine minimum flow-rate targets, pinch point allocation and resource allocation targets, they found that even if the membrane gives higher gas recovery compared with PSA, it may not necessarily be a better selection in minimizing the overall flow-rate of the hydrogen system in an oil refinery. Shahraki et al. (2005) have addressed the minimization of refinery hydrogen consumption by making use of pressure constraints and all the possible site-wide connections of the piping and optimum placement of purifiers. This approach is claimed to be suited for revamping industrial systems.

The practical conditions of recovery pressure, payback rate and network structure for purification process selection in refineries have been addressed by Smith et al. (2012). The work emphasize the effect caused by changes in hydrocarbon impurities in the make-up stream because of the assumption of fixed hydrogen to oil ratio and hydrogen partial pressure at the reactor inlet of the hydrogen consuming units. Such changes lead not only to change in the flow-rate and composition of recycle and purge hydrogen streams, but also a change in the vapour–liquid equilibrium in the downstream flash separation drum. The authors propose an approach that integrates the two general categories of hydrogen system optimization for either a new design or retrofit of the hydrogen system. Liao et al. (2010) have also established a state-space superstructure that includes the use of a shortcut method to represent all of the possible placements of compressors and purifiers. This work, however, makes use of a mixed integer nonlinear programming (MINLP) model considering PSA and membrane as the only purifiers used to quantify the total annual cost for better understanding of the trade-off between the operating and capital costs.

A by-product hydrogen system within a petrochemical complex has been designed by Jeong and Han (2011) to determine the minimum fresh hydrogen and recovery hydrogen needed for production. The system is described using a hydrogen pinch study by means of material exchange systems to address the hydrogen recycling system between the hydrogen producing plant and the hydrogen consuming plant. Other topics considering issues of strategic planning of the hydrogen supply chain under uncertainty in the operating costs (Almansoori and Shah, 2006; Sabio et al., 2010), transition towards a large-scale hydrogen supply infrastructure in the transportation sector (Murthy Konda et al., 2011) and electric system management through hydrogen production (Mansilla et al., 2012) have also been discussed.

4. Overview of oil refinery hydrogen network management and integration

Environmental legislation and safety policies, growing demands of product quality as well as increasingly competitive markets necessitate continuous development of hydrogen networks in oil refineries and petrochemical complexes (Gomes et al., 2009; Jeong and Han, 2011; Shariati et al., 2013). In addition, ever growing industrialization has made the business of hydrogen grow as the world's consumption of refinery products increases which is shown in Table 4. More likely, the need for enhanced environmental standards and cleaner automotive fuels call for improved hydrogen production and consumption within the refinery processes. The complexity and breadth of refinery units still make it very challenging to operate efficiently. On the other hand, efficient use of hydrogen in the refinery represents a major key part of operational management for refinery processes. Therefore, the attentions of management strategies, process modelling and day-to-day optimization of operational changes are critical for development opportunities and profitability (Sarabia et al., 2012). Oil refinery hydrogen management is rapidly developing as an area for research not only for educational but also for both commercial and industrial interests.

Table 4
World hydrogen demand, billion cubic metres.

				Annual growth	
Item	2003	2008	2013	2003–2008	2008–2013
World hydrogen demand	307.0	402.0	475.0	5.6	3.4
North America	96.0	122.9	132.0	5.1	1.4
Western Europe	61.0	73.3	78.0	3.7	1.3
Asia/Pacific	87.2	119.6	157.0	6.5	5.6
Other Regions	62.8	86.5	108.0	6.6	4.5

The state-of-the-art for hydrogen management strategies was first addressed by Simpson (1984), and since then the management of hydrogen has shown a high industrial impact and value in petrochemical complexes and refineries (Shariati et al., 2013; Jagannath et al., 2012). Therefore, many technologies and methodologies have evolved considerably over the last decades in response to energy saving and management. These methodologies can be illustrated under the umbrella of either targeting methods or mathematical approaches that are based on the superstructure for the design of the hydrogen system (Jia and Zhang, 2011).

Primary work addressing the graphical targeting process was developed by Towler et al. (1996), where the problem of process heat recovery was considered. That work provides a graphical insight into the idea of both refinery hydrogen profitability and the added value for hydrogen consuming processes. Another approach towards improved hydrogen utilization and management in the oil refinery is the study and design of a refinery hydrogen system by Alves (1999). The process is based upon pinch technology (Linnhoff and Flower, 1982), in which the identification of sources and sinks of hydrogen in the process are related to hot and cold streams identification in heat exchanger network synthesis reported by Hallale and Liu (2001). The pinch analysis in the aforementioned methods is illustrated by plotting the flow-rate vs the purity of all sources and need. Illustration of such an idea is known as the hydrogen composite curve. This two-dimensional plot of hydrogen flow-rate vs-purity yields the hydrogen surplus and debt profile, which is described by the hydrogen surplus plan.

Graphical methods in hydrogen network management give an important insight into the scope of refinery process integration and are accepted for dealing with the theoretical targeting for minimum hydrogen use (Agrawal and Shenoy, 2006; Foo and Manan, 2006). Moreover, several different methods have been published on the management of refinery hydrogen networks (Gutierrez et al., 2010; Khajehpour et al., 2009, Liu and Zhang, 2004) with different design superstructures supported by the use of hydrogen pinch analysis. However, many of these have the limitation of requiring an initial assumption of hydrogen flow-rate (Alves and Towler, 2002; Zhenmin, 2003). Some of the graphical methods for the targeting and design of hydrogen systems are rather tedious and iterative in nature because they require elaborate shifting of streams in the concentration vs flow-rate diagram (Nelson and Liu, 2008). These drawbacks can be overcome by mathematical programming techniques, particularly for large complex problems like a petroleum refinery that involve many constraints, such as pressure, numerous hydrocarbon components and reaction constraints. Therefore, it is arguably more important to take into account all the possible practical constraints as part of an integrated refiner process in order to exploit the interaction and synergies that may exist (Hallale and Liu, 2001; Salary et al., 2008).

Optimization of refineries is a complex business that involves different operations depending on the type of crude to be refined and the desired products to be processed. For these reasons, it is highly unlikely that two refineries are alike. Furthermore, crude oil refining is not a linear process there is a wide assortment of variables and parameters to manage. As a result, mathematical methods are required for refinery processes that are either hydrogen consumers or producers. Mathematical programming techniques have emerged principally to overcome the limitation of graphical methods which are restricted to flow-rate and purity constraints. The uncertainties of many impurities, hydrogen waste minimization, pressure constraints and optimal placement of new equipment such as compressor and purification units have been considered in a number of research articles (Liu and Hallale, 2001; Girardin et al., 2006; Fonseca et al., 2008; Jinsong and Zhaoxia, 2009; Khajehpour et al., 2009).

Some of these considerations have also taken into account the problem of reducing the cost of hydrogen consumption by means of unanticipated measures that may have an impact over a time frame. For example, Zhou et al. (2013) and Jiao et al. (2012c) discussed the reduction of greenhouse gases because of their environmental impact and also the uncertainties of an unsteady supply of hydrogen and the different hydrogen needs that could be tackled by reliable decision making scenarios. Refineries generally take several measures to improve on cost and energy savings. These measures might include increasing the yield of the hydrogen plant, expansion of the hydrogen plant, building new hydrogen plants, constructing new hydrogen purifiers, purchasing hydrogen and retrofitting the hydrogen system. Possible measures, with their associate investment, operating and total costs, are shown in Table 5.

The management and optimal utilization of refinery hydrogen systems can play a major role in contributing to the solutions related to the aforementioned measures because they draw increasing attention to an alternative energy source. Han et al. (2013) addressed an optimization model that is concerned about maximizing the total net profit of the hydrogen supply system that involves constraints of how much hydrogen can be produced and where it can be stored. Jiao et al. (2012a) proposed two improved systematic mathematical techniques that are based on a two-step approach (Putra and Amminudin, 2008) and a simultaneous optimization approach, respectively, to retrofit the hydrogen system in a refinery. The sequential two-step approach is used to optimize the hydrogen purification system with the goal of maximizing the value created by the purifiers and minimizing the operating cost and the annualized capital cost, and 2 to minimizing the total annual cost, including both annualized capital cost and operation cost. However, the simultaneous optimization method is used to optimize the hydrogen purification and hydrogen supply networks simultaneously for retrofitting the entire hydrogen system of the plant.

4.1. Methodologies for hydrogen management and integration in oil refineries

The goal of this section is to evaluate the background and development of hydrogen management issues, as well as interpret and introduce related work that has been performed by both academic and industrial sectors in the field of hydrogen management and utilization. The term "management" in this context refers to the requirement for accurate control and awareness of all possible issues and opportunities relating to the hydrogen distribution system (Liu and Zhang, 2004). It includes distribution, production, consumption, recovery and optimization of hydrogen. In addition, the development of capital investment and operational strategies, which often involve typical objectives of increased margins, lower hydrogen cost and lower capital cost are also included.

During the past two decades, many articles dealing with hydrogen system management have been published. In this time methodologies for better integration, utilization and management have been considerably developed (Shariati et al., 2013). Recent approaches include research work that deals with conceptual methods of pinch analysis subspace (Liu et al., 2005) and methods for multicomponent optimization models with linear and nonlinear programming models (Jia and Zhang, 2011; Liu and Hallale, 2001). In addition, deterministic optimization methods that discuss preprocessing of minimum hydrogen consumption data and merging of data sets (Jiao et al., 2012c) have also been considered. Furthermore, the synthesis and design of hydrogen superstructure with more comprehensive discussions and determination of total annual cost, taking into account constraints of flow-rate and pressure have been addressed by Jiao et al. (2013). Pinch analysis has also been extended to include supply chain management. Folorunso et al. (2011) proposed a methodology of pinch analysis as a knowledge management module for supply chain management, as a process of integrating/utilizing suppliers, manufacturers, warehouses and retailers, so that goods are produced and delivered with the right quantities and at the right time, while minimizing costs, as well as satisfying customer requirements.

The constant reduction of sulfur in refining products, economic changes shifting towards lighter fuels, processing of heavier crude oil and the upgrading of the heavy-end sourer crude in petroleum refineries have made process engineers struggle to find optimal use of oil refinery processing units. Therefore, the hydrogen system is considered as one of the most important refinery utilities that are in charge of providing both the a sufficient amount of hydrogen to supply hydrotreating units and to make use of cracking processes for lighter and cleaner fuel production. Consequently, the conservation principles of process integration techniques have been applied to a wide set of resources, including water and hydrogen. These principles are systematically oriented and integrated to industrial process design with two broad methodologies; one is based on the graphical methods of pinch analysis and the other is by making use of mathematical optimization techniques.

4.1.1. Graphical methods of pinch analysis

In the past 30 years, pinch analysis has been considered as one of the most practical tools to emerge in the field of process integration. It is a rigorous, structured approach that may be used to tackle a wide range of improvements related to process and site utility. The graphical representation and the simplicity of the

Table 5
Cost of various measures satisfying extra hydrogen demand (Jiao et al., 2012c).

Measures	Investment cost ($\$m^{-3}\ d^{-1}$)	Operating cost ($\$m^{-3}$)	Total cost (million \$)
Increasing yield of hydrogen plant	0	0.0706	7
Expansion of hydrogen plant	7.06–35.3	0.0741	8–10
Building new hydrogen plants	35.3	0.0671	9
Constructing new hydrogen purifiers,	3.53–14.12	0.0177–0.0353	2–4
Purchasing hydrogen	0	0.0353–0.177	3–17
Retrofitting the hydrogen system	3.53	0	0.3

calculation methods provided by pinch analysis not only give a good insight to engineers at the system syntheses phase, but also assist viewing the problem physically (Liu, 2004).

Alves and Towler (2002) proposed a systematic approach that sets the hydrogen distribution system with a minimum supply for hydrogen. Their assumption in the process is based on regular operating conditions for hydrogen consuming methods so that the refinery gas can be treated as a binary mixture of methane and hydrogen. Source and sink purity profiles are built based on the assumed value of fresh hydrogen consumption that acts as utility hydrogen. Another approach based on the use of pinch analysis technology has been proposed by Zhenmin (2003) to analyse the key factors that affect hydrogen consumption and its potential saving by establishing a hydrogen cost model. However, the intentional model optimizes the design of the hydrogen system on the basis of the pinch principle using hydrogen concentration composite. It is composed of both hydrogen resource and sinks that are formed by summing up resource and sinks with different concentrations.

Another graphical method was proposed by Zhao et al. (2006) to determine the minimum hydrogen system with any concentration using sink and source composite curves. The plots of pure hydrogen load vs flow-rate were used to determine the pinch point of the hydrogen network system. The minimum utility consumption and the minimum waste of the hydrogen source were identified from the plan. Bandyopadhyay (2006) has also proposed an approach called the source composite curve with the primary goal of a single graphical representation and algebraic method to address the problem of hydrogen management. This is achieved by either changing the concentration or the flow-rate of the internal needs of the process to reduce waste production. Integration of the hydrogen distribution system with many impurities was addressed by Zhao et al. (2007) through minimizing the utility consumption of hydrogen networks in the refinery. The process is based on plotting the impurity profile according to impurity concentration vs flow-rate of hydrogen. The pinch point of hydrogen is identified by using both the impurity profile and debt plan.

The idea of extending heat integration to mass integration along with pinch technology was used by Salary et al. (2008) as a guiding principle to achieve the minimum utility target. They applied the method in the design of a refinery hydrogen system, identification of the pinch point and potential recovery across the site. Wan Alwi et al. (2009), addressed another graphical method based on the need composite curves, source and demand allocation curve and network allocation diagram to target the minimum gas flow-rate and increase the maximum nitrogen recovery by proper guides of system design. The approach is appropriate for use in multiple pinch points. In addition, the Network Allocation Diagram mentioned in the approach allows designers to have a clear view of minimum targets or minimum streams, as well as the design of the optimized system.

Notwithstanding the benefits of the aforementioned methods, El-Halwagi et al. (2003) proposed a rigorous approach to reduce the use of fresh resources. This includes segregation, mixing and direct recycle/reuse strategies by formulating the problem mathematically to overcome the drawback of iteration. An elegant design for its noniterative approach to the problem of fixed flow-rates that involves regeneration to reduce waste discharge was proposed by Agrawal and Shenoy (2006). The unified conceptual approach for targeting the minimum make-up hydrogen used for reforming units in this work argues from the view of sustaining the penalty of the hydrogen utility. The algorithm is based on choosing the nearest available neighbouring source to satisfy a sink by means of source quantity and quality as parameters to meet the target for the hydrogen system. The idea of the algorithm has evolved to account for pressure and concentration constraints forced by compressors and enhanced by the purification process.

Foo and Manan (2006), addressed a scope-extending cascade analysis tool for targeting minimum flow-rates for water and property-based networks in a gas cascade study design. The process identifies the minimum flow-rate target, resource allocation target and the pinch point area for different types of gases, i.e. nitrogen, oxygen and hydrogen. The procedural approach is performed by locating sinks and sources of gas at their respective concentration level, cascading the net flow-rate surplus/deficit on the way down to a lower concentration level to obtain the cumulative surplus/deficit flow rate and then setting up the cumulative impurity load cascade. This process claimed to enable appropriate selection of gas purifiers and handles problems that involve multiple pinches.

The source composite curve method was further emphasized by Pillai and Bandyopadhyay (2007) using an algebraic methodology that combines both the simplicity of pinch analysis and the mathematically rigorous algorithm of optimization techniques to reduce the hydrogen resource requirement. Nelson and Liu (2008) utilized a pinch analysis process in an attempt to reduce the flow-rate of fresh hydrogen make-up and the off-gas discharge, using an automated pinch spreadsheet based on Excel. The Automated Hydrogen Pinch Spreadsheet is capable of identifying the surplus of hydrogen in the system and solving many pinch problems, purifier placement and approximations of how hydrogen can be saved. A three step numerical procedure was addressed by Liu et al. (2009) for designing and targeting a resource system with many inputs of different qualities. The authors argue from the perspective of costs for different types of resources i.e. hydrogen and water in industrial plants taking into consideration reduction in the consumption of resources with higher qualities because of their higher cost.

A targeting process for a refinery hydrogen system was addressed by Liao et al. (2011a). The process in this work imposes a rigorous algorithm to tackle the infrequent, but realistic, cases of threshold problems by establishing a connection between the pinch simplifications and mathematical modelling. Assuming that the hydrogen utility is cleaner than all of the process hydrogen, the authors claimed that the simplified formulation that relates to the problem of hydrogen pinch theory with the mathematical model can lead to an optimal analytical solution by mathematical deduction of the aforementioned formulations. Lou et al. (2013) incorporated the mechanism-based purifier model into graphical methods to find the initial location of the purifier in the hydrogen network to improve the pinch sliding approach of purifier integration and to simplify the procedures for shifting composite curves.

A graphical method for targeting the pinch point and reducing the hydrogen utility consumption, as well as the waste discharge, was proposed by Zhang et al. (2011). The process incorporates the reuse of the purification process by integrating its parameters, which include the feed concentration and recovery efficiency, with the hydrogen system in order to reduce the hydrogen utility consumption. However, the graphical representation is based on the developed triangular rule: for the purification process the material balance between its feed and product is represented by a polygon plotted using the hydrogen load vs flow-rate method. An extension of the water source diagram to a hydrogen source diagram was proposed by Borges et al. (2012). The process is non-iterative algorithmic procedure, constructed of eight steps, that calculates the minimal hydrogen consumption target for the refinery hydrogen system and simultaneously synthesizes the hydrogen system flowsheet. Another method to optimize the hydrogen system with purification reuse, based on the hydrogen surplus and pinch point method, was proposed by Liu et al. (2013a,b). The process addresses the effect of purification feed-flow rate on the hydrogen utility consumption. It analyses the characteristics of the pinch point location, hydrogen utility adjustment and identification of the pinch point.

4.1.2. Mathematical optimization techniques

The conceptually based methods of pinch analysis are helpful in obtaining physical insight and allowing different process design objectives to be screened prior to the detailed design in process integration problems, through their graphical representations. However, mathematical optimization-based methods far outweigh the benefits of pinch analysis-based techniques, and are much preferred to address several issues as synthesis tools in handling complex systems with different complex constraints. Many mathematical optimization techniques have emerged to tackle the limitations of pinch analysis-based techniques in hydrogen system integration and management. These limitations include issues of different components, cost optimality, flexibility and other issues that will be reviewed in this section.

Towler et al. (1996) proposed a Linear Programming (LP) method to expand the limited publication on cost correlation of hydrogen recovery from refinery off-gas for purification process units such as PSA and membrane diffusion. They exploited the analogy of refinery hydrogen recovery to that of process heat integration in which the utility requirement of the process is minimized by maximizing heat exchange between process streams. The information provided by the assessment of process heat integration is additional to that obtained by LP optimization of refinery operations. Girardin et al. (2006) addressed a multipurpose optimization model for the design of a minimum fresh hydrogen requirement. The design uses the pinch method as a preliminary procedure for the selection of compatible purification units based on the hydrogen flow-rate and purity profile. Then, the process combines the Mixed Integer Linear Programming (MILP) model for the hydrogen system design and evolutionary algorithm to identify the optimal solution for the design of purification units. Umana et al. (2014) have studied the interactions between the refinery hydro-processors and a multicomponent hydrogen network. The work investigated the varying hydrogen inlet conditions of refinery hydrogen consuming processes, optimizing refinery operating conditions and constrained process performance indicators. The authors argue that there is a potential saving of 2% from varying hydrogen inlet conditions and also 7% reduction can be attained by optimizing the temperature.

A case study of the hydrogen distribution system in the Porto Refinery was carried out by Fonseca et al. (2008) that discusses the limitation of using graphical methods directly to design a real hydrogen system. The graphical techniques can only help to identify the system modifications, thus, the authors proposed an assumption based on the use of a hydrogen surplus plan. They assumed that because the distribution system does not have a hydrogen pinch point it is possible to reduce the flow-rate of the hydrogen supplied to the system. The findings from a hydrogen surplus diagram guided them to the LP model. The LP model includes the pressure interactions by taking into consideration the make-up compressor as source and need. The process is claimed to be acceptable for utilities reductions and also applicable for working with reduced capacity in the case of plant shutdown. The equilibrium constraints for scheduling refinery hydrogen pipeline network between hydrogen producing and consuming units is addressed by Zhou et al. (2014). In this work the flow reversal inside a pipe and the dynamic performance of the pipe is considered by incorporating a detailed pipeline model into the optimization model allowing for flow reversal. The pipe model also considers the multi-component system, the compressibility factor and the safe operation of the pipe segments.

Liu and Hallale (2001) and Hallale and Liu (2001) have used the idea of global optimization of bilinear process networks with many components proposed in Quesada and Grossmann (1995) to retrofit refinery processes. The basic idea of the optimization of a bilinear process system that involves mixers, splitters or purifiers and linear processes is to rely on a relaxed LP model. Therefore, by establishing a clear relation between the component flow and composition model for mass balance problems, a relaxed LP can be obtained through reformulation of the linearization techniques (Quesada and Grossmann, 1995). The nonlinear nature of the restrictive problems for continuous optimization of process networks such as refinery hydrogen systems is challenging. The sources of non-convexities in these problems are the material balance equations in which bilinear terms for the flow and composition are unknown.

Gutierrez et al. (2010) addressed the concept of convexity for the nonlinear programming optimization in the problem of hydrogen distribution in an oil refinery. They argue from the point of view of the local minima if the standard non-global nonlinear programming algorithm is used. Yunqiang et al. (2011) proposed two-step sequential optimization process for the refinery hydrogen system. The method divides the hydrogen system optimization problem into two sub-problems. Optimization of the feed routes of the purification system and the feed routes of the hydrogen consumption units are the two steps proposed in the work. Then, the modelling of both the purification system and the consumption units is converted into a nonlinear programming (NLP) model using the linear interactive general optimizer. In contrast to Liu and Hallale (2001), Shahraki et al. (2005) proposed an NLP approach in which pressure constraints were also considered. Nevertheless, the design is based on optimizing a reducible superstructure of hydrogen within the refinery, and it is limited to feasible piping changes where there is no consideration for the installation of new equipment.

Another method addressed by Jiao et al. (2012a) extends the work of Yunqiang et al. (2011) by proposing two mathematical techniques that include 1) two-step optimization for the purification units and the hydrogen source and sink, 2) a simultaneous optimization process to retrofit the hydrogen system. Because of, the complexity and the large computational effort in optimizing the hydrogen distribution system in the refinery by the use of a mixed integer nonlinear programming (MINLP) model, the problem is tackled based on the two-step optimization process, where it is transformed into two smaller MINLP models. In addition, a simultaneous optimization process is set up to linearize the bilinear and trilinear terms that represent the hydrogen balance in the MINLP models, which could then be avoided by using MILP linearization techniques. Saleh et al. (2012) proposed a systematic MINLP optimization approach for the retrofit design of a Tehran refinery hydrogen system. The methodology of the approach is based upon the mathematical optimization of the superstructure for the two refinery sections. By incorporating a purification unit into the system, namely PSA, the authors claimed that the results of their approach show 20% reduction of hydrogen production in the north part and 31% in the south part of the refinery.

The static scheduling model of hydrogen balance in the refinery was addressed by Jinsong and Zhaoxia (2009). The authors proposed a hybrid particle swarm optimization algorithm for the balance between hydrogen production and utilization in the refinery. However, a chaos search mechanism was used along with an algorithm as the main body in NLP form to strengthen the capability of resolving the scheduling problem, as well as the minimization of hydrogen utilization cost for a time of three months. Jiao et al. (2012b) proposed an iterative algorithm for optimal scheduling of the hydrogen system in a refinery. The process is composed of eight periods of the scheduling horizon to address the occurrence of composition discrepancies in the hydrogen piping system. The mathematical formulation of the iterative process is to achieve a minimum operating cost to avoid suboptimal operation that occurs while using MINLP for the optimization of the hydrogen system. The inconsistency in the solution quality and the computational effort in the MINLP model are overcome in another approach by Jiao et al. (2013). The design is first formulated for the design and optimization of a flexible hydrogen system under many operating scenarios. Then, it is transformed into an MILP model based on a linearization process similar to the work published by Jiao et al. (2012a).

Modelling and optimization for multiperiod operation for hydrogen network design in a refinery was addressed by Ahmad et al. (2010). The model discusses optimization of a multiperiod design of a flexible hydrogen system that considers the maximum allowable capacity of existing compressors, pressure constraints and the optimal placement of new compressors, as well as the piping system. The work accounts for changes in the operating requirements of the refinery processes and the effect of these changes on the minimum total annualized cost of the hydrogen system. The MINLP model for the design of flexible multiperiod operation of a refinery hydrogen system in this work is transformed into an MILP by the use of bilinear terms for linearization of all nonlinear terms to obtain a proper initialization of the model. A chance constrained programming method for the optimization of the hydrogen system in a refinery was addressed by Jiao et al. (2012c). The process considers uncertain conditions including uncertain hydrogen supply, uncertain hydrogen market and uncertain prices of hydrogen, electricity and fuels. By considering these stochastic properties in the formulation of the MINLP model, some information and state constraints are complied with at predefined probability levels so that the relationship between profitability and reliability of the hydrogen distribution system could be solved with different confidence levels.

Khajehpour et al. (2009) have used a heuristic superstructure to reduce the hydrogen wastage to fuel gas system within the refinery hydrogen system. The reduced superstructure solution of the MINLP problem for the hydrogen distribution system in this model is solved using the Genetic Algorithm toolbox of Matlab. The work also considers the cost of hydrogen use in four different categories, namely, production, storage, distribution and end-user costs. Other decision-making constraints, such as purification technology, are not included because they are claimed to be influential in the results of the work as well as its formulation. Comparison of four mathematical programming techniques, namely LP, NLP, MILP and MINLP, was carried out by Kumar et al. (2010). To obtain the best selected results of these diverse comparisons, two case studies were carried out on hydrogen distribution in a refinery with respect to their total operating cost and the capital cost. The NLP model occurred because of the hydrogen balance constraint around the compressor. The difference between MINLP and LP is because of the variable inlet, outlet pressure and recycle rate of the compressor. In MINLP, these variables are declared to be binary variables to determine the existence of the stream entering the compressor, whereas, in the NLP model, they are declared as continuous variables. The result shows that the MINLP model is the most promising design because it includes consideration of flow combination, pressure constraints, source and sink constraints, compressor flowrate and purity and recycles constraints.

Jia and Zhang (2011) proposed a multicomponent optimization model for refinery hydrogen networks. The model takes into account the drawbacks of previous published pinch study work on refinery hydrogen systems. The simplified assumptions in the refinery hydrogen pinch analysis methods, namely, 1) the hydrogen-rich streams are a mixture of hydrogen and methane and 2) maintaining the hydrogen partial pressure constant, are optimally not proven. By incorporating the change of vapour–liquid equilibrium because of the composition change at the downstream separation unit, this design is claimed to be effective. It considers the detailed composition change of the hydrogen distribution in the refinery, and results in a reduction of hydrogen produced from the hydrogen plant by 6.19 Million Metric Standard Cubic Feet per Day MM scfd. The total exergy consumption of the hydrogen utility and compressor work in a refinery hydrogen system were studied by Wu et al. (2012). The authors argue from the point of view of hydrogen to hydrocarbon ratio, where it is necessary to satisfy the constraint of pure hydrogen load or the hydrogen oil ratio to achieve the desired oil conversion rate in the hydrogen consuming reactors. However, the mathematical optimization model of this work is only suitable for systems whose vapour–liquid equilibrium is not sensitive to change of the hydrogen to oil ratio. Hydrogen production from the correlation of both exergy efficiency and CO_2 emission aspects by steam-methane reforming (SMR) was investigated by Chen et al. (2012). The study is modelled based on a practical flow diagram that includes desulfurizer, furnace, separation unit and heat exchangers.

A multi-objective optimization model for simultaneous minimization of the operating cost and the investment cost of equipment in a refinery hydrogen system was addressed by Jiao et al. (2011). The model is formulated as an MINLP model in which the evaluation function method is used to obtain a Pareto curve (Zitzler and Thiele, 1999; Kim and de Weck, 2005). Hence, the relationship between the operating cost and the investment cost is explored for the assortment of the best possible solution from the curve. They claimed that they managed to reduce the total annual cost by 14.5 percent. The state-space superstructure representation of mass exchange network and heat exchanger network was modified by Zhou et al. (2012) to integrate the hydrogen sulfide removal unit into hydrogen system integration. The design of this work incorporates a simplified mass exchange network into the hydrogen distribution network using a set of optimization variables for the desulfurization ratio in the MINLP model. The optimization approach investigates the trade-off between the hydrogen distribution system cost and the mass exchange system cost. The optimal synthesis of sustainable hydrogen system integration was addressed by Zhou et al. (2013). The authors extended the hydrogen system integration to the sustainable hydrogen system integration problem in which a superstructure based on MINLP methodology is used to assess both the economic and environmental aspects of the hydrogen system. These features are tackled by calculating the total annual cost for efficiency evaluation of the economic aspects and the total CO_2 emission is also used to evaluate the environmental performance of the refinery hydrogen system.

Sarabia et al. (2012) presented a data reconciliation process to deal with the incomplete and unreliable information generated in the refinery control room in order to evaluate the performance of refinery processes and make decisions about its functioning. The problems of economic optimization and the interaction of the underlying basic control systems that are associated with the management of hydrogen networks in oil refineries are also significant. The authors propose an integrated framework derived from hydrogen mass balance of the entire process to optimize the available hydrogen producers in the consumers' facilities. The work covers the issues of uncertainty that might come from measurement systems such as instrument calibration, changes in flow because of variations of the composition using supervisory architecture to reconcile the data and optimize the system. de Prada et al. (2012) extended the data reconciliation concept addressed by Sarabia et al. (2012) to tackle the problems associated with reconfiguration after a sudden decrease of hydrogen production in the hydrogen producing units and plant failures. The inherent uncertainty caused by differences in the composition of the impurities, which creates a large change in the hydrogen stream's molecular weight and, therefore, the mass flow, are also considered. The MINLP model of the work provides a coherent estimation of all the process variables involved in the hydrogen system and the hydrogen consumption in the reactor with its light gases production. A robust optimization method for refinery's hydrogen network optimization was developed by Lou et al. (2014). A framework to optimize refinery's hydrogen network under uncertainty is employed. The method's robust optimization method is argued to be effective in dealing with the design and planning of hydrogen network sunder uncertainty in comparison with the deterministic approach and the stochastic programming method.

Liu et al. (2011) developed an evolutionary design methodology for creating resource allocation with many impurities for a refinery hydrogen system. In this process, the minimum utility consumption and the corresponding hydrogen system are identified by transforming the relevant sink and source data rooted in the integration of a matching matrix according to the hydrogen purity constraints and flow rate constraints. An automated targeting method for the estimation of a fresh hydrogen target in petrochemical plants was proposed by Shariati et al. (2013). The approach investigates the benefits of both graphical and mathematical optimization techniques. The superstructure-based optimization framework that accounts for the hydrogen plant as part of the entire hydrogen system is studied. The graphical design was carried out using a Resource Conservation Cascade Diagram (RCCD) that resulted in 16.7% reduction of fresh hydrogen consumption compared with the existing fresh hydrogen consumption. A mathematical optimization model that was based on NLP resulted in 9.6% reduction in the operating cost. The disturbance resistance ability of hydrogen and water networks with minimum utility consumption was addressed by Liao et al. (2014). The approach is based on measuring the concentration fluctuation of a single sink where both a graphical design method and its corresponding algorithmic method are proposed to design hydrogen and water networks with minimum utility consumption.

5. The challenges of future hydrogen network management

Hydrogen system management has been an active area of research for the past one and a half decades. Many think that the technology has reached a mature stage recently, especially for the insights based on techniques such as pinch analysis and mathematical optimization techniques. It is worth mentioning that there are no accessible review papers in the area of oil refinery hydrogen network management. However, many new methods and approaches have been published this century revealing that new research gaps are found, and more work has been carried out to address the limitations of the “old” techniques. The key strength in modelling hydrogen networks with the consideration of purification technologies is that they endogenously integrate the mass balance equation for the hydrogen content with the operating pressure in which they are mostly addressed as multipart nonlinear optimization models. Hydrogen production from conventional and supplementary sources falls into three general practices, namely, refinery catalytic naphtha reforming, steam reforming of natural gas or naphtha and partial oxidation. However, the measures taken for energy conservation in refineries frequently result in an excess of fuel gas, which in consequence is a waste of valuable hydrogen content.

The conceptual targeting representation of sources and sinks in the conceptual approaches of hydrogen pinch analysis are essential. They provide a better understanding of the analysis the hydrogen system distribution. In contrast, the real solution to the problem of hydrogen system management lies, first, in the interactions between the refinery process units, and second, in the importance of setting the target before the design. As a result, these eventually describe the performance of the system as a whole and should satisfy the following sensitivity analysis criteria for hydrogen design and distribution systems (Alves and Towler, 2002; Salary et al., 2008; Borges et al., 2012).

1. There should be an insight into each system element for appropriate selection of sources and sinks and target identification.
2. It is important to know the minimum hydrogen supply by the system in both designing a new hydrogen distribution system and the de bottlenecking of an existing system.
3. Identifying the target or the minimum hydrogen supply should allow different objectives of process design to be screened preceding the detailed process design.
4. There should be careful extraction of source and sink data for better identification of the operational constraints of the network elements.

Hydrogen pinch has a role to play in helping target project options. However, any hydrogen management solution must be tested, not just for economic viability, but for technical robustness, refinery integration and constructability. This test could be carried out using linear and nonlinear programming techniques. The output from such techniques is the key and a simple diagram of options for refinery development over various time frames.

The work on hydrogen network management has developed considerably in recent years particularly for eliminating the emission of greenhouse gases in industrial plants as well as to enhance the refinery's product quality. Studies in the literature have described a wide range of hydrogen systems, from relatively small, short-term projects such as to initiate the minimum hydrogen use to the production from internal refinery sources, for example, refinery off-gas as an effective solution to refinery hydrogen supply (Wang and Feng, 2013). Recently, the focus of most research has been shifting from modelling snapshots of hydrogen systems to modelling their evolution across time considering multiple impurities. It is, therefore, a sign of maturity to see this process underway in hydrogen management issues, with the contributions of Sarabia et al. (2012), Velázquez et al. (2013), Zhou et al. (2013) and Wang and Feng (2013) being some of the best examples of papers addressing transition processes in the refinery hydrogen system.

In the case of mathematical optimization studies, we would like to see inter-temporal models with simulated process flow diagrams able to grow across the refinery processing unit, similar to what we would expect to happen in reality. In particular, in order to be valuable for modelling early stages of transition to hydrogen utilization and management, it would be helpful if these models could incorporate the fact that different units of the refinery are expected to consume less hydrogen, with effluent treatment of crude oil products probably being the prototypes. A rigorous treatment of uncertainty, going beyond the attempts we have seen so far in the literature, would be a very useful improvement for holistic rules of process integration and data management.

6. Conclusion and recommendations for future studies

In this paper, we have assessed the modelling of hydrogen distribution in processing plants, especially in oil refineries, from extended pinch-based approaches to mathematical optimization models. The first conclusion that we can draw is that assessing the mass balance treatment of the hydrogen system and refinery hydrogen superstructure design received significantly more attention from the research community. Second, very few papers have been written attempting to model the hydrogen system in the refinery addressing the mechanism of naphtha-reforming reactions as compared with those that address pinch analysis and mathematical optimization techniques involving treatment of mass balance by either linear or nonlinear programming models. While we appreciate the data and the computational requirements of optimizing hydrogen systems across an industrial processing plant, we also see the capability of utility models in pinch technology.

We consider the information development of simultaneous syntheses of hydrogen networks and optimization as an important research area. Few hydrogen network optimization studies test the sensitivity of their analysis to assumptions on the impurities or the light gases containment in the recycled gas stream, where hydrogen has a high concentration. This information has an important effect on benchmarking the operation of the hydrogen system and can be grouped to estimate the potential benefits of different de-bottlenecking options. Demand trajectories are often assumed to be imminent, not situated within a broader energy system change, suggesting that there is value in linking hydrogen system models with optimization models. Hydrogen system models give an insight for representation of the dynamics of crude oil transitions from distillation to reforming stages, and allow returns-to-scale and optimize the system as a transition unfolds. There will be a shortcoming in using generalized models such as pinch analysis and that can be overcome if one is ready to model a non-linear or a linearize system.

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