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Targeting Model of Inter-Plant Hydrogen Network with Purification Reuse/Recycle

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Hydrogen resource can be saved by reuse and purification reuse between refineries. Thus, it's necessary to optimize the inter-plant hydrogen network in a petrochemical complex. In a petrochemical complex, purifiers are employed to recover hydrogen components from off gas. However, purifier models such as PSA, membrane units have non-linear characteristics. Even if minimizing hydrogen utility, mathematical programming models adopted by most of the researchers belong to the non-convex and non-linear models, not guaranteeing global optimal solution. In this paper, a novel mathematic programming model for the optimization of inter-plant hydrogen network with purification unit is presented. If only the hydrogen utility consumption is minimized, the model is linear, guaranteeing global optimal solution. If the object comes to economic benefits such as pipeline cost, the objective function is nonlinear. The hydrogen utilities in refinery can be reduced by the inter-plant optimization and the number of inter-plant connections can be optimized to minimum. Case study indicates that the model is effective.

1. Introduction

Sulfur, olefin as well as aromatic contents are important quality indicators for fuels such as gasoline and diesel. Experimental studies show excess amount of these contents may cause incomplete combustion of gasoline and diesel inside automobile engine. Consequently, more harmful substances such as SO_X, NO_X and Particulate Matter (PM) 2.5 are discharged. On the other hand, hydrotreating and hydrocracking reactions are effective in regulating these harmful substances. Hydrotreaters and hydrocrackers are widely employed in modern refineries. As a result, hydrogen resource plays an important role in gasoline and diesel product quality upgrading. Statistics (Dybkjær, 2005) show when the sulfur content in gasoline and diesel decreases from 500 ppm to 50 ppm, hydrogen consumption will be greatly increased. Then the shortage of hydrogen resources or the high cost of hydrogen resources is following. Therefore, the efficient use of hydrogen resources is essential for refineries to reduce costs and improve profits.
During the past two decades, process integration techniques have been introduced to manage refinery hydrogen resources efficiently. The superstructure method to hydrogen network was first proposed by Hallale and Liu (2001), and many superstructure-based methods have been developed since then for the hydrogen reuse cases. Liu and Zhang (2004) proposed a systematic methodology to select appropriate purifiers in the hydrogen network. They considered the trade-offs between hydrogen saving, cost of compressors and capital investment to build a superstructure-model including possible purification scenarios. Liao et al. (2010) presented a systematic approach for the integration of hydrogen networks with purifiers. Later, they (Liao, 2011a) developed a rigorous targeting approach with the pinch insight combined. This method is extended to hydrogen networks with purification reuse (Liao, 2011b). In addition, Zhou et al. recognized H_2S as a key contaminant and corresponding constraint (Zhou, 2012a) and inserted desulfurization processes in hydrogen network optimization (Zhou, 2012b). Wei et al. (2017a) introduced the Worst-Case Conditional Value-at-Risk concept to investigate the disturbance resistance ability of hydrogen network. What's more, they (Wei, 2017b) extended a

single impurity to the multiple impurity case to investigate the disturbance resistance ability of the hydrogen network.

The first graphical method for the assessment of hydrogen sources was proposed by Towler et al. (1996). The effectiveness of hydrogen reuse is investigated by comparing the hydrogen recovery costs against the added value generated by hydrogen-consuming process. In recent decades, the number of articles researching purification is increasing. For hydrogen network with purifiers, Alves (1999) analysed three possible placements of the purifiers (above the pinch, across the pinch and below the pinch), and reported that placing the purifier across the pinch is the best choice. However, this qualitative conclusion cannot give quantitative hydrogen utility target. Agrawal and Shenoy (2006) addressed the purification problem based on their limiting hydrogen profile. They illustrated that the system has two degrees of freedom and calculated the hydrogen utility consumption by fixing these two freedoms. Foo and Manan (2006) evaluated two different purification processes with given feed concentration by the GCA technique. Ng et al. (2009) employed an automated targeting approach to address this kind of purification problem. Lou et al. (2013) proposed a pinch sliding approach for targeting hydrogen and water networks with different types of purifier. Later, they (Lou, 2014a) introduced the robust optimization to optimize hydrogen network with uncertainty. Furthermore, Liao and Lou et al. (2015) proposed a new concept, Mixing Potential to improve the disturbance resistance ability of the networks in the design stage. Besides, they (Lou, 2014b) presented a novel conceptual method to design multi-contaminant hydrogen networks.

The previous literatures only considered in-plant hydrogen integration. However, the hydrogen utility can be further minimized through inter-plant hydrogen optimization. Gas Cascade Analysis is proposed to find the targets for the inter-plant hydrogen network with unassisted (Chew and Foo, 2010a) and assisted integration scheme (Chew and Foo, 2010b). Deng et al. (2015) proposed a systematic approach to determine the targets of inter-plant hydrogen networks. The generalized Improved Problem Table is developed to find the flow rate targets of individual and inter-plant hydrogen networks. Jeong and Han (2011) determined the minimum flow rate of hydrogen utility and designed the optimal hydrogen network for Yeosu Petrochemical complex in Korea via mathematical programming. Deng et al. (2017) presented the superstructure for optimal design of inter-plant hydrogen network with purification reuse/recycle. Inter-plant hydrogen integration has been applied in the industrial field. Three petrochemical plants exist in Nanjing Chemical Industrial Park of China, namely Nanjing Chemical Industry Company, Sinopec Yangzi Petrochemical and Sinopec Jinling Petrochemical, which all have hydrogen-producing and hydrogen-consuming units.

In this paper, a new transhipment model that combines that insights from hydrogen pinch with mathematical programming is developed to optimize the inter-plant hydrogen problem with purification reuse/recycle. The mathematical model is linear to guarantee global optimal solution. What's more, the model can also optimize the number of the inter-plant hydrogen network. To illustrate the application of the proposed model, a case study is presented before the conclusion of the work.

2. Problem statement

The problem for targeting the interplant hydrogen network can be stated as follows. Given a set of refineries in the petrochemical industrial park, there is a set of hydrogen consuming processes in each plant, and their outlet streams are treated as a set of process hydrogen sources while their inlets treated as a set of process hydrogen sinks. Each process hydrogen source is specified by its outlet flowrate and outlet hydrogen purity. Each process hydrogen sink has inlet flowrate and the lower bound of inlet hydrogen purity. Process hydrogen sources can be reused/recycled to fulfil the requirements of process hydrogen sinks. A set of external hydrogen sources or hydrogen utilities are needed for supplementary. The internal hydrogen sources should be recycle/reused as much as possible to reduce the consumption of hydrogen. The surplus process hydrogen sources are discharged to fuel system. Besides, every refinery has purifiers to recover the process hydrogen sources with the hydrogen purity. The recovery ratio of the purifier and the purification product purity are fixed. This paper aims to target inter-plant hydrogen network with purification reuse/recycle.

3. Mathematical model

As shown in Figure 1, the whole system can be divided into several concentration levels which correspond to the concentration of fresh hydrogen, purification product, purification tale and process sources/sinks. Following an analogy to the HEN problem (Papoulias and Grossmann, 1983), a transhipment model is developed in the form of considering hydrogen flow as a commodity. The hydrogen flow is shipped from process sources, purification product, purification tale and hydrogen utility to process demands, purification feed and fuel system mains through concentration levels. Figure 1 illustrates that hydrogen flows from process sources, purification product, purification tale and hydrogen utilities to the corresponding concentration level and then to process demands, purification feed and fuel system mains in the same concentration level with the remainder going to adjacent lower and higher concentration levels. The lowest concentration level is denoted by k=1, while the highest level is denoted by k=K.

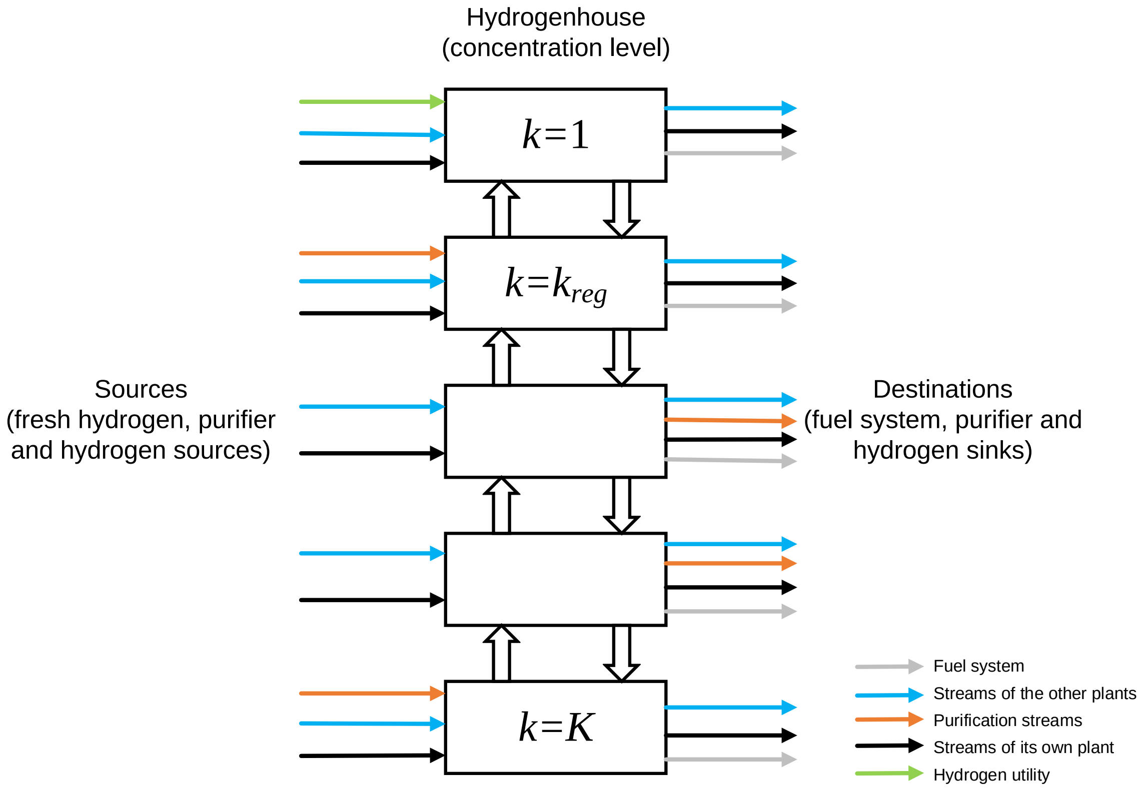

Figure 1: Transhipment model for inter-plant hydrogen minimization problem.

To simplify the model, the residual flows between level k and level k+1 can be regarded as one simple flow r_k whose flow rate is the residual flow from level k to level k+1 minus the residual flow from the opposite direction. As contaminated hydrogen streams are composed of hydrogen and impurity, the hydrogen concentration of level k can be expressed as:

$$Q_k = r_{k-1}(c_{k-1} - c_k) \qquad (1)$$

As detected from the pinch insight, the amount of hydrogen should meet the sinks of each concentration level. Thus, the cumulative surplus of hydrogen at each concentration level k should be nonnegative:

$$\sum_{k=1}^{k=m} Q_k \geq 0 \qquad \forall m = 2,3,4,\ldots\ldots,K \qquad (2)$$

By performing a total mass balance and hydrogen surplus constraint at each concentration level, the transhipment model for minimum fresh hydrogen cost of inter-plant hydrogen network is given by

$$\min \sum_{o\in O} \sum_{u\in U} F_u^o \qquad (3)$$

s.t.

$$\sum_{i\in I_k} F_i^o + \sum_{u\in U_k} F_u^o + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o + r_{k-1} = \sum_{j\in J_k} F_j^o + w_k + r_k + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o + \sum_{i\in i_k} F_{i,p}^o$$

$$\forall k = 2,3\ldots\ldots K-1, k \neq N, \forall o \in O \qquad (4)$$

$$\sum_{i\in I_k} F_i^o + \sum_{u\in U_k} F_u^o + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o + F_{reg}^o + r_{k-1} = \sum_{j\in J_k} F_j^o + w_k + r_k + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o \qquad k = N, \forall o \in O \qquad (5)$$

$$\sum_{i\in I_k} F_i^o + \sum_{u\in U_k} F_u^o + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o + F_r^o + r_{k-1} = \sum_{j\in J_k} F_j^o + w_k + r_k + \sum_{\substack{o'\in O\\ o'\neq o}} F_{o',k}^o \qquad k = K, \forall o \in O \qquad (6)$$

$$\sum_{i\in i_k} F_{i,p}^{o} = F_{reg}^{o} + F_{r}^{o} \qquad \forall o \in O \qquad (7)$$

$$R\sum_{i\in i_k} F_{i,p}^{o}\, c_i^{o} = F_{reg}^{o} c_{reg} \qquad \forall o \in O \qquad (8)$$

$$r_K = 0 \qquad (9)$$

$$F_u^{o}, F_{o',k}^{o}, w_k, F_{reg}^{o}, F_r^{o}, F_{i,p}^{o} \geq 0 \qquad \forall k = 1,2,3,\ldots\ldots, K, u \in U, o, o' \in O, i \in I \qquad (10)$$

where F_u^o is the hydrogen utility flowrate of *o*th refinery, w_k is the flow rate of fuel system out of level k. Equation (4) shows mass balance of a common concentration level. For the level *k*, the inlet streams include the process hydrogen sources itself, fresh hydrogen, the residual flow from level k-1 and process hydrogen sources from the other refineries, and the outlet streams include the process hydrogen sinks, waste hydrogen, the residual flow to level k+1, the purifier feed and the streams to the other refineries. Inlet and outlet flows should be equal. Equation (5) and (6) show mass balance of purifier product concentration level and tale gas concentration level. Equation (7) and (8) are mass balance for the purifier. Otherwise, Equation (9) indicates level K is the highest level.
The above equation is an LP problem, which can be easily solved to target the minimum hydrogen utility. Moreover, the location of a pinch point is indicated when the cumulative hydrogen surplus approaches zero, namely,

$$\sum_1^k Q_k = 0 \qquad (11)$$

On the other hand, the number of inter-plant connections is an important parameter for the demonstration of total network complexity. The binary variables $X_{o',k}^{o}$ are introduced to determine whether inter-plant streams exist. When $X_{o',k}^{o}$ equals to 1, it means that the inter-plant stream exists. When $X_{o',k}^{o}$ equals to 0, it means that the inter-plant stream does not exist.

$$X_{o',k}^{o} = \begin{cases} 1, & F_{o',k}^{o} > 0 \\ 0, & F_{o',k}^{o} = 0 \end{cases} \qquad \forall o, o' \in O, k = 1,2,3,\ldots\ldots, K \qquad (12)$$

The number of inter-plant connections (NC) can be determined by Eq. (13).

$$NC = \sum_{o\in O}\sum_{o'\in O}\sum_1^K X_{o',k}^{o} \qquad (13)$$

In addition, the minimum number of inter-plant connections can be determined by setting their upper bounds. The upper bounds can be increased one by one until the optimal flow rate of hydrogen utility does not tend to reduce. When optimizing hydrogen network, the number of inter-plant connections can be reduced to be the minimum.

4. Case study

This section adopts a case with two hydrogen networks from literature to prove feasibility and correctness of the MILP model. The data for plant A (Elkamel,2011), plant B (Alves, 2002) is extracted as shown in Table 1**.** It's assumed that the two plants exist in petrochemical industrial park. All the units for flow rate are converted to be Nm^3/h.
The mathematical model is applied to optimize the inter-plant hydrogen network with purification unit. In this case, the hydrogen purity for the product stream of PSA is given as 90% and the upper bound of its feed flow rate is set as 40,000 Nm3/h. The hydrogen recovery ratio of PSA is considered as a parameter, which is defined as 0.9.
The flow rate of hydrogen utility for plant A is optimized as 70,031 Nm^3/h. The optimal flow rate of the product stream of PSA is found as 20,516 Nm^3/h and the flow rate of its feed is optimized as 28,199 Nm^3/h. The optimal feed purity of PSA is found as 0.7275, which is less than the pinch purity (0.75) reported in the literature. Similarly, the flow rate of hydrogen utility for plant B is optimized to be 16,294 Nm^3/h. The product flow rate of the product of PSA is optimized as 6,730 Nm^3/h and the optimal feed flow rate is found to be 9,614 Nm^3/h. The optimal feed purity of PSA is determined as 0.7, which is equal to pinch purity of hydrogen network.
Similarly, the mathematical model be applied for the optimization of inter-plant A-B. The flow rate of hydrogen utility of plant A and plant B is optimized as 85,875 Nm^3/h. the optimal flow rate of the product stream of PSA is found as 22,966 Nm^3/h. Besides, the feed flow rate of PSA is found as 33,083 Nm^3/h and the optimal feed purity is found as 0.6942. What's more, only one inter-plant stream is allocated from plant A to plant B (31,162 Nm^3/h, 0.8). Based on the existence of this inter-plant stream, the total utility of plant A and Plant B is slightly decreased from 86,325 Nm^3/h (the sum of individual networks optimization) to 85,875 Nm^3/h (inter-plant network optimization).

For individual plant, the optimal flow rates of external hydrogen sources (or hydrogen utilities) for Plant A, B are determined as 70,031 Nm3/h, 16,294 Nm3/h listed in Table 2, which are less than those reported in the literature (Elkamel, 2011; Alves, 2002). Otherwise, the inter-plant hydrogen network is optimized which is not covered in the literature. At the same time, the number of inter-plant connections is also optimized to the minimum.

Table 1: hydrogen source and sink data for two plants (plant A and B)

Hydrogen network	Hydrogen sources	Purity (mole fraction)	Flowrate	Hydrogen sinks	Purity (mole fraction)	Flowrate
Plant A	CRU	0.8	17,303	HCU	0.8671	93,306
	HCU	0.8	60,678	GOHT	0.8358	82,656
	GOHT	0.75	55,281	RHT	0.8257	39,164
	RHT	0.75	25,870	DHT	0.7487	12,472
	DHT	0.7	8,004	NHT	0.7265	5,726
	NHT	0.65	3,840			
	HP-A	0.95	89,304(current)			
Plant B	SRU	0.93	50,303	HCU	0.8061	201,197
	CRU	0.8	33,530	NHT	0.7885	14,531
	HCU	0.75	145,305	DHT	0.7757	44,707
	NHT	0.75	11,177	CNHT	0.7514	58,117
	DHT	0.73	27,942			
	CNHT	0.7	36,885			
	HP-B	0.95	22,353(current)			

5. Conclusions

The paper adopts a mathematical model to target inter-plant hydrogen network with purification reuse/recycle. The model contains fresh hydrogen, process hydrogen sources, process hydrogen sinks, fuel system purifiers and all the possible connections between them. In case, two plants with purification reuse/recycle is optimized. It's obvious that the hydrogen utility for inter-plant scenario can be reduced further with the existence of the inter-plant streams.

Nomenclature

I	set of hydrogen sources
J	set of hydrogen sinks
O	set of refineries
U	set of hydrogen utilities
F	flowrate
c	concentration of hydrogen
r	hydrogen recovery ratio
K	the highest concentration level
r	the residual flows
Q	the cumulative surplus of hydrogen
w	waste hydrogen

Subscript

i	hydrogen source
j	hydrogen sink
p	purifier feed
k	hydrogen stream concentration level
reg	purifier product
r	purifier tale gas
u	hydrogen utility
o'	hydrogen streams from the o'th refinery

Superscripts

o	*hydrogen streams of the oth refinery*

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