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Relative Concentration based Mathematical Optimization for Improving Fresh Hydrogen Utilization Efficiency of Multi-Impurity Hydrogen Networks

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The traditional methods of hydrogen network integration generally minimize hydrogen consumption and rarely discuss the characterization of hydrogen utilization efficiency (HUE). And some definition of HUE are also not accurate enough. Based on relative concentration property, this paper constructs a nonlinear mathematical model encompassing all necessary constraints to synthesize the multi-impurity hydrogen networks. Result shows that this model is superior to all ones with absolute concentration basis. Through observation and analysis of hydrogen to oil ratio (HTO), a practical and important operation parameter of refinery, we can know the actual hydrogen demand of one hydrogen network. So, in this paper we compare two different kinds of HUE. Conclusion is: Efficiency$_2$, calculated by actual hydrogen demand, is more reasonable than Efficiency$_1$, calculated by hydrogen discharge.

1. Introduction

Nowadays refinery hydrogen consumption increases by about 8% a year. Therefore, hydrogen resources saving are of great significance. Alves and Towler (2002) used the diagram of surplus hydrogen to target minimum fresh hydrogen consumption. But this method computation efficiency is very low. El-Halwagi et al. (2003) put forward a graphical method with contaminant versus flow rate diagram. Then, Foo et al. (2006) put forward algebraic calculation instead of visual solution and ultimately achieved the purpose of maximum resource recovery. Zhang et al. (2011) put forward an improved graphical method for the hydrogen networks with purification. Liu et al. (2013) further performed the research on the relationship between the Pinch Point location and the inlet, outlet concentration of purification equipment. As for mathematical programming methods, they usually build superstructure and a corresponding mathematical model. Hallale and Liu (2001) first established a superstructure model to optimize the hydrogen network. Zhou et al. (2012) added a desulfurization device. Gradually, other factors such as uncertainty (Zuwei et al. 2010), multi-objective (Jiao et al. 2013), multi-period (Liang et al. 2016), etc. were considered. With reference to water networks, Zhang et al. (2016) established relative concentration constraints and further reduced hydrogen consumption.

In this paper, a multi-impurity, relative concentration basis, nonlinear programming mathematical model is established to analysis two different characterizations of HUE. According to the actual operation parameter, *HTO*, the best one is selected to explain and analyze the ability and potential of hydrogen resources saving.

2. Theory

2.1 Superstructure

Superstructure showed in Figure 1 includes hydrogen sources (*sr*), sinks (*sk*), and purifier *psa* (Pressure Swing Adsorption). {1,2,...i...a} is the number of sources and {1,2...j...b} is the number of sinks. Each source can supply hydrogen to any sink and each sink can also receive hydrogen from any source. Besides, *sr* could part or all be sent to one or more *sk*, or similarly to *psa* or fuel gas.

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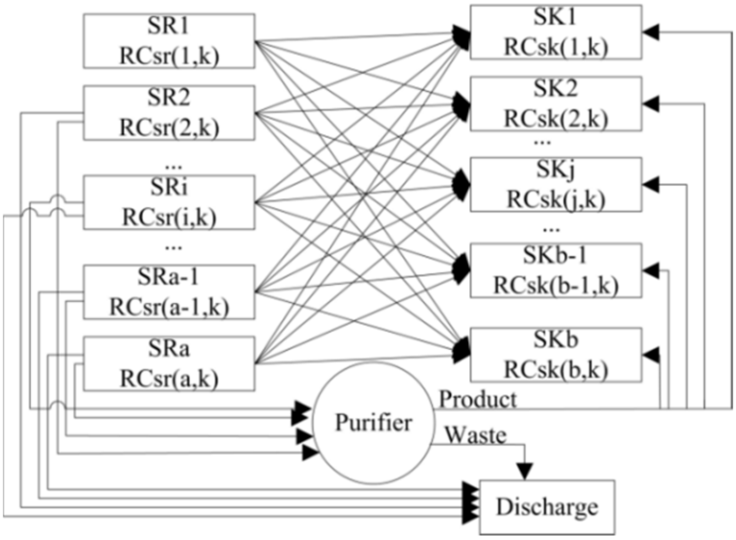

Figure 1: A superstructure of hydrogen network with purification

The hydrogen demand of sk_j can be satisfied by one or more sources and/or *psa*. This paper builds mathematical model with this superstructure, and the target is to minimize fresh hydrogen consumption and maximize HUE.

2.2 Mathematical model

2.2.1. Calculation of relative concentration

The main reference of relative concentration (*RC*) in this paper is research of Zhang et al. (2016).

$$RCsr_{i,k} = \frac{m_{i,k}}{ysr_i} \tag{1}$$

$$RCsk_{j,k} = \frac{n_{j,k}}{ysk_j} \tag{2}$$

$m_{i,k}$ and $n_{j,k}$ are absolute concentration (*AC*) of contaminant *k* in sr_i, and the ceiling *AC* of *k* in sk_j. $RCsr_{i,k}$, $RCsk_{j,k}$ are corresponding *RC*. Compared with *AC*, *RC* is used to quantify the impurity concentration and the hydrogen flow rate is employed to quantify flow rate.

It releases the total flow rate and concentration normalization constraints. In other words, it is relaxation, so it is superior to absolute basis.

2.2.2. Constraint equations of *sr*

The main constraint for every source is availability of hydrogen resource.

$$Hsr_i = Fsr_i \cdot ysr_i \tag{3}$$

$$Z_i \cdot s_i + \sum_{j=1}^{b} Hy_{j,i} \cdot d_{i,j} \leq Hsr_i \tag{4}$$

The sum of hydrogen flow rate Z_i (from sr_i to *psa*), $Hy_{j,i}$ (from sr_i to sk_j) should be less than total hydrogen flow rate of sr_i (Hsr_i). In Eq(4), s_i is the connection relationship between sr_i and *psa*; $d_{i,j}$ is matrix of network structure.

2.2.3. Constraint equations of *sk*

For every sink, there are two constraints: first is amount of hydrogen provided by *sr* and *psa* must meet the demand of sk_j; Second is the impurity concentration should be less than the upper limit of sk_j.

$$X_j = Xsk_j \cdot (1+e_j) \tag{5}$$

Xsk_j is the current oil processing capacity, when there is a fluctuation e_j in sk_j, it turns to X_j.

$$Hsk_j = l_j \cdot X_j + c_j \tag{6}$$

Hsk_j is the actual hydrogen demand of sk_j, and l_j, c_j are known correction coefficients.

$$O_j \bullet ss_j \bullet RCpsa_k + \sum_{i=1}^{a} Hy_{j,i} \bullet d_{i,j} \bullet RCsr_{i,k} \leq RCsk_{j,k} \bullet (O_j \bullet ss_j + \sum_{i=1}^{a} Hy_{j,i} \bullet d_{i,j}) \tag{7}$$

$$O_j \bullet ss_j + \sum_{i=1}^{a} Hy_{j,i} \bullet d_{i,j} \geq Hsk_j \tag{8}$$

O_j is hydrogen flow rate from *psa* to sk_j, ss_j is the connection relationship. $RCpsa_k$ is *RC* of contaminant *k* in *psa* outlet. Eq(7) is impurity constraint of sk_j. Eq(8) means the constraint of hydrogen demand of sk_j.

2.2.4. Constraint equations of psa

Constraint for *psa* is about mathematical relationship with its inlet and outlet.

$$\sum_{i=2}^{a} Z_i \bullet s_i \bullet psa_{\lambda 1} = \sum_{j=1}^{b} O_j \bullet ss_j \tag{9}$$

$$\sum_{i=2}^{a} Z_i \bullet s_i \bullet RCsr_{i,k} \bullet psa_{\lambda 2} = (\sum_{j=1}^{b} O_j \bullet ss_j) \bullet RCpsa_k \tag{10}$$

Eq(9) is *psa* hydrogen flow rate relationship and Eq(10) is impurity relationship both between *psa* inlet and outlet, where $psa_{\lambda 1}$, $psa_{\lambda 2}$ are constants of *psa* hydrogen recovery rate, impurities removal rate.

2.2.5. Objective function

In this paper, objective function is minimum hydrogen consumption: *FSRH*.

$$FSRH = \sum_{j=1}^{b} \frac{Hy_{j,1} \bullet d_{1,j}}{ysr_1} \tag{11}$$

And at the same time, we use following variables to do some useful analysis:

$$HTO_j = \frac{O_j \bullet ss_j + \sum_{i=1}^{a} Hy_{j,i} \bullet d_{i,j}}{X_j} \tag{12}$$

HTO_j is current hydrogen to oil ratio of sk_j.

$$efuel = \frac{\sum_{i=2}^{a} Z_i \bullet s_i - \sum_{j=1}^{b} O_j \bullet ss_j + (\sum_{i=2}^{a} Hsr_i - (Z_i \bullet s_i + \sum_{j=1}^{b} Hy_{j,i} \bullet d_{i,j}))}{FSRH \bullet ysr_1} \tag{13}$$

$$Efficiency_1 = 1 - efuel \tag{14}$$

efuel is fuel gas to fresh hydrogen consumption ratio. $Efficiency_1$ is one HUE.

$$Efficiency_2 = \frac{\sum_{j=1}^{b} Hsk_j - \sum_{i=2}^{a} Hsr_i}{FSRH \bullet ysr_1} \tag{15}$$

$Efficiency_2$ is HUE calculated by the actual hydrogen demand.

The relative concentration model consists of Eq(1) ~ Eq(15), which is a nonlinear programming (NLP) model.

3. Case studies

Case in this paper is based on actual data of a refinery. Table 1 shows the detail data of sources and sinks.

Table 1: Data of sr&sk in multi-impurity hydrogen network

	F (Nm^3/h)	y_{sr} (mol%)	$m_{j,i}$ (mol%)				Xsk_j (t/h)	y_{sk} (mol%)	$n_{j,i}$ (mol%)		
			H_2S	N	C				H_2S	N	C
Hydrogen sources						Hydrogen Sinks					
sr_1		0.995	0.0009	0.003	0.0011						
sr_2	16000	0.915	0.007	0.016	0.062	sk_1	35	0.92	0.032	0.017	0.031
sr_3	90000	0.899	0.0216	0.0634	0.016	sk_2	130	0.91	0.012	0.0554	0.024
sr_4	32000	0.88	0.037	0.041	0.042	sk_3	65	0.898	0.021	0.023	0.058
sr_5	7500	0.9	0.056	0.022	0.022	sk_4	22	0.918	0.032	0.021	0.029
sr_6	6500	0.87	0.026	0.083	0.021	sk_5	18	0.883	0.022	0.065	0.03
sr_7	24600	0.88	0.037	0.041	0.042	sk_6	50	0.898	0.021	0.023	0.058
sr_8	8864	0.87	0.026	0.083	0.021	sk_7	30	0.918	0.032	0.021	0.029
sr_9	14000	0.44	0.123	0.166	0.271						

When fluctuations in sk_2, sk_3, sk_5, use 2.2 model we get Table 2. Results of *AC* is got by traditional methods. There are 9 sources and sr_1 is fresh hydrogen, as well as 7 sinks. And it is a multi-impurity hydrogen network. Figure 2 (a), Figure 2 (b) and Figure 3 (a) are constructed by the results in Table 2, where the X axis is different X_j, Y axis is hydrogen utilization efficiency. In Figure 2, (a) is about $Efficiency_1$, while (b) is about $Efficiency_2$.

Table 2: Results by relative concentration basis and absolute concentration basis

	e_2, e_3, e_5	-0.2	-0.16	-0.12	-0.08	-0.04	0	0.04	0.08	0.12	0.16	0.2
RC	*FSRH* (Nm^3/h)	52,993	55253.	57513	59,773	62,033	64,293	66,553	68,813	71,285	73,364	76,783
	$Efficiency_1$	0.326	0.407	0.481	0.551	0.616	0.676	0.733	0.786	0.833	0.882	0.913
	$Efficiency_2$	0.326	0.407	0.481	0.551	0.616	0.676	0.733	0.786	0.833	0.882	0.913
AC	*FSRH* (Nm^3/h)	55,650	58044	60437	62,830	65,425	68,631	71,831	76,471	80,359	82,359	85,383
	$Efficiency_1$	0.396	0.4713	0.541	0.606	0.664	0.714	0.759	0.786	0.818	0.865	0.901
	$Efficiency_2$	0.3111	0.3881	0.459	0.526	0.585	0.636	0.681	0.710	0.743	0.790	0.825

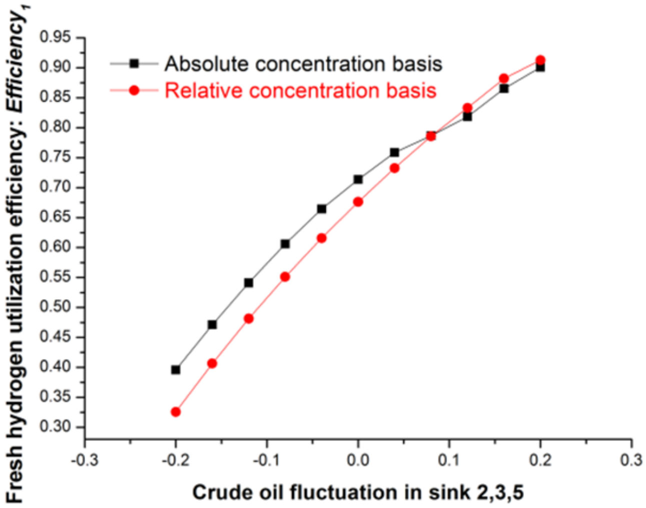

Figure 2: (a) Efficiency1 based on different X_j,

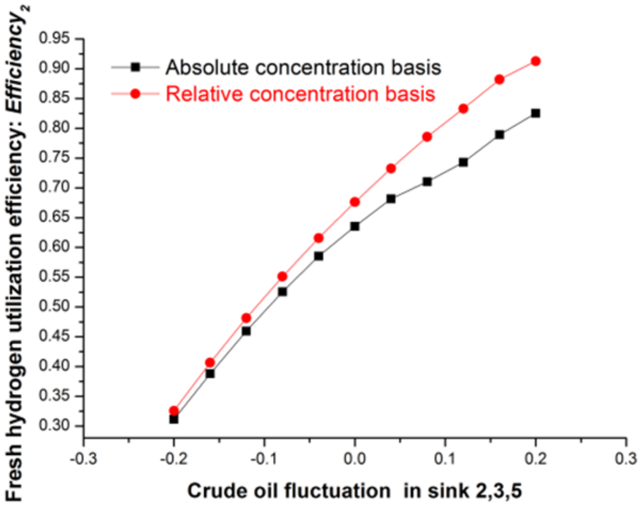

Figure 2: (b) Efficiency2 based on different X_j

Figure 3 (a) is HUE data with the same *FSRH*. The main reason of why *Efficiency*$_1$ is so close to *Efficiency*$_2$ based *RC* basis is, *RC* basis has almost reached the limit of hydrogen savings potential.

With the data of *HTO*$_j$, Figure 3 (b) shows the relationship among current, optimized and minimum *HTO*s in blue triangle, red round and black square curves in *sk*$_3$.

From Table 2 we can see that compared with absolute concentration basis, the *RC* model consumes less fresh hydrogen and it can increase HUE (*Efficiency*$_2$) by about 5%. So, *RC* constraint is more economical. Also in Figure 3 (a), we can see that with the same *FSRH*, efficiency of *RC* model is the highest. However, *Efficiency*$_1$ is higher than *Efficiency*$_2$ based on the same *AC* constraints. And Figure 2 (a) shows that for *Efficiency*$_1$, *RC* basis is smaller than *AC* basis. Consequently, *Efficiency*$_2$ is more logical.

Figure 3 (b) explains the origin of fresh hydrogen conservation, that is lowering *HTO* within feasible region. Specifically, for the same sink, the *HTO* curve based on *RC* is below absolute concentration basis while above the lower limit. This means that absolute concentration constraint always provides more hydrogen than that of *RC* constraint. *HTO* can make us easily know that how much the fresh hydrogen savings potential is.

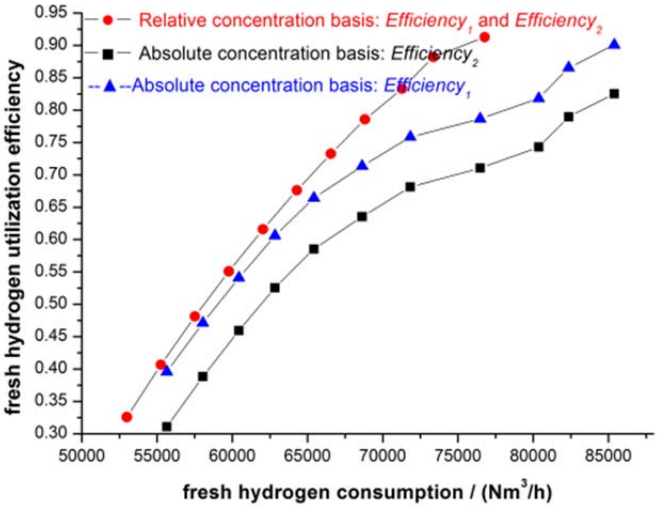

Figure 3: (a) Different efficiency based on the same FSRH

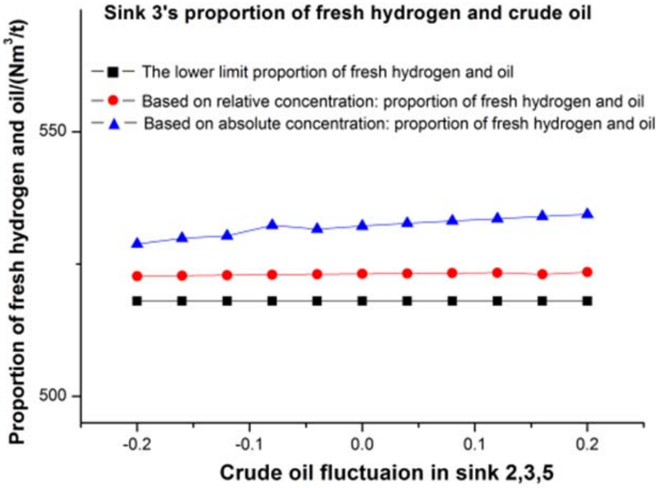

Figure 3: (b) The HTO data of sink 3

4. Conclusions

Through the analysis of the results we summarize that the NLP model proposed in this paper is superior to all absolute concentration based methods; Efficiency_2 calculated by the actual hydrogen demand is more reasonable; *HTO* is the primary reason to both amount and efficiency of fresh hydrogen consumption. Besides, the gap between current *HTO* and its lower bound represents the hydrogen saving potential of synthesis methods of hydrogen networks.

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