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Predicting Target Values of Hydrogen Networks with Purification Unit

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Hydrogen utility consumption can be reduced further when purification unit is introduced in a hydrogen network. However, targeting and design of the networks involving purification is more difficult than that of the ones involving reuse only. In this paper, when the purification pinch of a hydrogen network involving purification does not change, the target values at a certain purified concentration can be predicted easily and accurately based on the known target values at original purified concentration. The prediction is achieved by using a few relationships developed from the characteristics of hydrogen networks involving purification, and the balances of flow rate and impurity mass load below the purification pinch. The method proposed can simplify the targeting of hydrogen networks with purification unit.

1. Introduction

With crude oil becoming heavy-oriented and environmental regulation being more stringent, the demand for hydrogen increases sharply in refineries. Hydrogen network integration can increase the utilization efficiency of hydrogen resource. When purification units are introduced into a hydrogen network, hydrogen utility consumption and waste hydrogen emission can be reduced further.

Generally speaking, the methods of hydrogen network integration can be classified into Pinch methods and mathematical programming methods. Pinch method is a graphical-based one, with which hydrogen network targets can be determined by identifying pinch point. The point that corresponds to the minimum hydrogen utility consumption in composite curves is the pinch point (Foo and Manan, 2006). With mathematical programming methods, hydrogen utility consumption and network structure can be obtained simultaneously (Hallale and Liu, 2001). Whether with Pinch methods or mathematical programming methods, targeting and design of the hydrogen networks involving purification is much more complex.

For the hydrogen networks involving purification, the purification cost mainly depends on the purified concentration and the purification flow rate. Therefore, it is important to investigate the influence of the purified concentration on the purification target values. Currently, few researches include this subject.

For water networks of single contaminant involving regeneration, Xu et al. (2013) developed a few relationships between the regenerated concentration and regeneration pinch based on the balances of mass load and flow rate below the regeneration pinch. The regenerated stream targets can be predicted easily by the relationships developed. Similar to the work of Xu et al. (2013), this paper will develop a few relationships to predict the consumption of hydrogen utility and the flow rate of purified stream when the purification pinch does not change. This can simplify the targeting of hydrogen networks involving purification. A literature case is investigated to show the effectiveness of the method proposed.

2. Model of hydrogen purification unit

The common hydrogen recovery techniques include pressure swing adsorption (PSA), membrane separation, and cryogenic separation. Liu and Zhang (2004) presented a simplified model of purification device, as shown in Figure 1. In the model, impurity concentration and flow rate of hydrogen stream are involved, with other

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parameters such as temperature and pressure neglected. The balances of flow rate and impurity mass load of purification unit are shown in Eq (1) and Eq (2). The recovery ratio of hydrogen (R) is defined as Eq (3).

$$F_{in}=F_{reg}+F_{res} \tag{1}$$

$$F_{in}C_{in}=F_{reg}C_{reg}+F_{res}C_{res} \tag{2}$$

$$R=\frac{F_{reg}(1-C_{reg})}{F_{in}(1-C_{in})} \tag{3}$$

where C_{in}, C_{reg} and C_{res} are the concentrations of feed stream, purified stream and residual stream; F_{in}, F_{reg} and F_{res} are the flow rates of feed stream, purified stream and residual stream.

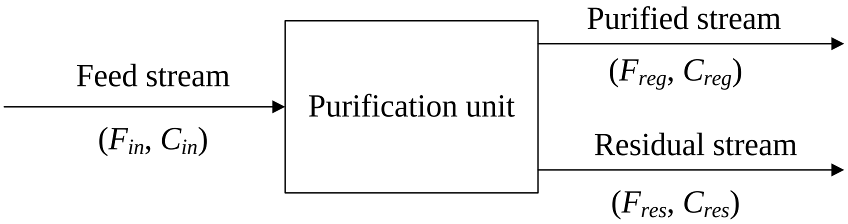

Figure 1.Simplified model of hydrogen purification unit

There is a main difference between hydrogen purification units and water regeneration units. For the regeneration unit in water network, the flow rate of feed stream is usually considered to be equal to that of outlet stream. However, for the purification unit in hydrogen network, the flow rate of residual stream should not be neglected, and the concentration change of residual stream will affect the hydrogen content of purified stream. The hydrogen purification units can be operated in the model of either fixed purified concentration or fixed hydrogen recovery ratio. The main purpose of this paper is to predict the target values of hydrogen networks with fixed purified concentration model.

3. Predicting the target values of hydrogen networks with purification unit

For water networks, Hallale (2002) pointed out that the pinch point can "sharply" divide the demands but cannot "sharply" divide the sources. The same is true for the hydrogen networks involving purification. Figure 2 shows the source-sink allocation of a hydrogen network with purification unit (Liu et al., 2016). If for the sinks just above a point, the purified streams are not used any more, the point is purification point and the source corresponding to the point is pinch-source stream. In Figure 2, S_p is the purification pinch source, D^B is the set of the sinks below the purification pinch, and D^A is the set of the sinks above the purification pinch. The cumulative flow rate and impurity mass load of the sinks below the purification pinch is equal to that of the sources which satisfy the sinks, as shown in Eq (4) and Eq (5):

$$F_{S0}+F_{reg}+\sum_{i=1}^{p-1}F_{Si,Dj}^{B}+F_{Sp}^{B}=\sum_{j}F_{Dj}^{B} \tag{4}$$

$$F_{S0}C_{S0}+F_{reg}C_{reg}+\sum_{i=1}^{p-1}F_{Si,Dj}^{B}C_{Si}+F_{Sp}^{B}C_{Sp}=\sum_{j}F_{Dj}^{B}C_{Dj} \tag{5}$$

where F_{S0}, F_{reg}, and F_{Sp}^{B} are the flow rates of hydrogen utility, purified stream, and purification pinch source allocated to the sinks below purification pinch; $\sum_{i=1}^{p-1}F_{Si,Dj}^{B}$ is the flow rates of the sources allocated to the sinks below purification pinch and $\sum_j F_{Dj}^B$ is the flow rates of sinks below purification pinch; C_{S0}, C_{reg}, C_{Si}, C_{Sp} and C_{Dj} are the impurity concentrations of hydrogen utility, purified stream, source *S_i*, pinch source, and sink *D_j*.

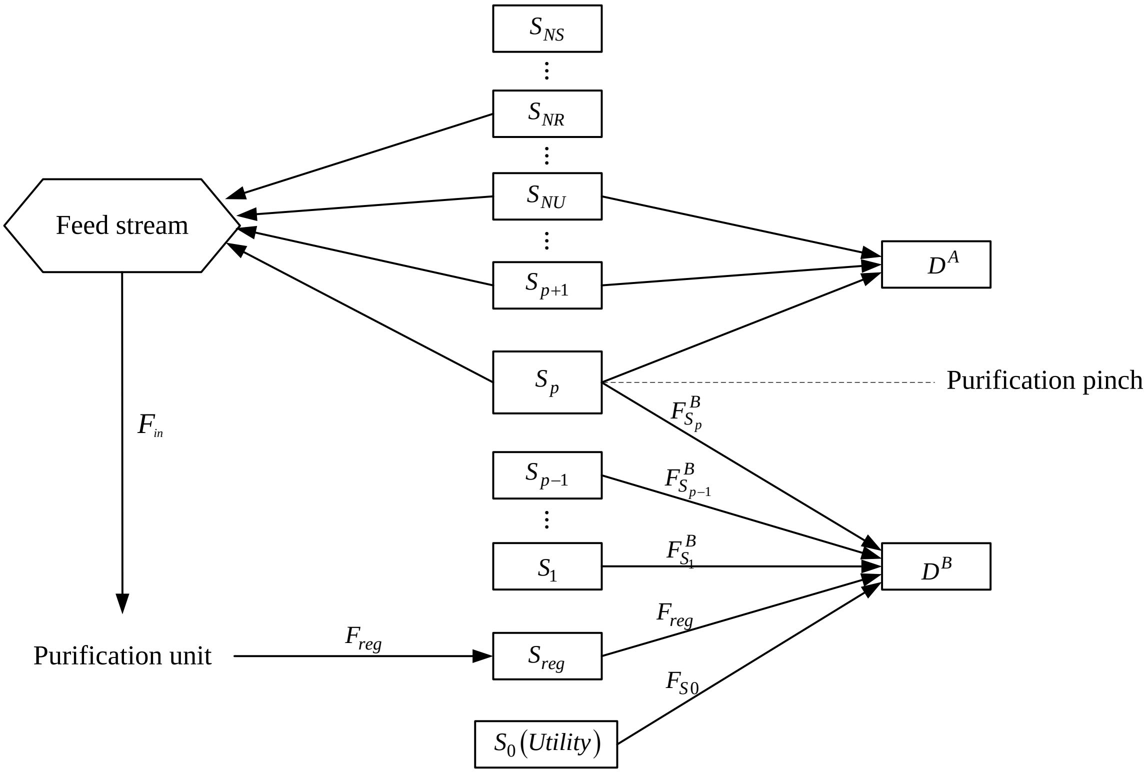

Figure 2.Source-sink allocation of hydrogen networks with purification unit

When the purification pinch does not change, from Eq (4) and Eq (5), we have:

$$F_{S0,0} + F_{reg,0} + F_{Sp,0}^B = F_{S0,1} + F_{reg,1} + F_{Sp,1}^B \tag{6}$$

$$F_{S0,0}C_{S0,0} + F_{reg,0}C_{reg,0} + F_{Sp,0}^B C_{Sp,0}^B = F_{S0,1}C_{S0,1} + F_{reg,1}C_{reg,1} + F_{Sp,1}^B C_{Sp,1}^B \tag{7}$$

where subscript “0” represents the given case and “1” the new case. When the purified concentration changes from $C_{reg,0}$ to $C_{reg,1}$, the parameters F_{S0}, F_{reg}, and F_{Sp}^B will change correspondingly. Thus, the purification targets cannot be predicted only by using Eq (6) and Eq (7).

The following information can also be obtained from Figure 2. For the whole source streams, one part is allocated to the sinks (including the sinks below and above purification pinch), another part is fed to purification unit, and the remaining with higher impurity concentration is exhausted as off-gas to fuel system. On the other hand, the demand streams are satisfied by hydrogen utility, purified stream and some sources. The balances of flow rates discussed above are:

$$\sum_{i=1}^{NS} F_{Si} = \sum F_{Si,Dj} + F_{in} + \sum_{i=NR+1}^{NS} F_{Si} \tag{8}$$

$$\sum_{j=1}^{ND} F_{Dj} = F_{S0} + F_{reg} + \sum F_{Si,Dj} \tag{9}$$

where *NS* is the number of sources, *ND* is the number of sinks, $\sum F_{Si,Dj}$ is the flow rates of sources allocated to sinks, and F_{in} is the flow rate of feed stream, which is composed of S_{p+1},…, S_{NR}, and part of S_p. The hydrogen streams whose concentrations are higher than S_{NR} will be discharged. The item of $\sum_{i=1}^{NS} F_{Si}$ in Eq (8) can be expressed as:

$$\sum_{i=1}^{NS} F_{Si} = \sum_{i=1}^{NR} F_{Si} + \sum_{i=NR+1}^{NS} F_{Si} \tag{10}$$

Under the new case, Eq (11) can be obtained from Eq (3), Eq (8), Eq (9) and Eq (10):

$$\sum_{i=1}^{NR} F_{Si} - \sum_{j=1}^{ND} F_{Dj} = \left[\frac{1-C_{reg,1}}{R_1\left(1-C_{in,1}\right)} - 1 \right] F_{reg,1} - F_{S0,1} \tag{11}$$

Solving Eq (6), Eq (7) and Eq (11) simultaneously, we can obtain the consumption of hydrogen utility and the flow rate of purified stream at the new purified concentration. With the method proposed, when the purification pinch of a hydrogen network does not changed, the target values at a certain purified concentration can be predicted easily based on the known target values.

4. Case study

The data for this example taken from Liao et al.(2011) are shown in Table 1.The hydrogen recovery ratio is 95 % and the purified concentration is 2 mol %.

Table 1: Data for Example

Demand			Source		
Stream	C_{Dj}/mol %	F_{Dj}/mol·s^{-1}	Stream	C_{Si}/mol%	F_{Si}/mol·s^{-1}
D_1	19.39	2,495.00	S_0	5	
D_2	21.15	180.20	S_1	25	1,801.90
D_3	24.86	720.70	S_2	25	138.60
D_4	22.43	554.40	S_3	30	457.40
			S_4	27	346.50
			S_5	7	623.80
			S_6	20	415.80

With the iterative method of Yang et al. (2013), the following parameters at purified concentration of 2mol % can be obtained: the purification pinch source is S_3 with impurity concentration of 30 mol %, the hydrogen utility consumption is 196.76 mol·s^{-1}, the flow rate of purified stream is 64.33 mol·s^{-1}, and the flow rate of pinch source S_3 allocated to the sinks below the purification pinch is 362.60 mol·s^{-1}.

Let us predict the purification targets at purified concentration of 4 mol%. Substituting the given data into Eq (6), Eq (7) and Eq (11), we have:

$$\begin{cases} 196.76 + 64.33 + 362.60 = F_{S0,1} + F_{reg,1} + F_{S3,1}^{B} \\ 196.76 \times 0.05 + 64.33 \times 0.02 + 362.60 \times 0.30 = 0.05F_{S0,1} + 0.04F_{reg,1} + 0.30F_{S3,1}^{B} \\ 3{,}784 - 3{,}950.3 = \left[\dfrac{1-0.04}{0.95\times(1-0.30)} - 1 \right] F_{reg,1} - F_{S0,1} \end{cases} \tag{12}$$

It can be obtained that $F_{S0,1}$=196.95 mol·s^{-1} and $F_{reg,1}$ =69.10 mol·s^{-1}. The results obtained agree with that of Yang et al (2013) which includes five iterations. Similarly, the purification targets at other purified concentrations can be obtained with the proposed method. Table 2 lists the results predicted by this work and those obtained by the method of Yang et al. (2013). The above calculations and the results in Table 2 show that the proposed method effectively simplifies the targeting procedure of hydrogen networks involving purification.

For clarity, the relationship between F_{S0} ($F_{reg,1}$) and C_{reg} in Table 2 is depicted in Figure 3. It can be seen from Figure 3 that for this example, when the purified concentration increases, the hydrogen utility consumption increases slightly, but the flow rate of purified stream increases significantly. It means that a trade-off between operating performance and device capability should be carried out in designing purification unit. Hence, the proposed method can provide good insight for the determination of appropriate purified concentration.

Table 2: Results predicted by this work and those obtained by the method of Yang et al. (2013)

C_{reg}/mol·s^{-1}	Results by this work		Resultsby the method of Yang et al. (2013)	
	F_{S0}/ mol·s^{-1}	F_{reg}/mol·s^{-1}	F_{S0}/ mol·s^{-1}	F_{reg}/mol·s^{-1}
4	196.95	69.10	196.95	69.10
5	197.05	71.76	197.06	71.76
7	197.28	77.75	197.28	77.76
10	197.71	89.88	197.71	88.89
15	198.77	116.74	198.77	116.75

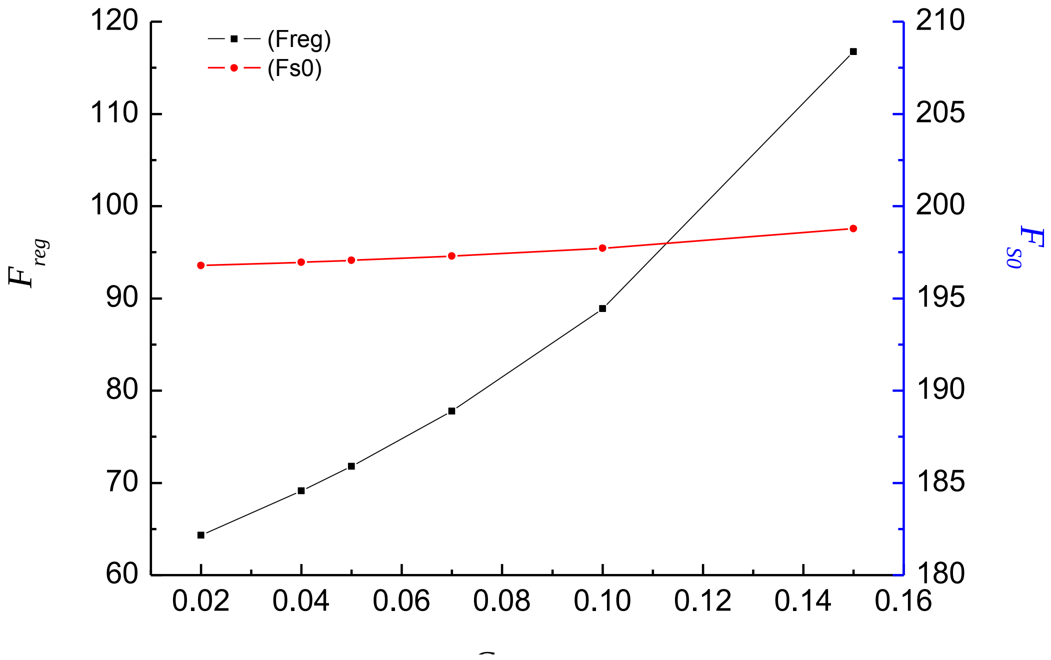

Figure 3.Relationship between F_{S0} ($F_{reg,1}$) and C_{reg} for the example

5. Conclusions

This paper presents a simple and effective method to predict the target values for hydrogen networks involving purification. The prediction is based on a few relationships developed from the balances of flow rate and impurity mass load below purification pinch, together with the characteristics of source-sink allocation of hydrogen networks involving purification. The method proposed can simplify the targeting of hydrogen networks involving purification. It should be noted that the proposed method is not suitable for the situation when the change of purified concentration leads to the change of purification pinch, which is our future work.

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