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A procedure for design of hydrogen networks with multiple contaminants☆

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ABSTRACT

It is necessary to reduce hydrogen consumption to meet increasingly strict environmental and product-quality regulations for refinery plants. In this paper, the concentration potential concepts proposed for design of water-using networks are extended to synthesis of hydrogen networks with multiple contaminants. In the design procedure, the precedence of processes is determined by the values of concentration potential of demands. The usage of complementary source pair(s) to reduce utility consumption is investigated. Three case studies are presented to illustrate the effectiveness of the method. It is shown that the design procedure has clear engineering meaning.

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1. Introduction

Hydrogen resources are widely used in hydrogenation process and desulfurization process to upgrade crude oil to light transportation fuels. Since crude oil is getting heavier and environmental regulations are becoming stricter, large amounts of hydrogen resources are required for hydro-treating processes. To reduce hydrogen consumption, one of the methods is synthesis of hydrogen networks.

A hydrogen network has several hydrogen consumers, such as hydro-cracking and hydro-treating processes, and hydrogen producers, such as catalytic reforming process. The inlet streams are called as the demands (or sinks), the outlet streams as the internal sources, and the hydrogen stream from outside suppliers as the external source (or utility).

Two widely used techniques to synthesize hydrogen networks are pinch analysis and mathematical programming methods. The pinch-based methods present clear physical insight, but can deal with single impurity hydrogen system only. Alves and Towler [1] proposed a method for calculating the minimum flow rate of hydrogen utility required, in which purity profile and hydrogen surplus diagram were employed to identify hydrogen pinch and the minimum amount of hydrogen utility. Liu *et al.* [2] analyzed the characteristics of pinch point and the effect of purification to the hydrogen surplus diagram [1], with the flow rate of purification feed given, the pinch location of the hydrogen system can be identified qualitatively. El-Halwagi *et al.* [3] developed a non-

iterative graphical technique for targeting the minimum utility consumption. Ding *et al.* [4] constructed average pressure profiles of sources and demands to design the hydrogen networks with pressure constraints. Zhao *et al.* [5] proposed a simple graphical method to identify the minimum utility consumption and pinch point. Based on ternary composition diagram [6] for three component systems, Wang *et al.* [7] proposed a graphical method for optimizing the match between multiple sources and one demand. Zhang *et al.* [8] proposed a graphical method for targeting and design of resource conservation networks with multiple contaminants, in which a triangle (polygon) rule was proposed to illustrate the match between multiple sources and a demand. Based on the ranked order of sources and demands, the material recovery pinch diagram for multiple contaminants was constructed. The resulting group of matching polygons gave the target and corresponding network.

The mathematical programming approaches are also important in the design of hydrogen networks. These methods can deal with complex system and achieve utility target, but the solving process is often complicated. Many mathematical models, including linear programming (LP), non-linear programming (NLP), mixed-integer linear programming (MILP) and mixed-integer non-linear programming (MINLP) models, have been developed to optimize hydrogen distribution systems [9–15]. Hallale and Liu [9] proposed a superstructure based approach for hydrogen networks accounting for pressure constraints and economic issues. By using the newly developed understanding of network interactions in refineries, Zhang *et al.* [10] proposed a method for overall refinery optimization through integration of hydrogen network and utility system with the material processing system. Khajepour *et al.* [11] proposed heuristic rules to reduce the complexity of the superstructure of hydrogen network,

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and the reduced superstructure was used to optimize hydrogen system. Kumar *et al.* [12] employed mathematical modeling technique to optimize the hydrogen distribution network and analyzed the characteristics of LP, NLP, MILP and MINLP models. Based on the automated design approaches of Hallale and Liu [9] and Liu and Zhang [13], Ahmad *et al.* [14] proposed a method for multi-period design of hydrogen networks. Jiao *et al.* [15] proposed two mathematical methods based on two-step approach and simultaneous optimization approach to make efficient use of hydrogen streams. Liao *et al.* [16,17] proposed a method for hydrogen systems by combining pinch insight and rigorous mathematical optimization technique.

In this paper, the approach developed for water-using networks with multiple contaminants [18] is extended to hydrogen networks with multiple contaminants. The method proposed in this study considers constraints on flow rate and concentrations simultaneously. First, operation order is determined based on the values of concentration potential of the demands. Then, a few rules satisfying demands with sources are deduced. Based on the source–demand matches, utility consumption and corresponding network can be obtained.

2. The Concentration Potentials of the Source and Demand Streams

A water network with single contaminant can be designed by satisfying the demands in their ascending order of impurity concentrations, but it is difficult to determine the concentration order of the streams with multiple contaminants. In the design of water-using network with multiple contaminants, Liu *et al.* [18] proposed to use concentration potentials of demand streams (CPDs) and sources (CPSs) to determine the concentration order of the streams. Here we give a brief introduction about them.

The limiting quasi-allocation ratio of S_i for unit amount of D_j is

$$R_{i,j} = \frac{C_{D_j,KC}^{\lim}}{C_{S_i,KC}} = \min_{m=1,2,\dots,NC} \left(\frac{C_{D_j,m}^{\lim}}{C_{S_i,m}} \right) \quad (1)$$

where $C_{D_j,m}^{\lim}$ is the limiting concentration of contaminant m in demand D_j , $C_{S_i,m}$ is the concentration of contaminant m in source S_i , and NC is the number of contaminants.

The contaminant(s) determining the value of $R_{i,j}$ is called as the reuse key contaminant(s) (KCs). The concentration of KC will reach its limiting value when D_j is satisfied by S_i . The concentration potential of demand D_j is the sum of the $R_{i,j}$ values for all the source streams:

$$CPD(D_j) = \sum_{i=1}^{NS} R_{i,j} = \sum_{i=1}^{NS} \min_{m=1,2,\dots,NC} \left(\frac{C_{D_j,m}^{\lim}}{C_{S_i,m}} \right) \quad (2)$$

where NS is the number of source streams. The value of $CPD(D_j)$ reflects the overall possibility of demand D_j to reuse the source streams.

Similarly, the value of $CPS(S_i)$ reflects the overall possibility of source S_i to be reused by demand streams,

$$CPS(S_i) = \frac{1}{\sum_{j=1}^{ND} \min_{m=1,2,\dots,NC} \left(\frac{C_{D_j,m}^{\lim}}{C_{S_i,m}} \right)} \quad (3)$$

where ND is the number of demand streams.

3. The Rules Satisfying a Demand with Complementary Source Pair(s)

3.1. The concept of complementary source pair

The concept of complementary sources [19] will be applied to hydrogen networks in this paper. When satisfying a demand, if the limiting quasi-allocation ratio for contaminant m of source S_i is larger than

unity and that of the same contaminant of another source, say source S_k , is smaller than unity, the two sources are called as complementary source pair. If the complementary source pair is used to satisfy the corresponding demand, utility consumption may be reduced [19].

We will use an example to show its application with the limiting data in Table 1. In Table 1, the limiting impurity mole concentrations of demand D_2 are 0.1% and 8.57%, with those of source S_1 as 0 and 9.2 and S_2 as 0.21 and 6.18. The limiting quasi-allocation ratios of S_1 and S_2 for D_2 are $R_{1,2} = \min\{\infty, 0.93\} = 0.93$ and $R_{2,2} = \min\{0.48, 1.39\} = 0.48$, which are lower than unity, so that utility is needed. For contaminant H_2S , the limiting quasi-allocation ratio of S_1 for D_2 is infinity (∞), which is larger than unity, while that of S_2 for D_2 is 0.48, lower than unity. For contaminant A , the limiting quasi-allocation ratio of S_1 for D_2 is 0.93, which is lower than unity, while that of S_2 for D_2 is 1.39, larger than unity. For this case, if S_1 with an amount of $166.57 \text{ mol} \cdot \text{s}^{-1}$ and S_2 with an amount of $151.43 \text{ mol} \cdot \text{s}^{-1}$ are used to satisfy D_2 , it is not necessary to use the utility, so that utility consumption can be reduced.

Table 1
Limiting data for case 1 [20]

		$F/\text{mol} \cdot \text{s}^{-1}$	$H_2/\text{mol}\%$	$H_2S/\text{mol}\%$	$A/\text{mol}\%$
Sources	Utility		99.99	0	0.01
	S_1	262	90.8	0	9.2
	S_2	295	93.61	0.21	6.18
	S_3	104	86.63	1.03	12.34
Demands	D_1	122	92.81	0	7.19
	D_2	318	91.33	0.1	8.57
	D_3	197	90	1	9

3.2. The rules satisfying a demand with complementary source pairs

When satisfying a demand with complementary source pairs, utility consumption may be reduced, as discussed above. If a few complementary source pairs are available, we should determine the one to be used. We will use the following rules to choose complementary source pair.

- (1) When satisfying demand D_j , if a few complementary source pairs are available, arrange the pairs in the ascending order of $R_{h,j}$ values, where $R_{h,j}$ is the higher value of the quasi-allocation ratio of a complementary source pair; if a few complementary source pairs have the same $R_{h,j}$ value, arrange them in ascending order of $R_{l,j}$ values, where $R_{l,j}$ is the lower value of the quasi-allocation ratio of a complementary source pair.
- (2) Consider the complementary source pairs in the above order. If one of the complementary source pairs can satisfy the demand without utility, this pair will be used to satisfy the demand. If none of the pairs can satisfy the demand without utility, choose the complementary source pair corresponding to the minimum amount of utility consumption to satisfy the demand.

The procedure of selecting complementary sources to satisfy a demand is shown in Fig. 1.

4. Design Procedure

We will use similar rules for design of water-using networks proposed by Liu *et al.* [18]. The design procedure proposed in this study is as follows.

- (1) Calculate the values of CPD and CPS based on the limiting concentrations;
- (2) The demand with the lowest CPD value will be satisfied first. If a few processes have the same CPD values, the process with the highest hydrogen purity will be satisfied first;
- (3) If a source stream is used up, it will not be considered in the following steps;

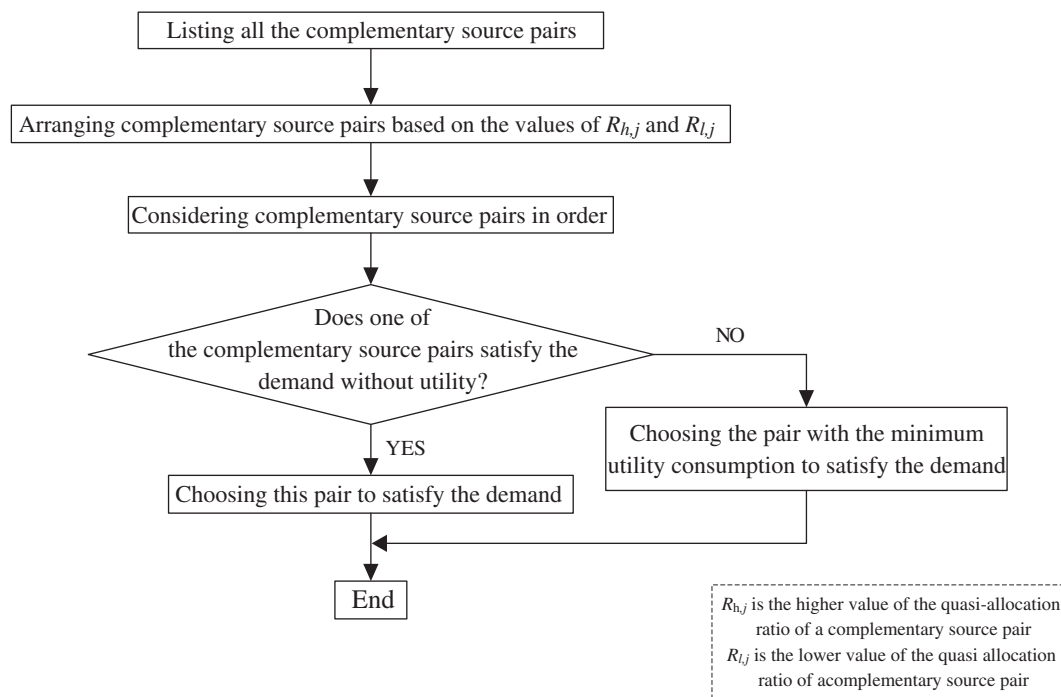


Fig. 1. The procedure of satisfying a demand with complementary source pairs.

- (4) Allocate the source streams to the demand identified;
- (5) Repeat Steps (2)–(4) until all the demands are satisfied.

The procedure of allocating source streams to a demand is as follows.

- (a) If a source with $R_{i,j} = 1$ is available, it will be used to satisfy the demand, which will often reduce connection number; otherwise, go to Step (b);
- (b) If there is complementary source pair(s) for a demand, the demand can be satisfied by using the rules of complementary source pairs; otherwise, the demand can be satisfied by the source with the largest quasi-allocation ratio and utility; if a few sources have the same quasi-allocation ratios, the source with the highest CPS value will be selected (saving the sources with lower CPS value may reduce utility consumption for the downstream processes).

The design procedure is shown in Fig. 2.

5. Case Studies

5.1. Case 1

This case is taken from Zhao *et al.* [20], with the limiting data shown in Table 1. The values of CPD and CPS based on the limiting concentrations are listed in Table 2. In this case, demand D_1 is satisfied first, because its CPD value is the minimum. For demand D_1 , the quasi-allocation ratio of S_1 is the largest, so S_1 is used first. Because $R_{1,1} < 1$, demand D_1 will be satisfied by S_1 and utility. The consumption amounts of S_1 and utility are 95.32 and $26.68 \text{ mol} \cdot \text{s}^{-1}$, respectively. Based on the CPD values, D_2 is satisfied now. A complementary source pair is for D_2 , S_1 and S_2 ($R_{1,2} = \min\{\infty, 0.93\} = 0.93$, $R_{2,2} = \min\{0.48, 1.39\} = 0.48$). The calculation shows that complementary source pair S_1 and S_2 can satisfy D_2 without utility consumption. The consumption amounts of S_1 and S_2 are 166.57 and $151.43 \text{ mol} \cdot \text{s}^{-1}$, respectively. Demand D_3 is satisfied then. S_2 and S_3 are complementary source pair for D_3 ($R_{2,3} = \min\{4.76, 1.46\} = 1.46$, $R_{3,3} = \min\{0.97, 0.73\} = 0.73$). Based on mass balance of contaminant A and flow rate balance, the

consumption amounts of S_2 and S_3 are 106.81 and $90.19 \text{ mol} \cdot \text{s}^{-1}$, respectively.

The total utility consumption is $26.68 \text{ mol} \cdot \text{s}^{-1}$. The final concentrations are shown in Table 3, with the maximum concentrations shadowed. The design is shown in Fig. 3. The connection number of this work is 6. The design obtained is the same as that of Zhao *et al.* [20], in which iterative method was used and their calculation effort was heavier.

5.2. Case 2

The limiting data for Case 2 are shown in Table 4. There are three impurities, seven source streams (including utility) and six demand streams. The values of CPD and CPS based on the limiting concentrations are listed in Table 5. Demand D_1 is satisfied first, because its CPD value is the smallest. The consumption of utility is $219 \text{ mol} \cdot \text{s}^{-1}$. Then, D_4 is satisfied and source S_2 is used. Because $R_{2,4} < 1$, D_4 is satisfied by S_2 and utility. The consumption amounts of S_2 and utility are 105.77 and $169.23 \text{ mol} \cdot \text{s}^{-1}$, respectively. Demand D_6 is satisfied now. A few complementary source pairs are available for D_6 , S_3 and S_2 , S_3 and S_4 , S_3 and S_1 . However, none of the pairs can satisfy D_6 without utility. When pair S_2 and S_3 is used, utility consumption is the lowest. The consumption amounts of S_2 , S_3 and utility are 90.41 , 275.66 and $24.93 \text{ mol} \cdot \text{s}^{-1}$, respectively. Then demand D_2 is satisfied. There are a few complementary source pairs (S_4 and S_2 , S_4 and S_3 , S_4 and S_5 , S_4 and S_6 , S_4 and S_1 , S_2 and S_1 , S_3 and S_1). The pair of S_2 and S_4 is used to satisfy demand D_2 . The consumption amounts of S_2 and S_4 are 108.86 and $18.14 \text{ mol} \cdot \text{s}^{-1}$, respectively. For demand D_5 , complementary sources S_2 and S_3 are used. Because the amount of S_2 is not sufficient, the consumption amounts of S_2 and S_3 are 122.96 and $192.04 \text{ mol} \cdot \text{s}^{-1}$, respectively. For D_3 , its complementary sources, S_3 and S_6 , are used. The consumption amounts of S_3 and S_6 are 68.21 and $122.79 \text{ mol} \cdot \text{s}^{-1}$, respectively.

The concentrations of the streams are shown in Table 6, with the maximum concentrations shadowed. The design is shown in Fig. 4. The total utility consumption is $413.16 \text{ mol} \cdot \text{s}^{-1}$. The design without considering matches of complementary sources is shown in Fig. 5, and the total utility consumption is $434.23 \text{ mol} \cdot \text{s}^{-1}$. Compared to the design in Fig. 4, the connection number for the design in Fig. 5 is decreased by one, but the utility consumption increases by 4.85%.

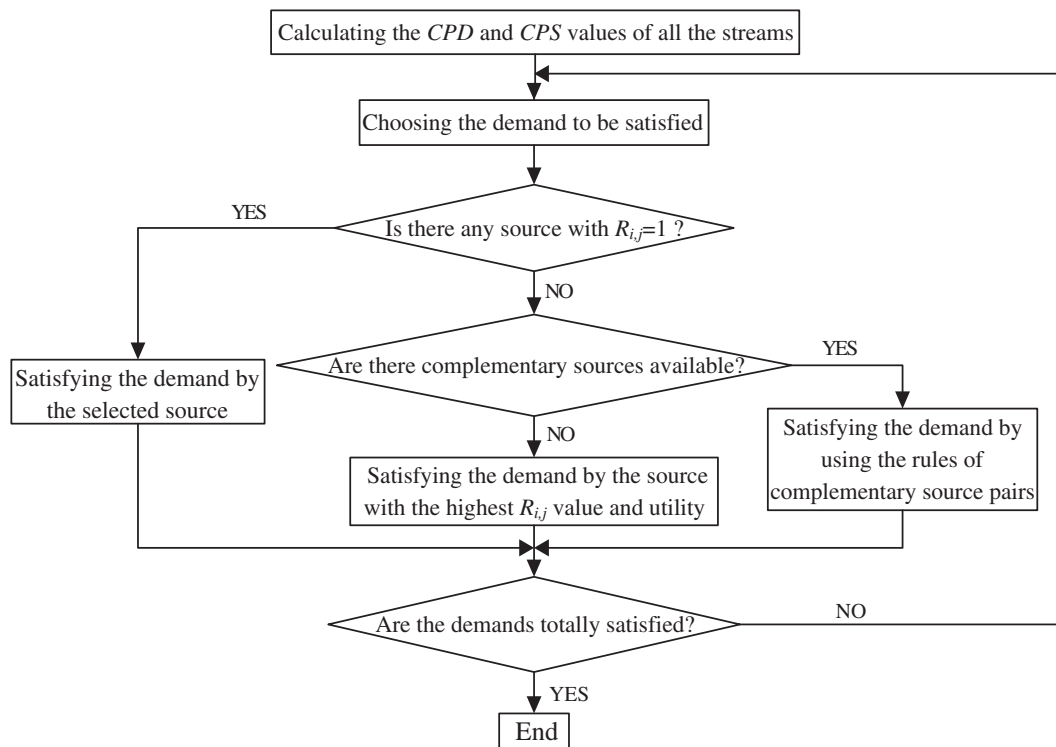


Fig. 2. Design procedure.

Table 2

Concentration potential values of Case 1

D_j	D_1	D_2	D_3
$CPD(D_j)$	0.78	1.5	3.16
Order	1	2	3
S_i	S_1	S_2	S_3
$CPS(S_i)$	0.37	0.52	1.21
Order	1	2	3

Table 3

Final concentration of the streams of Case 1

Demands	$F/\text{mol} \cdot \text{s}^{-1}$	$\text{H}_2/\text{mol}\%$	$\text{H}_2\text{S}/\text{mol}\%$	$A/\text{mol}\%$
D_1	122	92.81	0	7.19
D_2	318	92.14	0.1	7.76
D_3	197	90.41	0.59	9

Table 4

Limiting data for Case 2

		$F/\text{mol} \cdot \text{s}^{-1}$	$\text{H}_2/\text{mol}\%$	$\text{H}_2\text{S}/\text{mol}\%$	$N/\text{mol}\%$	$C/\text{mol}\%$
Sources	Utility		99.99	0	0.01	0
	S_1	176	87.2	1	2.3	9.5
	S_2	428	95.4	1.3	1.5	1.8
	S_3	576	97.2	1.7	0.5	0.6
	S_4	295	93.22	0.6	5.14	1.04
	S_5	104	85.4	2.26	7.23	5.11
Demands	S_6	271	89.4	4.5	2.6	3.5
	D_1	219	82.3	0	7.8	9.9
	D_2	127	91.33	1.2	5.4	2.07
	D_3	191	87.1	3.5	3.9	5.5
	D_4	275	98.2	0.5	0.6	0.7
	D_5	315	84.7	3	1.2	11.1
	D_6	391	96.8	1.5	0.7	1

Table 5

Concentration potential values of Case 2

D_j	D_1	D_4	D_6	D_2	D_5	D_3
$CPD(D_j)$	0	1.06	1.96	3.57	3.95	7.31
Order	1	2	3	4	5	6
S_i	S_3	S_2	S_4	S_6	S_1	S_5
$CPS(S_i)$	0.18	0.19	0.44	0.53	0.67	0.78
Order	1	2	3	4	5	6

Table 6

Final concentration of the streams of Case 2

Demands	$F/\text{mol} \cdot \text{s}^{-1}$	$\text{H}_2/\text{mol}\%$	$\text{H}_2\text{S}/\text{mol}\%$	$N/\text{mol}\%$	$C/\text{mol}\%$
D_1	219	99.99	0	0.01	0
D_2	127	95.09	1.2	2.02	1.69
D_3	191	92.19	3.5	1.85	2.46
D_4	275	98.23	0.5	0.58	0.69
D_5	315	96.5	1.54	0.89	1.07
D_6	391	96.96	1.5	0.7	0.84

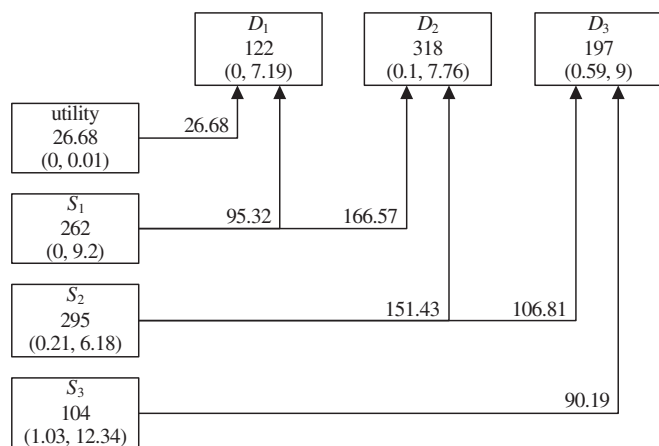


Fig. 3. Network design of Case 1.

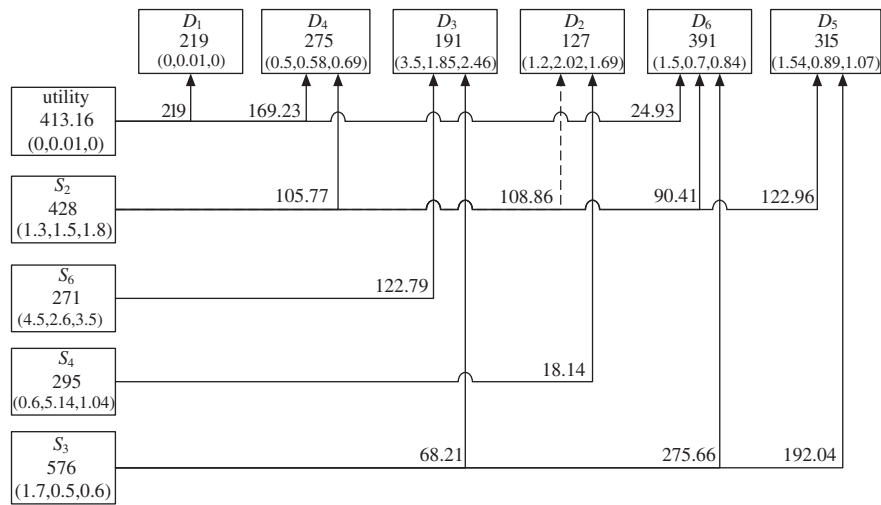


Fig. 4. Network design for Case 2.

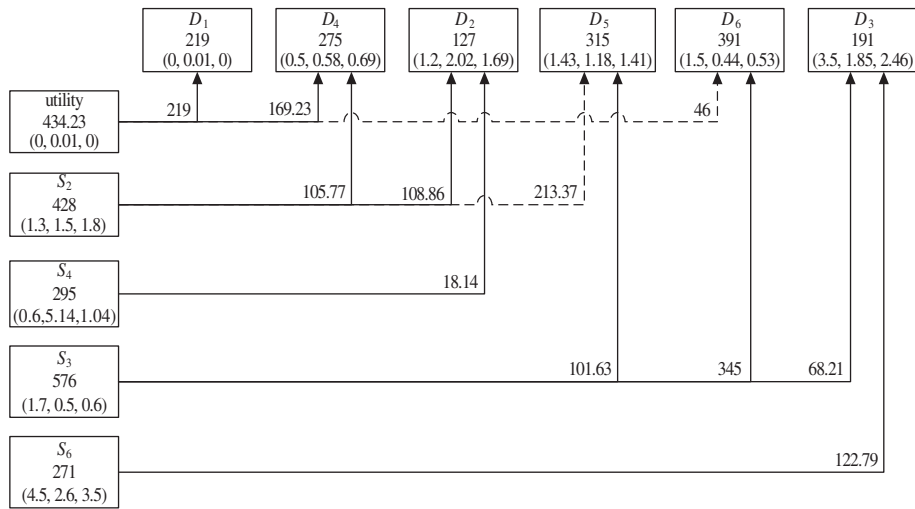


Fig. 5. Network design for Case 2 without considering matching of complementary sources.

5.3. Case 3

This case is taken from a refinery. In the original system, the utility is pure hydrogen. The limiting data for Case 3 are shown in Table 7. The values of CPD and CPS based on the limiting concentrations are listed in Table 8. Demand D₁ is satisfied first, and the consumptions of S₁ and utility are 23.04 and 72.96 mol·s⁻¹, respectively. Then, demand

Table 7
Limiting data for Case 3 [21]

		F/mol·s ⁻¹	Salt/mol%	H ₂ S/mol%	Hydrocarbon/mol%
Sources	Utility		0	0	0
	S ₁	110	0.5	4.30	1.82
	S ₂	770.13	1.03	4.2	4.07
	S ₃	68	1.4	8.17	2.52
	S ₄	115	3.52	1.2	9.09
	S ₅	45	5.1	6.75	3.2
Demands	S ₆	28	7.25	5.2	10.44
	D ₁	96	0.12	1.42	4.04
	D ₂	21	1	3.56	2.6
	D ₃	351	2.1	2.84	4.1
	D ₄	579	3.48	4.00	4.3
	D ₅	52	5.6	1.33	6.2
	D ₆	45	6.82	8.5	1.37

D₅ is satisfied. A few complementary source pairs are for D₅, but none of them can satisfy the demand without utility consumption. It is found that the pair of S₁ and S₄ is the best. The consumption amounts of S₁, S₄ and utility are 6.56, 34.14 and 11.30 mol·s⁻¹, respectively. Demand D₆ is satisfied by S₁ and utility, and the consumption amounts are 33.87 and 11.13 mol·s⁻¹, respectively. Demand D₂ is satisfied by S₁ (16.64 mol·s⁻¹), S₄ (2.68 mol·s⁻¹) and utility (1.68 mol·s⁻¹). Demand D₃ is satisfied by S₂ (220.29 mol·s⁻¹), S₄ (59.68 mol·s⁻¹) and utility (71.03 mol·s⁻¹). Demand D₄ is satisfied by S₂, S₄ and utility, and the consumption amounts are 546.14, 18.5 and 14.36 mol·s⁻¹, respectively.

The design obtained is shown in Fig. 6. The final concentrations are shown in Table 9. The total utility consumption is 182.46 mol·s⁻¹, which is slightly larger than the utility consumption 182.38 mol·s⁻¹

Table 8
Concentration potential values of Case 3

D _j	D ₁	D ₅	D ₆	D ₂	D ₃	D ₄
CPD(D _j)	0.52	1.92	2.34	2.52	2.84	3.85
Order	1	2	3	4	5	6
S _j	S ₁	S ₂	S ₄	S ₃	S ₅	S ₆
CPS(S _j)	0.27	0.33	0.482	0.484	0.54	0.8
Order	1	2	3	4	5	6

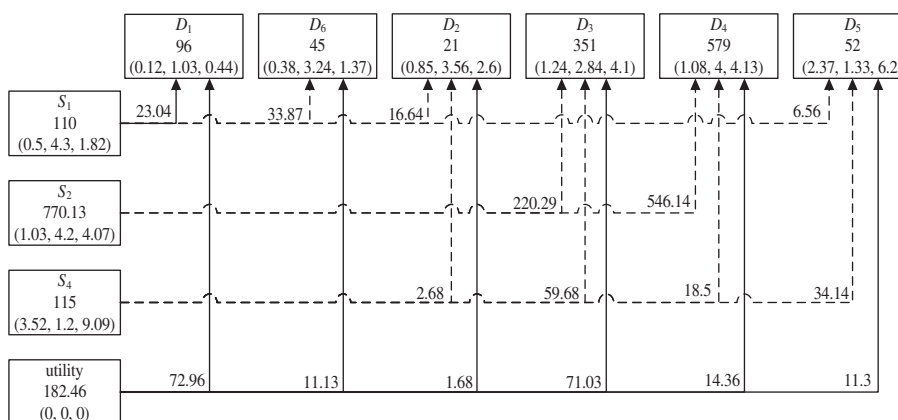


Fig. 6. Network design of Case 3.

Table 9

Final concentration of the streams of Case 3

Demands	$F/\text{mol} \cdot \text{s}^{-1}$	Salt/mol%	$\text{H}_2\text{S}/\text{mol}\%$	Hydrocarbon/mol%
D_1	96	0.12	1.03	0.44
D_2	21	0.85	3.56	2.6
D_3	351	1.24	2.84	4.1
D_4	579	1.08	4	4.13
D_5	52	2.37	1.33	6.2
D_6	45	0.38	3.24	1.37

reported by Liu *et al.* [21], who used an LP programming to solve the problem. The connection number of this work is 16 and that of the design of Liu *et al.* [21] is 17.

6. Conclusions

A method based on concentration potential concepts is proposed for design of hydrogen network with multiple contaminants. The usage of complementary source pair(s) to reduce utility consumption is investigated. The values of concentration potential of demands are used to determine the allocation order of processes. The illustrated examples show that the results obtained in this work are comparable to that in the literature.

Nomenclature

C	concentration of impurity, mol%
CPD	concentration potential of demand
CPS	concentration potential of source
D	hydrogen demand
F	flow rate of hydrogen stream, $\text{mol} \cdot \text{s}^{-1}$
R	quasi-allocation ratio
S	hydrogen source

Superscripts

lim	limiting data
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Subscripts

h	source with higher quasi-allocation ratio of a complementary source pair
i	process of hydrogen source
j	process of hydrogen demand
k	process of hydrogen source
l	source with lower quasi-allocation ratio of a complementary source pair
m	contaminant

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