

# Flexibility Analysis and Design of Heat Exchanger Network for Syngas-to-methanol Process

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## Research

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# Abstract

The heat exchanger network (HEN) of syngas-to-methanol process was designed and optimized based on pinch technology under the stable operation conditions to balance the energy consumption and economic gain. Inevitably, the fluctuations of production affect the stable operation of HEN in real industrial processes. The flexibility analysis of HEN was carried out in this study to minimize such disturbances by using the downstream paths method. The results show that 2/3 downstream paths cannot meet flexibility requirements, indicating that HEN doesn't have enough flexibility to accommodate the disturbances in the actual production. The flexible HEN was then designed with the methods of dividing and subsequent merging, which led to 13.89% and 20.82% reduction in energy consumption and total cost, respectively. Thanks to enough area margin and additional alternative heat exchangers, the flexible HEN is found to be able to resist interferences and maintain the production stability and safety, only sacrificing the increase of total cost increase by 4.08%.

## 1 Introduction

Methanol is one of the most important raw materials in the chemical industry for producing formaldehyde, acetic acid, methyl formate, etc., and it also has been used as the fuel for methanol automobiles(Li et al., 2010; Riaz et al., 2013). Along with the rapid development of the coal chemical industry in recent years, methanol has played as an important intermediate in the comprehensive coal utilization processes, including coal-to-olefins, coal-to-dimethyl ether, and other coal-to-chemicals conversions (Galadima and Muraza, 2015; Gao et al., 2018). Generally, the syngas from coal gasification technology is used to synthesize methanol via the catalytic processes. Coal-based syngas-to-methanol technology has been the main route for methanol production, especially in China, due to its special status energy resources: rich in coal, poor in oil and natural gas(Li et al., 2010).

Several works have focused on the energy utilization efficiency of coal-based syngas-to-methanol process since it is a remarkable energy-intensive process(Riaz et al., 2013; Bessa et al., 2012; Cui et al., 2017; Rashid et al., 2011; Sun et al., 2012). Taking the whole process of coal-to-olefins as an example, which contains six sub-processes, including coal mining, transportation, coal-to-methanol (CTM), methanol-to-olefins (MTO), products delivery, and carbon capture and storage (CCS), respectively(Gao et al., 2018). The subsection of coal-to-methanol consumes the most energy in the whole process of coal-to-olefins, which dominates 71.04% of total energy consumption and it is five times more than CCS subsection (13.81%)(Gao et al., 2018). Extensive efforts have been made to develop the energy-saving strategies of coal-to-methanol process, among which the purification technology of crude methanol has attracted much attentions. A five-column heat integrated methanol distillation scheme was proposed by adding a medium-pressure column, and its total energy loss can be conspicuously reduced by 21.5% comparing to the previous four-column purification scheme(Sun et al., 2012; Chien et al., 2005). Besides, a hybrid methanol purification process was built to improve energy efficiency, which elaborately combined heat pump distillation with double-effect thermal integration by designing an intermediate heater to shunt the heat load of the reboiler(Bessa et al., 2012; Douglas and Hoadley, 2006). Although these above-mentioned strategies contributed to energy conservation on certain subsections in syngas-to-methanol process, other energy-saving alternatives regarding the overall process should also be raised more attention.

The study of heat exchanger network (HEN) can obtain a comprehensive strategy of heat exchange for the whole process of syngas-to-methanol by considering energy utilization, energy efficiency, operation cost, equipment cost, etc. Energy consumption is able to be reduced obviously by using optimized heat exchange strategy according to the result of global HEN analysis(Kang and Liu, 2019). Currently, the optimal calculation of HEN is performed based on the pinch technology which was developed for analyzing the potential energy-saving and economic gain(Rashid et al., 2011). Some previous studies(Kang and Liu, 2019; Payet et al., 2018) have discussed that the energy consumption and

operation cost of syngas-to-methanol process can be saved through restructuring HEN referring to the result of pinch analysis. However, the optimization of HEN was always carried out under the stable heat transfer operating conditions, which ignored some inevitable uncertainties arisen from various external and internal factors (for example, feed status, product output, heat transfer coefficients, fouling, and so on). In order to make the optimal HEN from pinch analysis more precise and practical, the flexibility analysis was introduced with considering those uncertain factors under real operation status (Payet et al., 2018; ZHU et al., 1996). However, only very few reports have focused on the study of the flexibility analysis of HEN for methanol production so far.

In this work, the process of syngas-to-methanol was firstly simulated with optimized HEN, which is conducted by pinch technology with the assumption of stable operating conditions. Then, the flexibility analysis of optimized HEN was implemented by using the heuristic method. The “downstream paths” approach was used to determine the variables for building flexible HEN. Besides, the flexible HEN was improved to meet the requirements for actual production with enough area margin and heat exchangers amount. This work proposed a feasible strategy to save energy and equipment cost as well as operation cost of syngas-to-methanol by performing the flexible analysis on optimized HEN.

## **2 Methodology And Simulation**

### **2.1 Pinch technology**

Pinch technology is a methodology for minimizing energy consumption of chemical processes which was put forward by Linnhoff (LINNHOFF and HINDMARSH, 1983). Pinch technology has been widely used in the new design and construction of HEN as well as in the energy-saving modification of old equipment. The mechanism of pinch analysis is based on thermodynamics with the computation method of topology to analyze the distribution of energy flows along with the temperature in the process. The objects of pinch analysis are to find the heat integration bottleneck, namely “the pinch”, and offer a solution to debottleneck for the energy consumption process (Binosi and Papavassiliou, 2009; Tan et al., 2014). Aspen Energy Analyzer software is utilized here to compute and optimize HEN according to pinch technology.

### **2.2 Flexible analysis**

Generally, the design and optimization of HEN are based on stable operation conditions. However, the inevitably uncertainties affect the stability of HEN in the actual production. Uncertain factors (e.g., feed status fluctuation) are able to cause the operation of HEN to deviate from the optimal state and even stop operating. Thus, for HEN design and optimization, it is necessary to meet the flexibility requirements under fluctuated operation conditions, other than only considering the stable operation conditions. The concept of the flexible index was first proposed by Swaney and Grossmann in 1987 to measure the maximum deviation of uncertain factors from stable operation conditions in the feasible region, and also the strict mathematic model was established (SWANEY and GROSSMANN, 1985). The flexibility analysis contains mathematical programming method and heuristic method (Grossmann and Kravanja, 1995; Linnhoff and Kotjabasakis, 1986). The mathematical programming method is always used to study the feasibility of HEN in the specified disturbed domain under certain constraints, while the heuristic method is mainly applied to the detailed structure analysis of HEN to determine feasibility in the whole disturbed domain based on the experience in chemical engineering (Grossmann and Kravanja, 1995; Linnhoff and Kotjabasakis, 1986). The heuristic method is much simpler and more convenient than the mathematical programming method, which is used more widely in the field of chemical engineering, particularly in a relatively short chemical process. Besides, the feasible and precise strategy of HEN can be obtained by the heuristic method, whereas the mathematical programming method is only used for testing the feasibility of existing HEN under the different disturbing conditions. In the heuristic method, the concept of downstream paths was developed to solve the balance between flexibility and economics of HEN, and downstream

paths had been used as a convenient approach for determining the disturbed and controlled variables of HEN during flexibility analysis (Linnhoff and Kotjabasakis, 1986). A path was defined as the uninterrupted connection between any two nodes in the grid diagram of HEN, meanwhile, a downstream path was defined as the directed path which was always in the same direction of streams through which it passed (ZHU et al., 1996). Thus, the disturbance occurring in HEN can only affect the controlled variables along with the downstream paths between disturbed variables and controlled variables.

## 2.3 Process simulation

The technical route of this work is shown in **Fig. S1**. The process simulation was carried out by Aspen Plus V10. The original HEN, traditional optimal HEN, and flexible HEN were designed and optimized by Aspen Energy Analyzer V10 based on the theories of pinch technology and heuristic method. The whole process of syngas-to-methanol was divided into two parts for simulation, one part was named as methanol synthesis, and another was termed as methanol purification as shown in Fig. 1. The information of all blocks and streams are listed in **Table S1**. Lurgi low-pressure methanol synthesis process was adopted to simulate the methanol synthesis process. The synthetic reaction was carried out at 250 °C and 5 MPa in a Lurgi reactor, modeled by combining RPLUG reactor block with RSTOIC reactor block. RPLUG reactor block and RSTOIC reactor blocks were used to model two main reactions and six side reactions, respectively. The reaction equations and kinetic parameters are displayed in **Table S2**. The parts of methanol synthesis and methanol purification were connected by the stream PCF-IN. In the methanol purification process, the small amount of light components (i.e. dimethyl ether and methyl formate, etc.) dissolved in crude methanol were removed in the pre-distillation column (Block PC). The double-effect distillation, constituted by combining high-pressure column (Block HC) with atmospheric column (Block CC), was applied to remove other impurities including water, ethanol, n-butanol, etc. The purity of obtained methanol product reached 99.98 wt.%. The compositions and volume fractions of syngas and crude methanol are shown in **Table S3** and **Table S4**, respectively.

## 3 Design And Optimization Of Hen

### 3.1 The determination of energy targets

Table 1  
The datasheet of streams involving in heat exchange

Steam No.	Steam name	$T_{in}/^{\circ}\text{C}$	$T_{out}/^{\circ}\text{C}$	Enthalpy /(kJ/h)
H1	REA-OUT_To_28	250.0	40.0	$1.52 \times 10^8$
H2	HCW-OUT_To_CCF-IN	129.2	80.3	$5.86 \times 10^6$
H3	HCD-OUT_To_49	122.3	121.8	$1.46 \times 10^8$
H4	24_To_16	122.3	40.0	$5.58 \times 10^6$
H5	27_To_W-WATWER	107.3	40.0	$1.80 \times 10^6$
H6	To Condenser@CC_TO_CCD-OUTDuplicate	74.9	71.0	$1.46 \times 10^8$
H7	PCD-OUT_To_S1-IN	73.3	64.0	$1.55 \times 10^7$
H8	CCD-OUT_To_31	71.0	40.0	$2.22 \times 10^6$
H9	S1D-OUT_To_S2-IN	64.0	40.0	$1.71 \times 10^6$
C1	10_To_REA-IN	124.0	230.0	$5.51 \times 10^7$
C2	To Reboiler@HC_TO_HCW-OUTDuplicate	126.9	129.2	$1.46 \times 10^8$
C3	21_To_HCF-IN	81.7	127.0	$8.76 \times 10^6$
C4	52_To_CCW-IN	107.3	110.9	$1.46 \times 10^8$
C5	To Reboiler@PC_TO_PCW-OUT	81.1	81.7	$1.85 \times 10^7$
C6	23_To_PCF-IN	20.4	75.0	$8.65 \times 10^6$

The datasheet of process simulation obtained by Aspen Plus is imported directly into Aspen Energy Analyzer for designing HEN. The temperature and enthalpy of each stream participating heat exchange are listed in Table 1. Through the computation of Aspen Energy Analyzer, the total cost index is maintained at a low level when the minimum heat transfer temperature difference ( $\Delta T_{min}$ ) located between 4 °C and 11 °C (**Fig. S2**). Here the value of  $\Delta T_{min}$  is set to 11 °C since the empirical value of  $\Delta T_{min}$  generally situated in the range of 10 °C to 20 °C by considering the factors of utilities and heat exchanger equipment cost, heat-exchange media, heat transfer coefficient, operating flexibility, and other factors (Kang and Liu, 2019). The composite curves and grand composite curve are shown in **Fig. S3** and **Fig. S4**, respectively. The pinch point temperatures of hot stream and cold stream were 135 °C and 124 °C from the composite curves (see **Fig. S3**) when  $\Delta T_{min}$  was set as 11 °C. Besides, the minimum hot utility ( $Q_{H,min}$ ) target was  $1.460 \times 10^8$  kJ/h, while the minimum cold utility ( $Q_{C,min}$ ) target was  $2.396 \times 10^8$  kJ/h. According to **Fig. S4**, the average temperature to process pinch point was confirmed as 129.5 °C when the corresponding enthalpy was 0 kJ/h.

## 3.2 Original HEN and traditional optimal HEN

The original HEN of syngas-to-methanol process generated by Aspen Energy Analyzer is shown in **Fig. S5**. The original HEN includes two process-process heat exchangers (white matches), nine coolers (blue matches), and five heaters (red matches). The original HEN was optimized under the assumption of stable operation conditions to gain traditional optimal HEN, shown in **Fig. S6**. The optimized HEN contains seven process-process heat exchangers, seven coolers, and two heaters.

## 3.3 Flexible HEN

### 3.3.1 Flexibility analysis of HEN

The disturbed variables and controlled variables of HEN in syngas-to-methanol process were determined by the downstream paths approach. Disturbed variables fluctuate disorderly due to the changes in the external environment, equipment operation, production load, etc. In the actual production, the temperatures of several streams are determined as the disturbed variables, i.e. the feedstock stream and outlet stream of a reactor, the mixing stream of crude methanol with water, and the bottom outlet stream of the pre-distillation column (Rashid et al., 2011). Here,  $\theta$  is denoted as the disturbed variables.  $\theta_1$  is the temperature of the feedstock stream  $C1$ ;  $\theta_2$  is the temperature of the outlet stream ( $H1$ ) of the reactor;  $\theta_3$  is the temperature of the mixing stream ( $C6$ ) of crude methanol with water, and  $\theta_4$  is the temperature of bottom outlet stream ( $C3$ ) of the pre-distillation column. Controlled variables are defined as the ones that are strictly manipulated to maintain stability and ensure safety in the actual production. In syngas-to-methanol system, the inlet stream temperature of the reactor needs to be strictly controlled to ensure the stability of the production load and feedstock conversion rate. Also, the control on the feed stream temperature of the pressured column and the atmospheric column is highly required, due to the sensitive inlet temperature requirements for double-effect distillation. Therefore, the temperatures of (1) the reactor inlet stream, (2) the feed streams of pressured column and atmospheric column, were set as the controlled variables. Here,  $z$  was termed as the controlled variables,  $z_1$  was the temperature of the reactor inlet stream ( $C1$ ),  $z_2$  was the temperature of feed stream ( $C3$ ) of the pressured column, and  $z_3$  was the temperature of feed stream ( $H2$ ) of the atmospheric column. Figure 2 shows the positions of disturbed variables and controlled variables in the grid diagram of traditional optimal HEN.

Table 2  
The node adjacency matrix of traditional optimal HEN

$j \setminus i$	1	2	3	4	5	6	7	8	9	HE1	HE2	CE1	CE2	$\theta_1$	$\theta_2$	$\theta_3$	$\theta_4$	$z_1$	$z_2$	$z_3$	
1	*									*					*						
2	*	*									*										
3			*					*									*				
4				*																	
5					*			*													
6						*	*														
7							*									*					
8		*						*													
9			*		*				*												
HE1										*				*							
HE2											*										
CE1									*			*									
CE2								*					*								
$\theta_1$														*							
$\theta_2$															*						
$\theta_3$																*					
$\theta_4$																	*				
$z_1$	*																	*			
$z_2$			*																	*	
$z_3$						*															*

A method for determining downstream paths was developed by using the node adjacency matrix that represented the connection of units along the direction of HEN streams (ZHU et al., 1996). Units, splitting, and mixing points as well as various variables in HEN are all thought as nodes. In the adjacency matrix, both heading row and heading column represent the above-mentioned nodes. The  $(i, j)$ th entry of the matrix is filled with a '\*' if node  $j$  can directly reach node  $i$  in the direction of a stream; otherwise, it leaves a blank. The nodes of coolers or heaters in the matrix which are not adjacent to other nodes are deleted for the sake of simplicity (Table 2). In this case, the determination of downstream paths between disturbed variables and controlled variables can be transformed into finding the downstream paths between the two nodes,  $i$  and  $j$ . The detailed approach for finding a downstream path is described as follows: (Step 1)  $\theta_i (i = 1, 2, \dots, I)$  and  $z_j (j = 1, 2, \dots, J)$  were used to represent the number of disturbed variables and controlled variables of HEN, respectively. (Step 2)  $S$  was defined as a set of nodes, and the serial number of these nodes should start from  $i = 1$ . (Step 3) For the last node  $p$  in  $S$ , the node  $q$  that can be directly reached from the node  $p$  along with the direction of stream needs to be confirmed, i.e., if  $(p, q)$ th entry is filled with '\*' in the node adjacency matrix, then add  $q$  to set  $S$ . Step

Step 4 will stop if  $S$  contains all nodes of HEN or  $(p, q)$ th entry is blank in the node adjacency matrix for each  $q$ . If node  $z_j$  is included in the set  $S$ , then there is a downstream path between node  $\theta_i$  and node  $z_j$ . Otherwise, let  $i = i + 1$  and go back to Step 4 until  $i > l$ . For the traditional optimal HEN, the results of identifying downstream paths between all disturbed variables and controlled variables are given in Table 3, and each downstream path is plotted in Fig. 3.

Table 3  
Downstream paths between disturbed variables and controlled variables

Number of downstream paths	Initial node	Process nodes	Final node
1	$\theta_1$	HE1, 1	$z_1$
2	$\theta_1$	HE1, 1, 2, 8, 3	$z_2$
3	$\theta_2$	1	$z_1$
4	$\theta_2$	1, 2, 8, 3	$z_2$
5	$\theta_3$	7, 6	$z_3$
6	$\theta_4$	3	$z_2$

### 3.3.2 Flexibility design of HEN

For a HEN with temperature fluctuations, heat recovery will reach the maximum when the inlet temperatures of all hot streams take the upper limit of fluctuations and meanwhile the inlet temperatures of all cold streams perform the lower limit. With this assumption, it is possible to get the maximum design margin and subsequently to meet the requirements for varying operation conditions in actual production. The stream dividing principle is used to design flexible HEN (Zhihong and Ben, 1999), and the details are shown as follows: (1) For hot stream  $i$ , its outlet temperature is denoted as  $T_{i, out}$  and inlet temperature is in the range of  $T_{i, min} \sim T_{i, max}$ .  $i$  can be divided into the high-temperature stream and low-temperature stream. In the corresponding high-temperature stream, the inlet and outlet temperatures should be  $T_{i, max}$  and  $T_{i, min}$ , respectively. In the low-temperature stream here, the inlet temperature is  $T_{i, min}$  and the outlet temperature is  $T_{i, out}$ . (2) Similarly, for cold stream  $j$ , its outlet temperature is referred to as  $T_{j, out}$  and the inlet temperature is in the range of  $T_{j, min} \sim T_{j, max}$ .  $j$  can be divided into the high-temperature stream and low-temperature stream. In the high-temperature stream of  $j$ , the inlet and outlet temperatures are  $T_{i, max}$  and  $T_j$ , respectively. For the low-temperature stream, its inlet temperature is  $T_{i, min}$  and outlet temperature is  $T_{j, max}$ . The fluctuation ranges of disturbed variables are determined according to the case of actual production, as shown in Table 4 (Zamora and Grossmann, 1997).  $\theta_N$  is set as the reference value.  $\Delta\theta^+$  and  $\Delta\theta^-$  are the expected deviations.

Table 4  
The reference value of disturbed variables and their expected deviation

disturbance variable	$\theta^N / ^\circ\text{C}$	$\Delta\theta^+ / ^\circ\text{C}$	$\Delta\theta^- / ^\circ\text{C}$
$\theta_1 = T_{C1}$	124.0	10	10
$\theta_2 = T_{H1}$	250.0	10	10
$\theta_3 = T_{C6}$	20.4	10	0
$\theta_4 = T_{C3}$	82.7	5	5



Table 5  
The datasheet of process streams obtained by the  
dividing principle

<b>New No.</b>	<b>Old No.</b>	$T_{IN}/^{\circ}C$	$T_{OUT}/^{\circ}C$	$q/(kJ/h)$
h1	H1 <sup>g</sup>	260	240	$9.72 \times 10^6$
h2	H1 <sup>d</sup>	240	40	$1.48 \times 10^8$
h3	H2	129.2	80.3	$5.86 \times 10^6$
h4	H3	122.3	121.8	$1.46 \times 10^8$
h5	H4	122.3	40	$5.58 \times 10^6$
h6	H5	107.3	40	$1.80 \times 10^6$
h7	H6	74.9	71	$1.46 \times 10^8$
h8	H7	73.3	64	$1.55 \times 10^7$
h9	H8	71	40	$2.22 \times 10^6$
h10	H9	64	40	$1.71 \times 10^6$
c1	C1 <sup>g</sup>	134	230	$4.99 \times 10^7$
c2	C1 <sup>d</sup>	114	134	$1.02 \times 10^7$
c3	C2	126.8	129.8	$1.46 \times 10^8$
c4	C3 <sup>g</sup>	86.7	127	$7.85 \times 10^6$
c5	C3 <sup>d</sup>	76.7	86.7	$1.76 \times 10^6$
c6	C4	107.3	110.9	$1.46 \times 10^8$
c7	C5	81.1	81.7	$1.85 \times 10^7$
c8	C6 <sup>g</sup>	30.4	75	$7.13 \times 10^6$
c9	C6 <sup>d</sup>	20.4	30.4	$1.52 \times 10^6$

The process streams with disturbed variables were divided according to the dividing principle, and the data are listed in Table 4. Besides, the segmented streams were treated as independent streams. Then, the data of all streams shown in Table 5 were imported into Aspen Energy Analyzer for designing flexible HEN. The final flexible HEN was obtained after merging the segmented streams and relaxing the energy load, and the detailed procedure is shown as follows: (1) the segmented process streams were merged firstly to obtain the network structure shown in Fig. 4. (2) the load loops were relaxed along with the direction of the downstream paths of the merged streams. (3) the flexible HEN was obtained by adjusting the parameters of each heat exchanger as shown in Fig. 5. Segmented streams merge was conducted based on the design results under the conditions of maximum heat recovery, and energy relaxation can reduce the number of heat exchangers in the whole process. Consequently, the final flexible HEN is optimized.

## 4 Results And Discussion

From Fig. S5, the strategy of original HEN employs the excessive number of sixteen utilities, which is able to be attributed to two reasons. On one hand, the situation, that a cooler crosses two pinch points and a heater crosses the process pinch point, violates the rule of forbidding across-pinch. On the other hand, there are three heaters below the pinch point and a cooler above the pinch point, which violates other rules of the pinch technology: no heating utility below the pinch point and no cooling utility above the pinch point are allowed. It can be concluded that the original HEN has massive energy expected to be recycled. Compared with the original HEN scenario, the traditional optimal HEN significantly improves the heat recovery capacity of the system and the number of heat exchangers decreases sharply. Although two process-process heat exchangers across the pinch point, there is no cooling utility above the pinch point and no heating utility below the pinch point (Fig. S6). In the flexible HEN (Fig. 4), 6 downstream paths were obtained in total, and the results of the flexibility analysis can be concluded as follows. (1) For the downstream path 1 and 2 with the disturbance variable  $\theta 1$  as the initial node, it is possible to stop  $\theta 1$  from affecting the controlled variables  $z 1$  and  $z 2$  by adjusting the usage of the heating utility, since these two paths pass through the node  $HE 1$ . (2) For the downstream path 3 and 6, there is only one node of heat exchanger between the disturbance variables and the control variables, and the disturbance variables have a direct effect on the controlled ones. Some differences between path 3 and path 6 are observed. Both variables on the downstream path 3 locate in different streams. In contrast, those variables in the downstream path 6 are on the same stream. (3) For the downstream path 4 and 5, there are many nodes on each of them and no node of utilities, so the fluctuation of the disturbance variable will be transferred to the controlled variable along with the downstream path direction. In summary, the controlled variables will be affected as long as the disturbance variables of the downstream path 3–6 fluctuate, so it is difficult for this regular optimized HEN to meet the flexibility requirements in the practical scenario.

Table 6  
Parameters of original HEN and flexible HEN

Parameter	Unit	Original HEN	Flexible HEN	Reduction/%
Heating utilities	kJ/h	$1.819 \times 10^8$	$1.508 \times 10^8$	17.12
Cooling utilitie	kJ/h	$2.756 \times 10^8$	$2.432 \times 10^8$	11.76
Number of shell	-	55	37	32.73
Total area	m <sup>2</sup>	$2.066 \times 10^4$	$1.191 \times 10^4$	42.35
Capital cost	Cost	$5.099 \times 10^6$	$3.080 \times 10^6$	39.60
Operating cost	Cost/s	0.1053	0.0931	11.53
Total cost	Cost/s	0.1573	0.1246	20.82

Table 6 shows the comparison of major parameters from the original HEN and the flexible HEN. The total energy consumption of the flexible HEN is reduced by 13.89% compared with that of the original HEN, indicating the prominent energy recycling efficiency. Moreover, the capital cost, operating cost and total cost in the flexible HEN scenario are individually reduced by 39.60%, 11.53%, and 20.82% as well.

Table 7  
Parameters of traditional optimal HEN and flexible HEN

Parameter	Unit	Regular optimized HEN	Flexible HEN	Reduction/%
Heating utilities	kJ/h	$1.468 \times 10^8$	$1.508 \times 10^8$	-2.72
Cooling utilitie	kJ/h	$2.405 \times 10^8$	$2.432 \times 10^8$	-1.13
Number of shell		32	37	-15.63
Total area	m <sup>2</sup>	9209	$1.191 \times 10^4$	-29.35
Capital cost	Cost	$2.445 \times 10^6$	$3.080 \times 10^6$	-25.97
Operating cost	Cost/s	0.0947	0.0931	1.69
Total cost	Cost/s	0.1197	0.1246	-4.08

Table 7 shows the comparison of major parameters from the traditional optimal HEN and the flexible HEN. The cooling and heating utilities in the flexible HEN scenario are increased slightly with the increment of 2.72% and 1.13%, compared with the regular optimized HEN. And the number and total area of heat exchangers are increased significantly by 15.63% and 29.35%. This illustrates the essence of the HEN flexibility design in our work is to increase the area margin and appropriately set up alternative heat exchangers. However, the operating cost in the flexible HEN strategy is reduced by 1.69%, implying that increase the HEN flexibility can reduce the frequency and difficulty of operations. Although the total cost merely has slight increase (4.08%), the dramatic enhancement on the HEN flexibility guarantees the stability and safety of production. The flexible HEN can lengthen the cleaning cycle and the service life of heat exchangers so that the operating cost will be reduced. In the long term, the prolonged lifespan of heat exchangers will offset partial capital cost, which is caused by extra heat exchangers and additional area margin.

## 5 Conclusion

In this study, the overall HEN of syngas-to-methanol was designed and optimized based on pinch technology under the stable operating conditions. The flexibility analysis of traditional optimal HEN was implemented by using the downstream paths method. 6 downstream paths between disturbance variables and control variables are determined by the modified procedure with the node adjacency matrix. The results show that 2/3 downstream paths cannot meet flexibility requirements, indicating the optimized design of the HEN under stable operating conditions is not flexible enough to resist disturbances in the actual production. The flexible HEN is realized under the condition of maximum heat recovery by the method of dividing and subsequent merging. Although the total cost of the flexible HEN is 4.08% higher than that of traditional optimal HEN, the flexible HEN has enough area margin and extra alternative heat exchangers to avoid the production disturbances and maintain its stability and safety.

## Abbreviations

$\Delta T_{\min}$

minimum heat transfer temperature difference

$Q_{H, \min}$

minimum hot utility

$Q_{C, \min}$

minimum cold utility

$\theta$

disturbed variable

$z$

controlled variable

$(i, j) \& (p, q)$

elements of a node adjacency matrix

$S$

a set of nodes

$T_{i, \text{out}}$

outlet temperature of hot stream

$T_{i, \text{max}}$

inlet temperature of hot stream

$T_{j, \text{out}}$

outlet temperature of cold stream

$T_{j, \text{max}}$

inlet temperature of cold stream

$\theta_N$

reference value

$\Delta\theta^+ \& \Delta\theta^-$

expected deviations

## Declarations

### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

### Authors' contributions

**Jinchang Liu:** Conceptualization, Methodology, Formal analysis, Experiment, Writing - Review & Editing; **Pingping Zhang:** Writing - Original Draft; **Qiang Xie:** Writing - Review & Editing, Supervision; **Dingcheng Liang:** Writing - Review & Editing; **Lei Bai:** Formal analysis, Writing - Review & Editing.

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## References

1. Li Z, Gao D, Chang L, Liu P, Pistikopoulos EN (2010) Coal-derived methanol for hydrogen vehicles in China: Energy, environment, and economic analysis for distributed reforming. *Chem Eng Res Des* 88:73–80. DOI:10.1016/j.cherd.2009.07.003
2. Riaz A, Zahedi G, Klemeš JJ (2013) A review of cleaner production methods for the manufacture of methanol. *J Clean Prod* 57:19–37. DOI:10.1016/j.jclepro.2013.06.017
3. Galadima A, Muraza O (2015) From synthesis gas production to methanol synthesis and potential upgrade to gasoline range hydrocarbons: A review. *J Nat Gas Sci Eng* 25:303–316. DOI:10.1016/j.jngse.2015.05.012

4. Gao D, Qiu X, Zhang Y, Liu P (2018) Life cycle analysis of coal based methanol-to-olefins processes in China. *Comput Chem Eng* 109, 112–118. DOI:10.1016/j.compchemeng.2017.11.001
5. Bessa LCBA, Batista FRM, Meirelles AJA (2012) Double-effect integration of multicomponent alcoholic distillation columns. *Energy* 45:603–612. DOI:10.1016/j.energy.2012.07.038
6. Cui C, Sun J, Li X (2017) A hybrid design combining double-effect thermal integration and heat pump to the methanol distillation process for improving energy efficiency. *Chem Eng Process* 119:81–92. DOI:10.1016/j.cep.2017.06.003
7. Rashid SRA, Ibrahim UK, Alauddin SM (2011) Retrofit Design of Heat Exchanger Network (HEN) on Synthesis and Purification Unit of Methanol Plant. 2011 IEEE Colloquium on Humanities, Science and Engineering Research (CHUSER 2011) Dec 5–6 2011, 33–36. DOI: 10.1109/CHUSER.2011.6163746
8. Sun J, Wang F, Ma T, Gao H, Wu P, Liu L (2012) Energy and exergy analysis of a five-column methanol distillation scheme. *Energy* 45:696–703. DOI:10.1016/j.energy.2012.07.022
9. Chien IL, Teng Y-P, Huang H-P, Tang YT (2005) Design and control of an ethyl acetate process: coupled reactor/column configuration. *J Process Control* 15:435–449. DOI:10.1016/j.jprocont.2004.07.003
10. Douglas AP, Hoadley AFA (2006) A process integration approach to the design of the two- and three-column methanol distillation schemes. *Appl Therm Eng* 26:338–349. DOI:10.1016/j.applthermaleng.2005.07.001
11. Kang L, Liu Y (2019) Synthesis of flexible heat exchanger networks: A review. *Chin J Chem Eng* 27:1485–1497. DOI:10.1016/j.cjche.2018.09.015
12. Payet L, Thery Hétreux R, Hétreux G, Bourgeois F, Floquet P (2018) Flexibility Assessment of Heat Exchanger Networks: From a Thorough Data Extraction to Robustness Evaluation. *Chem Eng Res Des* 131, 571–583. DOI:10.1016/j.cherd.2017.11.036
13. ZHU J, HAN Z, RAO M, CHUANG KT (1996) Identification of heat load loops and downstream paths in heat exchanger networks. *The Canadian Journal of Chemical Engineering* 74:876–882. DOI:10.1002/cjce.5450740609
14. LINNHOFF B, HINDMARSH E (1983) The pinch design method for heat exchanger networks. *Chem Eng Sci* 38:745–763. DOI:10.1016/0009-2509(83)80185-7
15. Binosi D, Papavassiliou J (2009) Pinch technique: Theory and applications. *Phys Rep* 479:1–152. DOI:10.1016/j.physrep.2009.05.001
16. Tan YL, Ng DKS, El-Halwagi MM, Foo DCY, Samyudia Y (2014) Floating pinch method for utility targeting in heat exchanger network (HEN). *Chem Eng Res Des* 92:119–126. DOI:10.1016/j.cherd.2013.06.029
17. SWANEY RE, GROSSMANN IE (1985) An index for operational flexibility in chemical process design. Part I Formulation and theory. *AIChE J* 31:621–630. DOI:10.1002/aic.690310412
18. Grossmann IE, Kravanja Z (1995) Mixed-integer nonlinear programming techniques for process systems engineering. *Comput Chem Eng* 19:189–204. DOI:10.1016/0098-1354(95)00072-A
19. Linnhoff B, Kotjabasakis E (1986) Downstream paths for operable process design. *Chem Eng Prog* 5:263–281
20. Zhihong L, Ben H (1999) Synthesis of flexible heat exchanger networks with stream no-splitting(I) based on ranges of stream supply temperatures. *Journal of Chemical Industry Engineering (China)* 50:317–325. DOI:10.3321/j.issn:0438-1157.1999.03.005
21. Zamora JM, Grossmann IE (1997) A comprehensive global optimization approach for the synthesis of heat exchanger networks with no stream splits. *Comput Chem Eng* 21:S65–S70. DOI:10.1016/S0098-1354(97)87480-7

## Figures



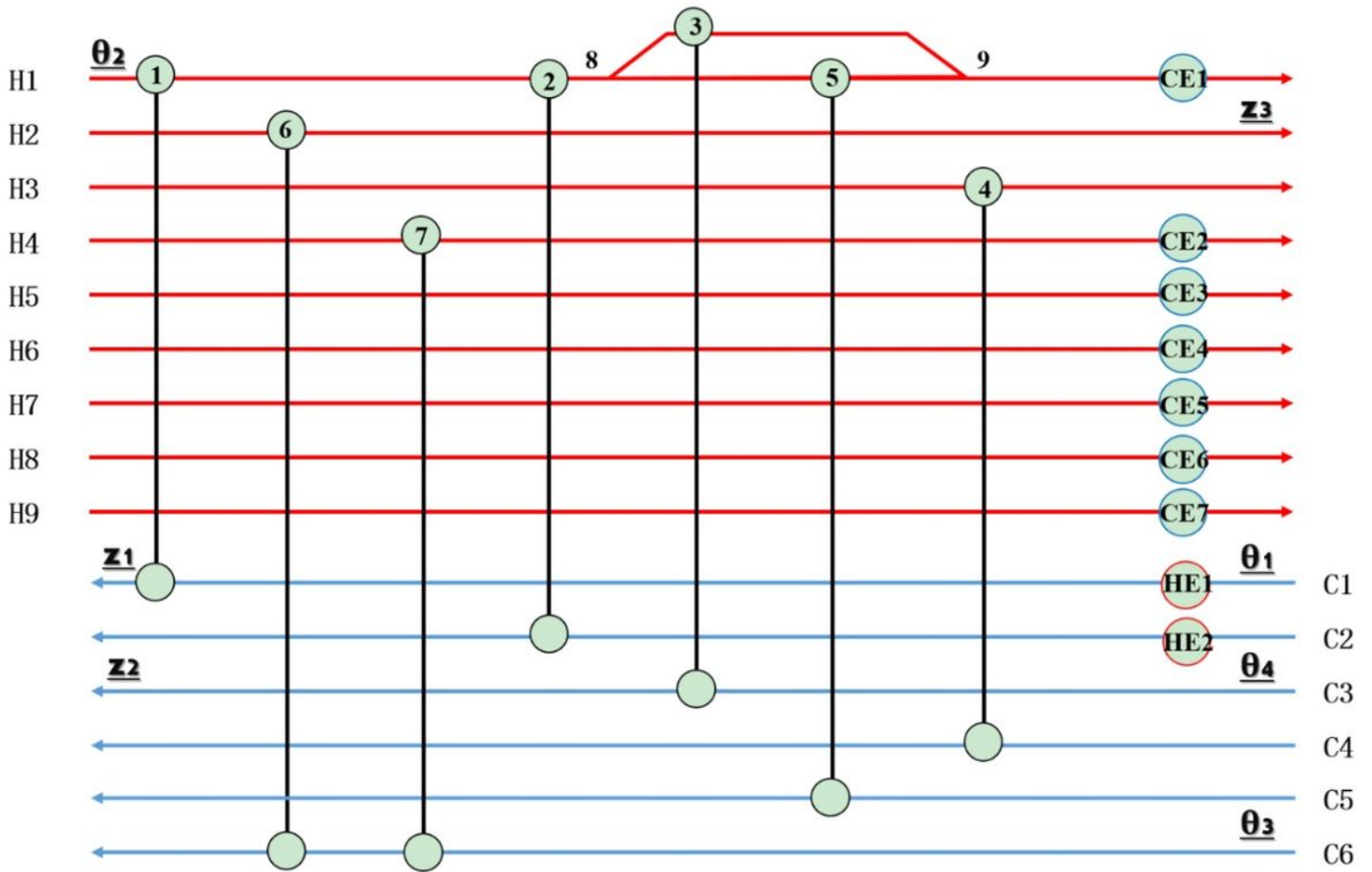


Figure 2

Disturbed variables and controlled variables in the grid diagram of traditional optimal HEN

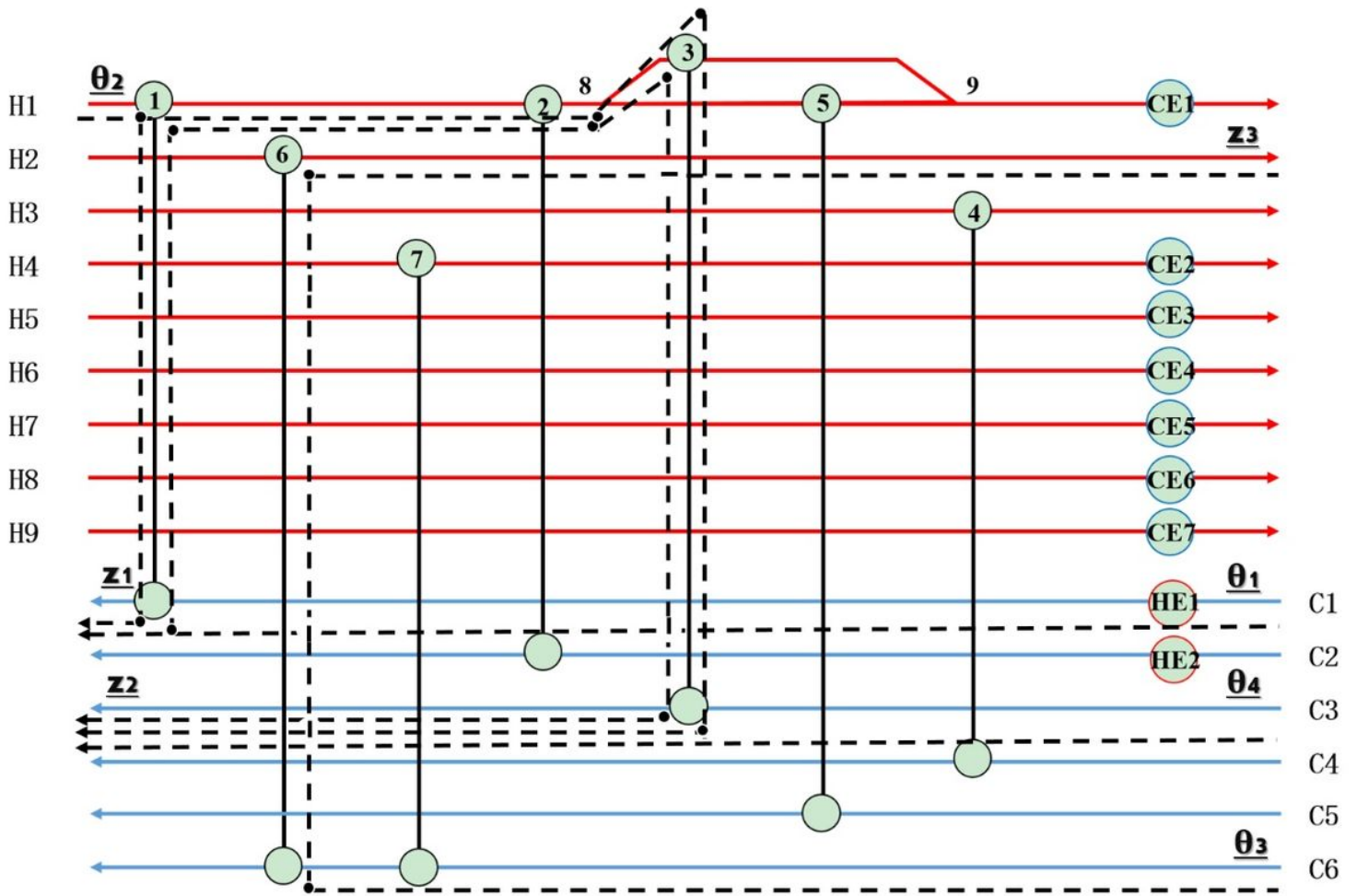
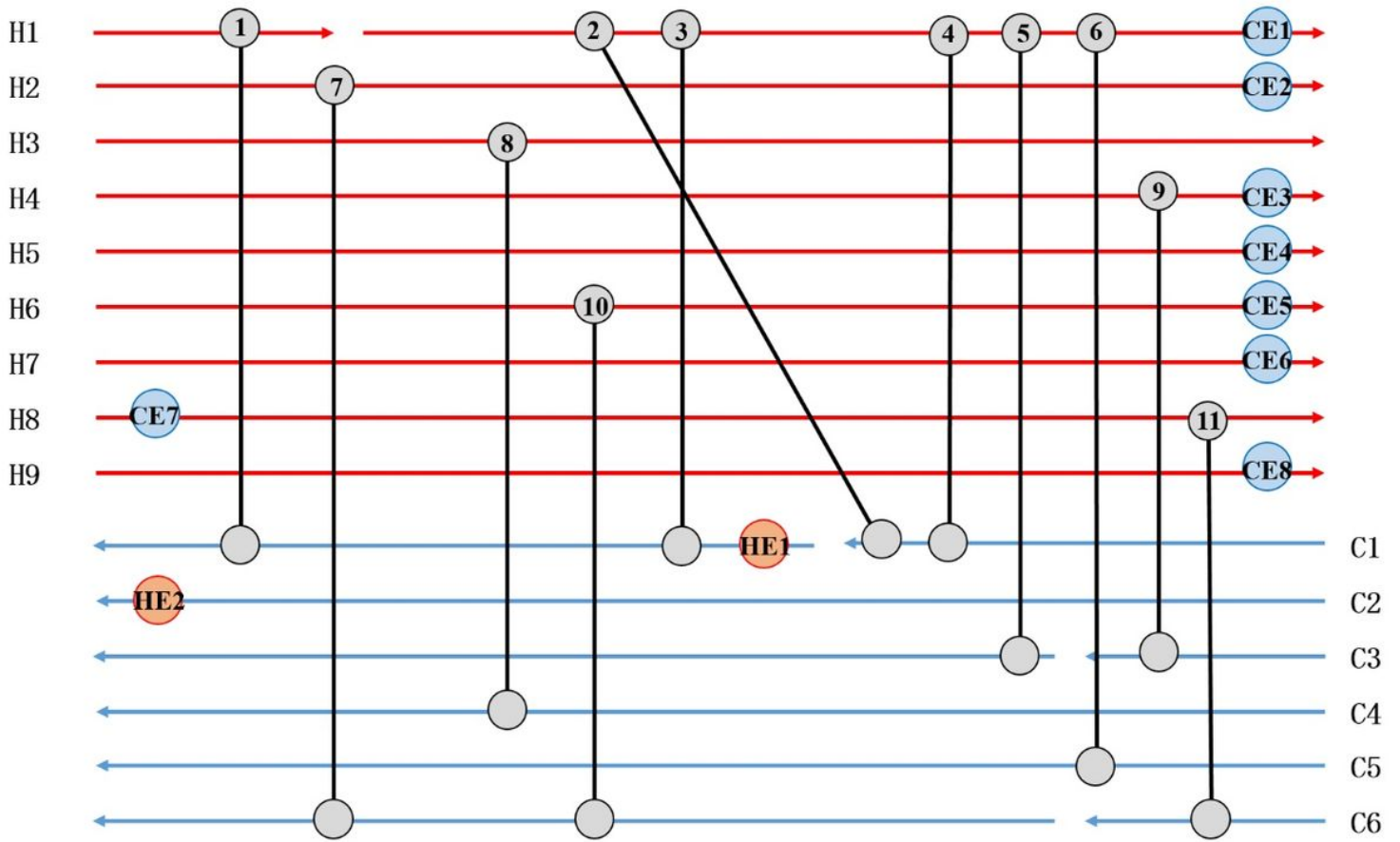


Figure 3

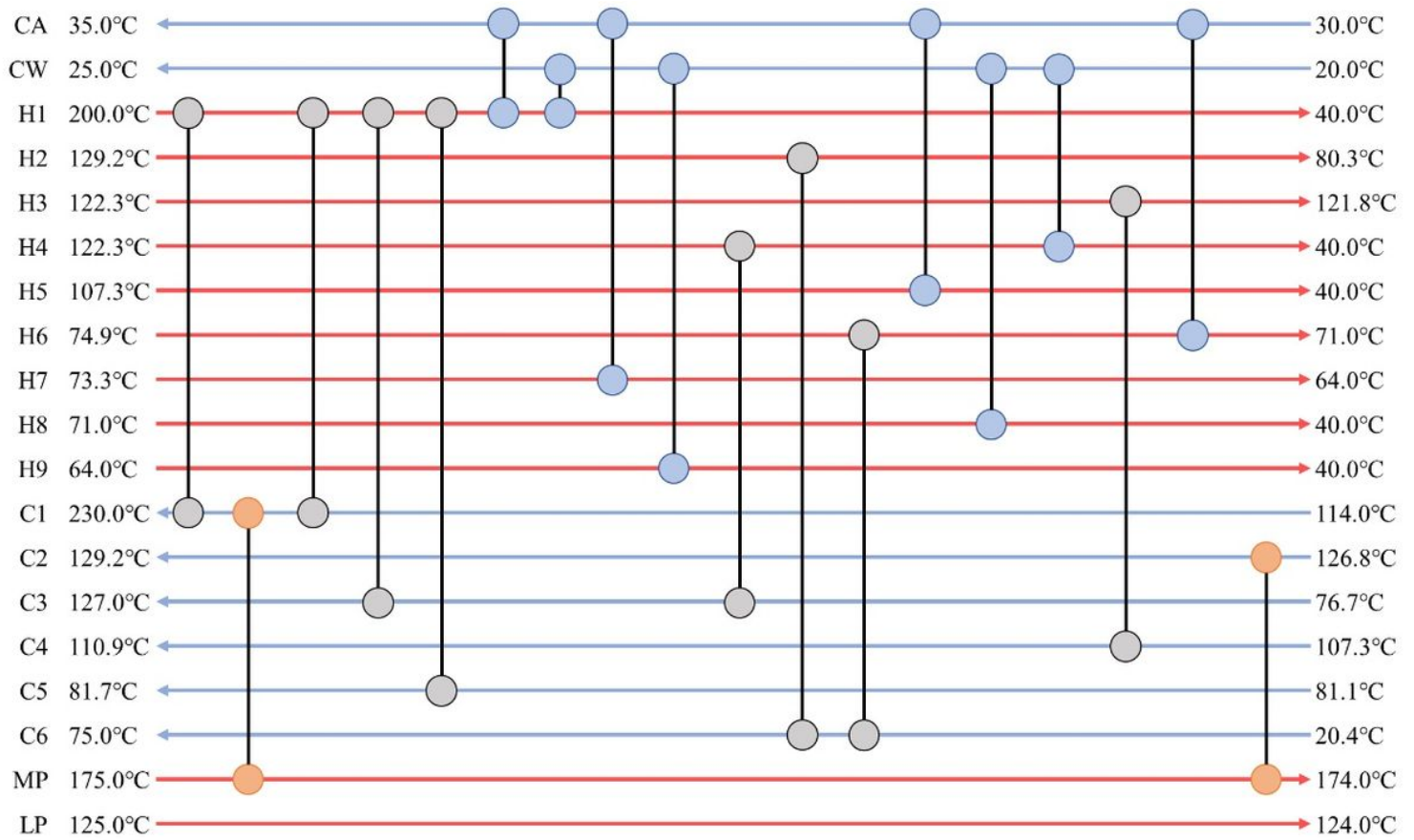
Downstream paths between disturbed variables and controlled variables





**Figure 4**

The grid diagram of flexible HEN with merging the segmented streams



Note: CA-Cooling Air, CW-Cooling Water, MP-Middle Pressure Steam, LP-Low Pressure Steam

**Figure 5**

Final flexible HEN

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