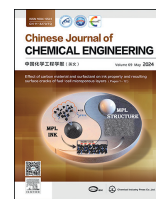




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Full Length Article

Process synthesis for the separation of coal-to-ethanol products

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ABSTRACT

The coal-to-ethanol process, as the clean coal utilization, faces challenges from the energy-intensive distillation that separates multi-component effluents for pure ethanol. Referring to at least eight columns, the synthesis of the ethanol distillation system is impracticable for exhaustive comparison and difficult for conventional superstructure-based optimization as rigorous models are used. This work adopts a superstructure-based framework, which combines the strategy that adaptively selects branches of the state-equipment network and the parallel stochastic algorithm for process synthesis. High-performance computing significantly reduces time consumption, and the adaptive strategy substantially lowers the complexity of the superstructure model. Moreover, parallel computing, elite search, population redistribution, and retention strategies for irrelevant parameters are used to improve the optimization efficiency further. The optimization terminates after 3000 generations, providing a flow-sheet solution that applies two non-sharp splitting options in its distillation sequence. As a result, the 59-dimension superstructure-based optimization was solved efficiently via a differential evolution algorithm, and a high-quality solution with a 28.34% lower total annual cost than the benchmark was obtained. Meanwhile, the solution of the superstructure-based optimization is comparable to that obtained by optimizing a single specific configuration one by one. It indicates that the superstructure-based optimization that combines the adaptive strategy can be a promising approach to handling the process synthesis of large-scale and complex chemical processes.

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1. Introduction

Ethanol is an essential feedstock of the chemical industry and energy supply chain [1–3]. The coal-to-ethanol (CTE) technology provides not only ethanol products but also a route for the clean and low-carbon utilization of coal. Especially in China, where coal dominates the energy share and results in intensive carbon emission [4], the CTE route is of great significance [5,6]. Among the categories of CTE, the route from dimethyl ether to ethanol (DMTE) via carbonylation and hydrogenation is highly attractive because it does not need precious metal catalysts and directly produces anhydrous ethanol [7,8].

The effluents from the reactor in the DMTE route are a mixture of ethanol, methanol, ethyl acetate, and methyl acetate. In industrial practice, the process combinedly uses conventional and swing-pressure distillation due to the presence of azeotropes [9] shown in

Table 1. Swing-pressure configurations introduce extra columns, resulting in a large-scale separation system with at least eight columns. The distillation system involves intensive energy consumption, which may hinder the DMTE technology from being a promising route for coal's clean and low-carbon utilization [10]. Therefore, finding competitive configurations becomes a necessity.

Designing energy-efficient and economical configurations for large-scale separation processes remains a significant challenge. Considering the significance of ethanol in this system, the prevalent industry approach is to prioritize its separation, which means purifying ethanol at the beginning of the product separation [11]. Nevertheless, the presence of azeotropes in the process necessitates the repeated evaporation and condensation of methanol, resulting in additional energy demand. In previous work, four other alternative separation strategies were proposed by the empirically based means of focusing on methanol and ethyl acetate. After optimizing these configurations with a single-process stochastic algorithm, two of the distillation sequences stood out, namely splitting methanol and ethyl acetate with ethyl acetate priority [11]. However, it does not encompass all potential distillation sequences, and there could be the probability of ignoring some competitive

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Table 1

Azeotropes in the separation unit of coal-to-ethanol products at 100 kPa

Azeotropes (mass fraction)	Boiling point/°C
Methyl acetate (0.1762) -Methanol (0.8238)	53.27
Methyl acetate (0.9981) -Ethanol (0.0019)	56.68
Ethyl acetate (0.5377) -Methanol (0.4623)	61.91
Ethyl acetate (0.7043) -Ethanol (0.2957)	71.44

alternatives. In addition, distillation sequences usually significantly affect the energy and economic performance of the separation [12]. Therefore, it is essential to perform comprehensive, considered all potential distillation sequence comparisons, including sharp and non-sharp splits.

Compared to parameter optimization, the screen of distillation sequences is more complicated. The diversified sequences lead to a tremendous amount of candidates, which could hardly be exhaustively compared. As a countermeasure, superstructure-based optimization, which optimizes distillation sequences and parameters simultaneously, has recently drawn increasing attention as an efficient process synthesis tool [13].

As an abstract model that encapsulates various alternative processes based on specific rules, superstructures are widely used to form the total set of all process candidates [13]. Among various superstructures, state-task network and state-equipment network (SEN) are the primary ones. They identify candidate configurations by activating specific branch nodes and selecting partial subnetworks [14,15]. This feature leads to the convenience of transforming superstructure models into pure mathematic forms, which introduces integer variables (usually binary variables) to determine the branch selection [16]. Solving the extended mixed integer nonlinear programming (MINLP) could provide competitive configurations. Currently, the superstructure-based optimization has been a mature route for process synthesis and performs considerably for process synthesis using simplified models (*e.g.*, short-cut columns) [17–19], but some challenges remain as rigorous models were used. Since the equations and constraints related to inactivated subprocesses are included in the MINLP problem, the solving procedure encounters a sparse matrix with many zero values, raising difficulties for the gradient-based algorithms [14,20]. More challenges are caused by the increasing problem dimensions. Although stochastic algorithms are more robust for nonconvex problems, degradation of solution quality and search efficiency are still observed when dealing with large-scale process synthesis problems. However, the optimum of rigorous models is still necessary because the error resulting from model simplification is too large and unreliable for industry practice. From this perspective, further improvements in the superstructure-based process synthesis are still engaged.

The strategy that adaptively selects branches and nodes of the SEN according to some logical disjunction could be helpful in reducing the complexity of process synthesis [21]. By embedding logical disjunction into specific modules, the simulation-based optimization could also implement this strategy for process synthesis [22]. Based on the logical correlations between the downstream separation task and the calculation results of upstream modules, the explicit discrete variables representing branches and nodes could be transformed into implicit dependent variables figured by disjunction. Including fewer discrete variables in solution vectors, the complexity of the superstructure model significantly reduces. The advantages of this method have been demonstrated in some industrial practices, for instance, the benzene/cyclohexane/cyclohexane separation [23] and the close-boiling organosilicon monomers separation [24]. However, all those reported cases have relatively small dimensions lower than 30, which is not enough to validate the feasibility of the improved method in larger-scale process systems yet.

In this study, a superstructure-based optimization framework was applied for process synthesis to find competitive configurations of ethanol distillation. High-performance computing technology was used to accelerate the optimization, and the adaptive strategy was used to lower the complexity of superstructure models. Moreover, considering the performance of stochastic algorithms will degrade as the size of the optimization problem increases [25]; some slight modifications were also used to improve the solution quality and iterative efficiency, *e.g.*, the inactivated variable retention strategy, elite-search, and population redistribution. Some reported CTE-optimized configurations were introduced as the comparison [11].

In summary, this paper further develops the energy-efficient and economic ethanol distillation process for the CTE route via the superstructure-based process synthesis, providing a promising solution for the clean utilization of coal. Meanwhile, the comparison between the results of superstructure-based optimization and those of single-process optimization could also help validate the feasibility of the adaptive branch selection strategy in larger-scale process systems. In addition, this study provides a systematic framework for large-scale process synthesis, which extends the applications of classical optimization methods. The paper is organized as follows. Section 2 introduces the process optimization and synthesis methods. In Section 3, the CTE product separation process is detailed. The results and discussion are provided in Section 4. Finally, Section 5 concludes.

2. Optimization and Modeling Methods

The simultaneous optimization approach using the stochastic algorithm and parallel computing has successfully addressed the rigorous simulation optimization of CTE single process of 42 dimensions [11]. The CTE single-process rigorous optimization belongs to the single-process optimization, which constructs separate rigorous process simulations based on different separation strategies, *i.e.*, different distillation sequences. In contrast, the rigorous model-based process synthesis belongs to the superstructure optimization, which encompasses all possible distillation sequences and is a merger of the single-process optimization. The superstructures are implemented through a splitter embedded in the simulation software and the self-written FORTRAN programming language for specific process selection. The dimensionality and complexity of the rigorous model-based process synthesis are greater. Moreover, the increasing sparsity of equations caused by inactivated modules leads to more significant challenges. Therefore, some modifications are made to the algorithm and model construction.

2.1. Retention of the inactivated variables

During the iteration of the superstructure-based optimization, some variables are not activated as the corresponding modules or branches are not selected. The dimensionality of the variables is kept constant to ensure the robust implementation of optimization algorithms but raises the difficulty of handling the inactivated variables.

A stochastic algorithm iterates its population via selecting advantageous individuals from the original population and the generated one. As the selection criterion is usually “<” or “>”, the individuals with the same activated variables and different inactivated ones will have the same fitness function values, promoting the new individual to replace the old one. In other words, individuals could move randomly without any confirmation along the inactivated dimensions, which may lower the search efficiency or cause unexpected influences on the optimization. To guarantee the robustness of the optimization, a retention strategy of the

inactivated variables, which makes the old individual only accept variables from the activated dimensions of the new dominant individual, is introduced to the crossover operation. Considering the inactivated dimensions of individuals are known before the selection operation, the retention strategy could be implemented as follows:

- (1) Predetermine the set of indexes of the activated variables in each column and all the possible sequences of this process synthesis.
- (2) Establish the mapping of sequence number to the index set of relevant variables and the mapping of code to sequence number, where the encoding method is based on the state of the splitter.
- (3) Determine the sequence number of the specific subprocess based on the obtained code.
- (4) Preserve the variable values of the corresponding variable index set according to the sequence number during the selection operation. The schematic diagram is shown in Fig. 1, where Fitness indicates the total annual cost (TAC) obtained, which is based on Luyben's work [26], and the calculation method is shown in Supplementary Material S2; num represents the sequence number; $\mathbf{X}$ is the target vector, and $\mathbf{U}$ is the trial vector; mapObj indicates the index of the variable to maintain.

2.2. Elite search and population redistribution

The superstructure is the encapsulation of each alternative process; the dimensionality and the range of the variables are the unions of all subprocesses; therefore, the search efficiency is significantly worse than single-process optimization. The elite-search strategy is implemented to expedite the optimization process, which involves narrowing the search range based on the contemporary optimal individuals. As exemplified in Fig. 2 a new search range is established based on optimal individuals from the current generation. Subsequently, an assessment is performed to determine whether the new range is within the original feasible domain. In the event of compliance, the original feasible domain is replaced; conversely, the boundary values of the initial feasible domain are used. However, it will result in the risk of reducing the population diversity. As a countermeasure, population redistribution is introduced [27]. After a given number of generations, a portion of the worst individuals reinitialize, and the search range

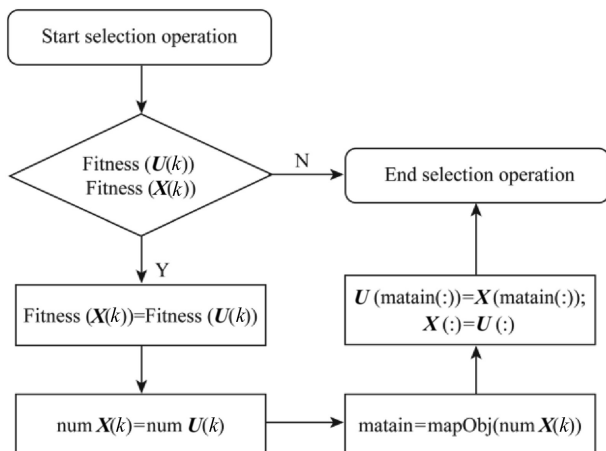


Fig. 1. Schematic diagram of retention strategies for inactivated parameters.

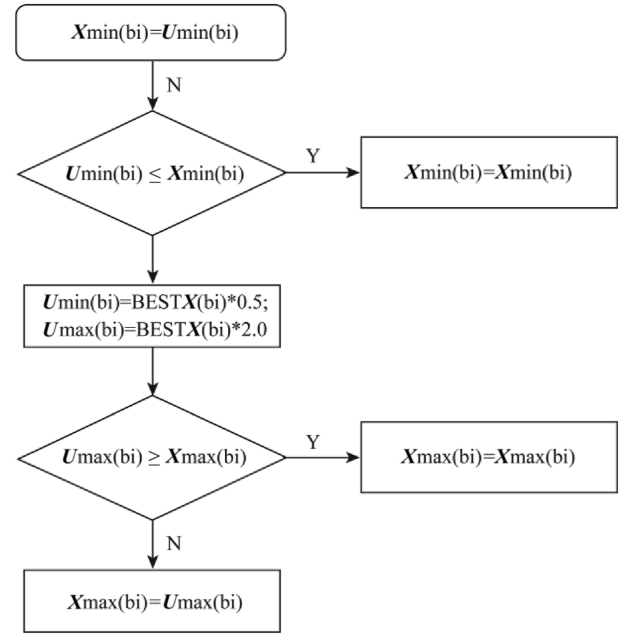


Fig. 2. Schematic diagram of a strategy for elite-search.

is reset to the magnitude before the optimization begins. Figs. 2 and 3 show the elite-search and population redistribution [28] strategies.

2.3. Automatic generation of the heat exchange network

The improved simultaneous optimization approach combined with parallel computation is shown in Fig. 4 (adapted from Ref. [29]), and the improved strategy has been described in detail earlier. In addition, the simultaneous optimization approach depends on the sorting and matching of cold and hot streams for heat integration structure generation based on the minimum heat

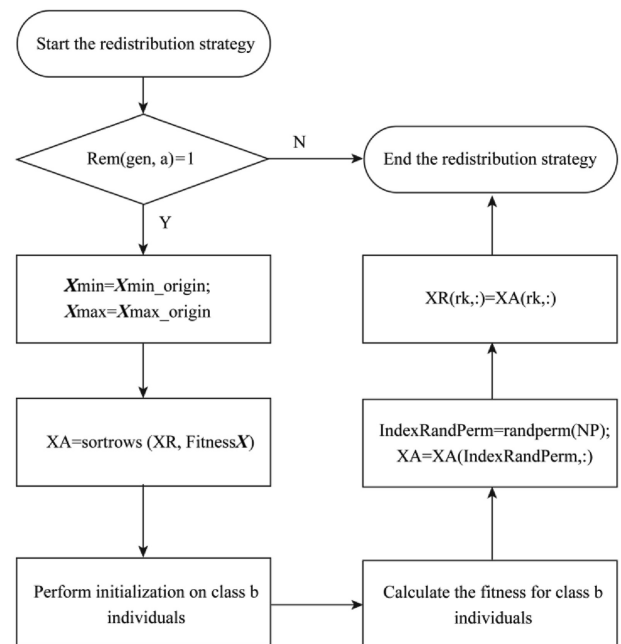


Fig. 3. Schematic diagram of a strategy for population redistribution.

Table 2
ABCD four-component distillation and swing-pressure distillation combinatorial problem

Component	Mass fraction/%	NBP/°C	Number
Argon	0.1	−185.9	Light components
Dimethyl ether	0.5	−29.5	
Methyl acetate–methanol azeotrope		53.27	A
Methyl acetate [⊙]	3.4	56.8	
Ethyl acetate–methanol azeotrope		61.91	B
Ethyl acetate [⊙]	1.4	77.2	
Methanol [⊙]	41.5	64.8	C
Ethanol [⊙]	51.7	78.3	D
N-propanol	0.1	95.8	Heavy components
Water	1.0	100	

Note: Feed temperature, pressure, and mass flow rate are 68.7 °C, 301.25 kPa, and 122104 kg·h^{−1}, respectively.

[⊙] Indicates the end products.

eliminated in the light-end removal column (T1), whereas the heavy component (N-propanol, water, and other trace impurities heavier than D) is collected from the bottom of the ethanol recovery column (T7). Since the Normal Boiling Point (NBP) of light-ends and heavy-ends are pretty different from other components, the top distillate rate of the light-end removal column (T1) and the bottoms-to-feed ratio of the ethanol recovery column (T7) are approximately constant and could be fixed for simplicity.

Meanwhile, a partial condenser with a distillate vapor fraction of 1.0% (mol) is set up in a methyl acetate atmospheric column to remove the residual non-condensable products. Here, CY

structured packing of 1000 USD·m^{−3} is used as the column internals, and the pressure drop and the height equivalent of each theoretical plate are assumed to be 0.1 kPa and 0.2 m, respectively. Moreover, a pressure drop of 10 kPa and a subcooling degree of 3 °C is set for the condenser.

The superstructure of CTE product separation is shown in Fig. 6, which is constructed based on SEN. In contrast to the conventional eight- and nine-column processes, it comprises 11 columns. Six splitters exist in this superstructure, whose possible states are shown in Table 3, where the number in the bracket represents the splitter flow direction, and the invalid diversion is represented by 0.

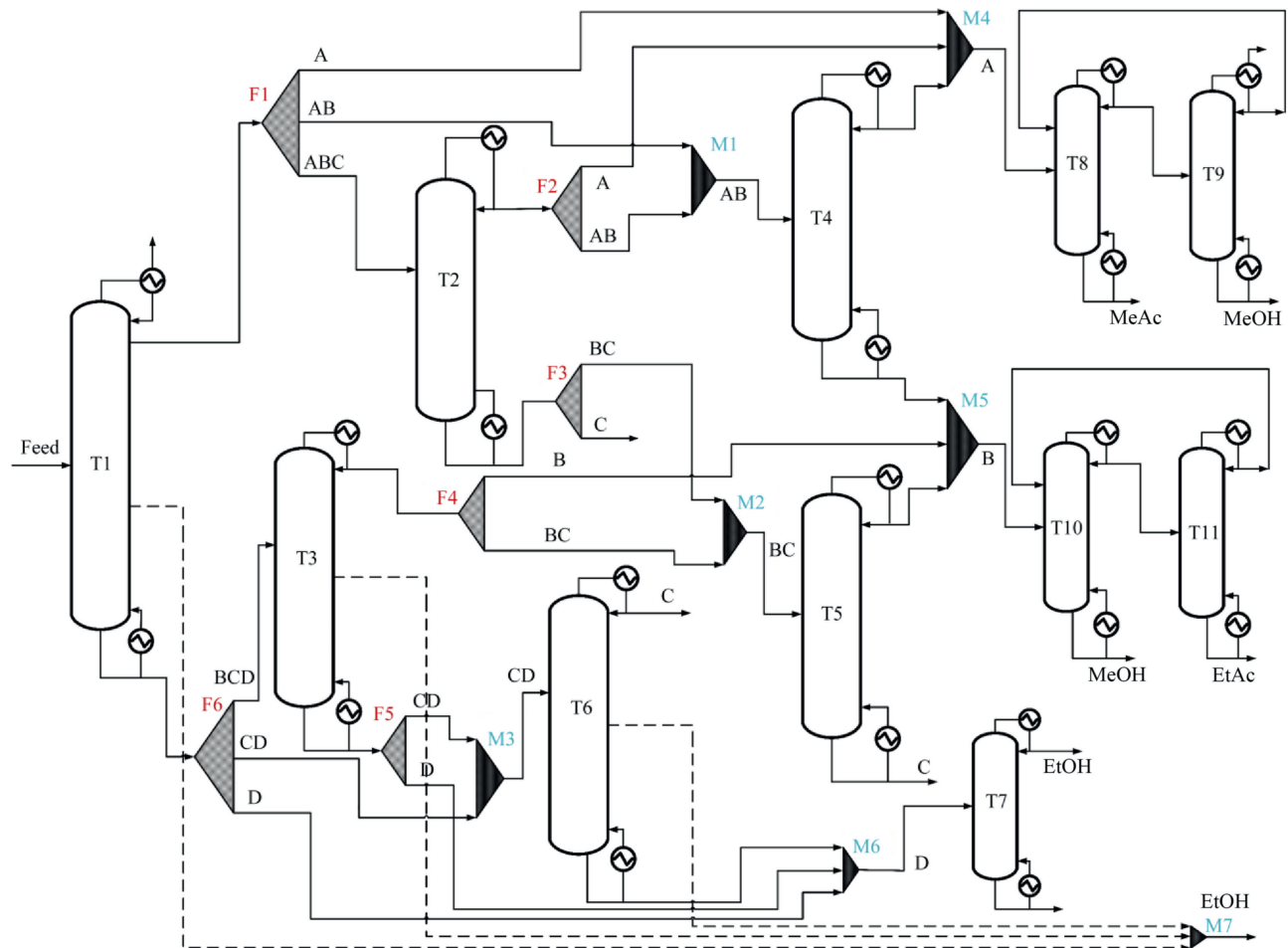


Fig. 6. The superstructure of coal-to-ethanol product separation.

Table 3

Selector module status log table

F1	F2	F3	F4	F5	F6
A (1)	A (1)	C (1)	B (1)	D (1)	D (1)
AB (2)	AB (2)	BC (2)	BC (2)	CD (2)	CD (2)
ABC (3)	– (0)	– (0)	– (0)	– (0)	BCD (3)

In addition, the conditions for its logical judgment are illustrated in detail in Table 4. Considering the presence of azeotropic compositions and the constant changes in operating pressure, it is infeasible to judge the boundary between the two azeotropic compositions and the pure component exceedingly accurately. Consequently, the judgment conditions for F1 and F4 are empirical values obtained by debugging on the optimization results reported in Ref. [11]. For F2, F5, and F6, the content of more than 1 represents the presence and that of less than 1 means the absence; if greater than 0.999, it means that the product is a qualified withdrawal without the need for subsequent separation in F3. The side draw of the system is for the ethanol, which means that the bottom of the side draw column is incapable of collecting a component with a lower volatility point than the ethanol, and the judgment of side draw will be explained in detail later. Table 5 illustrates 22 alternative configurations without considering column merging, where the sequential arrangement of numbers can be associated with subprocess sequence numbers. In addition, the process diagram of sequence ‘1’ is shown as an example in Fig. 7.

According to the manufacturer's request for quotation, the ethanol product's refining is divided into two parts: vapor side draw and distillate. This superstructure includes three candidates for the side draw of ethanol: T1, T3, and T6, but only one side draw can be selected. The probability value was fixed at 0.15, considering that A cannot be obtained directly from T1, and three configurations of side draw in T1 are available. In addition, the activation status of equipment is considered first and foremost when the side draw of T1 is absent. The result is straightforward when only one is activated; if both are activated, giving both the same probability of competition. A schematic diagram of the processing algorithm for side stream is shown in Fig. 8.

Design variables include total stages (NT), feed stage (NF), distillate or bottoms-to-feed ratio (D:F or B:F), reflux ratio (RR), operating pressure (*P*), recirculating product feed stage (NR), side draw stage (NS), and side draw to feed ratio (S:F). It raises an optimization problem with 59 variables. The detailed variables declaration of the superstructure and relationship between degrees of freedom and variables can be found in Supplementary Material S1.

The constraint contains equation constraints and inequality constraints. The commercial solvers automatically satisfy the

Table 5

The mapping of code to sequence number

Sequence	Code	Sequence	Code
1	200002	12	200213
2	312001	13	200223
3	321001	14	312123
4	100123	15	312213
5	100213	16	312223
6	322001	17	321123
7	100223	18	321213
8	312002	19	321223
9	322002	20	322123
10	200123	21	322213
11	321002	22	322223

former when the process simulation converges. A programming environment can handle the latter or be embedded in the commercial solvers as design specifications [30]. Nevertheless, due to loops in the system, the iterative design specification will exponentially increase computational complexity with a significant risk of non-convergence. The product-quality requirement represents the most considerable inequality constraint, which is guaranteed by the penalty function method within the MATLAB environment [32]. More details can be found in Table 6 and Fig. 9, where the penalty factor is denoted by α .

4. Results and Discussion

The optimization of process synthesis is implemented on 12 parallel threads of an Intel (R) Xeon (R) W-2295 (3.00 GHz) CPU. The optimization progress curve of TAC is given in Fig. 10. Figs. 11 and 12 show the process configuration diagram of the optimal sequence and the heat integration structure diagram of the optimal solution for the superstructure, respectively, and the temperature-enthalpy graph is shown in Fig. 13.

After 3000 generations of differential evolution, the stand-out sequence is sequence ‘13’, with the identification code 200223. The description of the identification code is provided in Table 5. It can be observed in Fig. 10 that the optimization process tends to converge after approximately 3000 generations. Furthermore, the difference between the average and optimal values diminishes over time. It ultimately reaches an absolute difference of less than 2%, a remarkable result for a large-scale optimization problem.

In contrast to the five reported strategies, the process is characterized by a 10-column configuration, as shown in Fig. 11. Sequence ‘13’ uses non-sharp splitting twice: first for ethyl acetate in T1 and then for methanol in T3. Its distillation brings out the remaining ethyl acetate and part of the methanol product, and the bottom of the column involves the remaining methanol, ethanol,

Table 4

Logical judgment condition of splitter

Splitter	Logical judgment condition	Direction of split
F1	Mass flow of the stream to splitter F1 (MF1)	MF1 ≤ 5050 5050 ≤ MF1 ≤ 23000 Other A AB ABC
F2	Mole flow of ethyl acetate in the stream to splitter F2 (MF2)	MF2 ≥ 1 Other AB A
F3	Mass fraction of methanol in the stream to splitter F3 (MF3)	MF3 ≥ 0.999 Other C BC
F4	Mole flow of methanol in the stream to splitter F4 (MF4)	MF4 ≥ 300 Other BC B
F5	Mole flow of methanol in the stream to splitter F5 (MF5)	MF5 ≤ 1 Other D CD
F6	Mole flow of ethyl acetate in the stream to splitter F6 (MF6); side draw of ethanol from T1 (SIDE1)	SIDE1 is present MF6 ≥ 1 Other D BCD CD

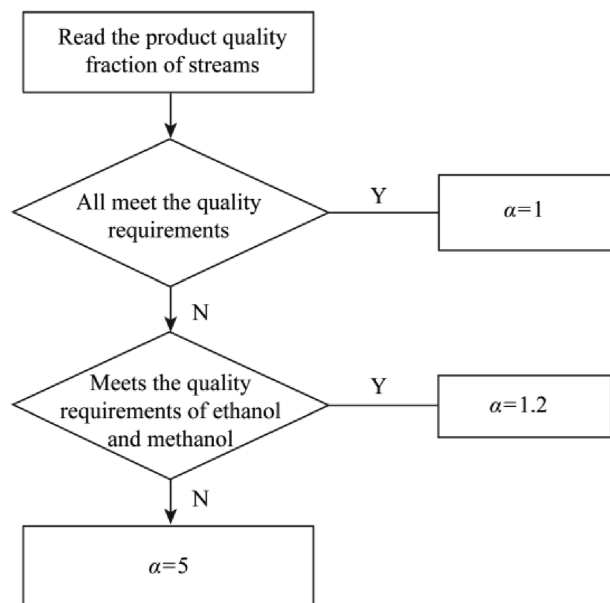


Fig. 9. Schematic diagram of external penalty function processing method.

columns. The temperature of the reboiler stream of T6, T7, T8, and T11 is too high to receive the latent heat of condensation, as shown in Fig. 13, which necessitates the use of heat utilities.

On the other hand, the condenser stream of T1, T3, T4, T9, and T10 fail to be utilized as the latent heat of the reboiler stream due to their low grade and require cold utilities. The latent heat of the T5,

T6, T7, T8, and T11 condenser stream is fully recovered. Due to the latent heat received, no additional reboilers are required for T1, T4, and T10, whereas the reboilers of T3, T5, and T9 receive part of the latent heat from condensation and still need utility. After heat integration, the structure has an equal actual heat exchange ratio (AHER) and theoretical maximum heat exchange ratio (TMHER), reaching 54.68%, which is slightly lower than the best value of 55.55% reported in previous work, indicating good heat utilization. AHER is the ratio of the utilized latent heat of the condenser to the total latent heat of the condenser. TMHER is the ratio of the latent heat of the condenser above the pinch to the full latent heat of the condenser.

The optimal sequence '13' with the obtained identification code 200223 is individually optimized to verify the superstructure optimization performance. Its constraint variable declarations are shown in Supplementary Material S1. The numbering of columns is consistent with that of the superstructure. The single-process optimization for sequence '13' also terminates in 3000 generations with the same approach and equipment as in the superstructure optimization. As shown in Fig. 14, the curve converges after 2000 generations of evolution. The gap between the average and optimal values steadily decreases, and the final gap is of only 1.48%, comparable to the optimization results obtained for the superstructure.

The optimal heat integration structure of sequence '13' is shown in Fig. 15, and the detailed column parameters are shown in Supplementary Material S2. In contrast, the temperature–enthalpy diagram is shown in Fig. 16. The final optimal heat integration structures obtained are “T7&T8&T11→T5→T4&T10”, “T7→T1&T5”, “T6→T1&T3&T4&T9”, and “T6→T1&T3&T4&T9”, as seen in Fig. 15. Fig. 16 illustrates that similar to the superstructure optimization, the latent heat from the

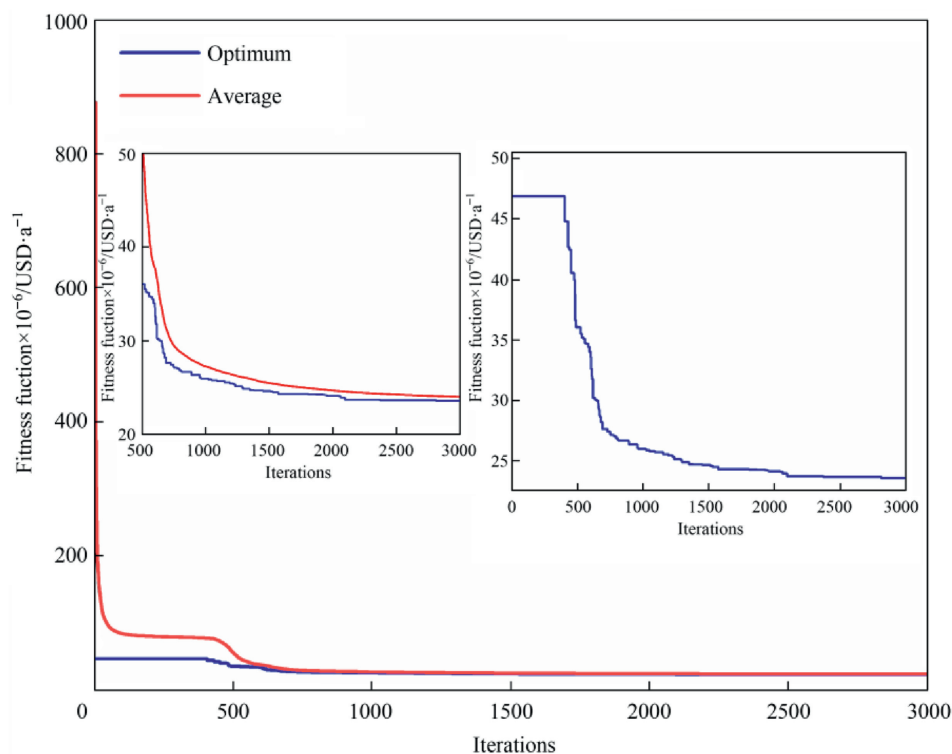


Fig. 10. The optimization progress curves of TAC for process synthesis.

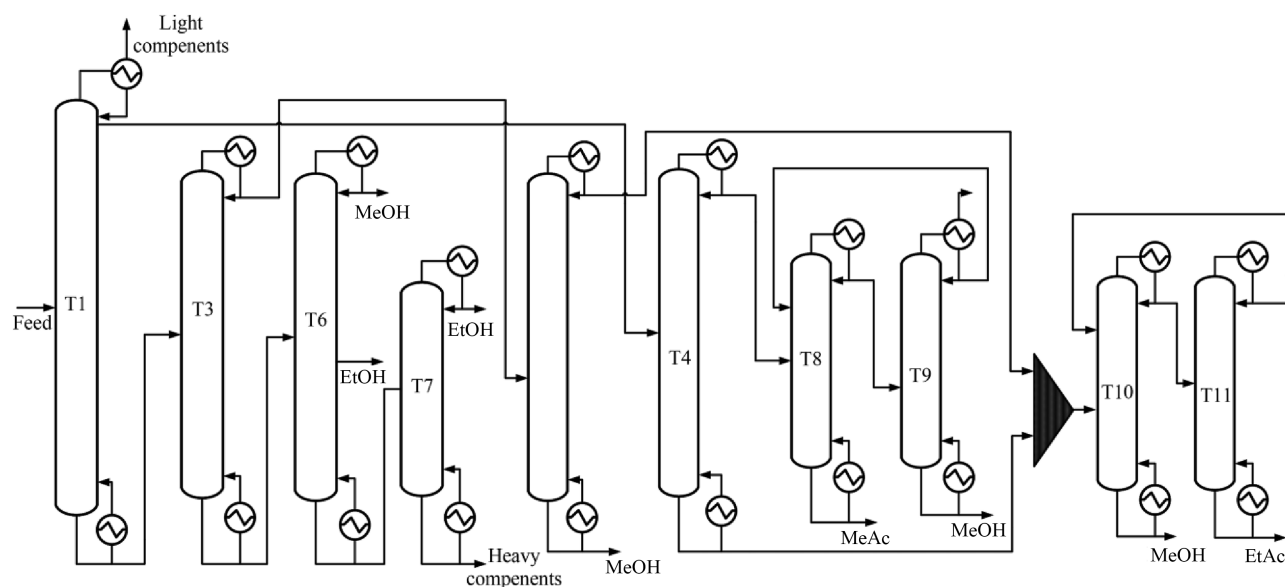


Fig. 11. Process configuration of the optimal sequence (code: 200223).

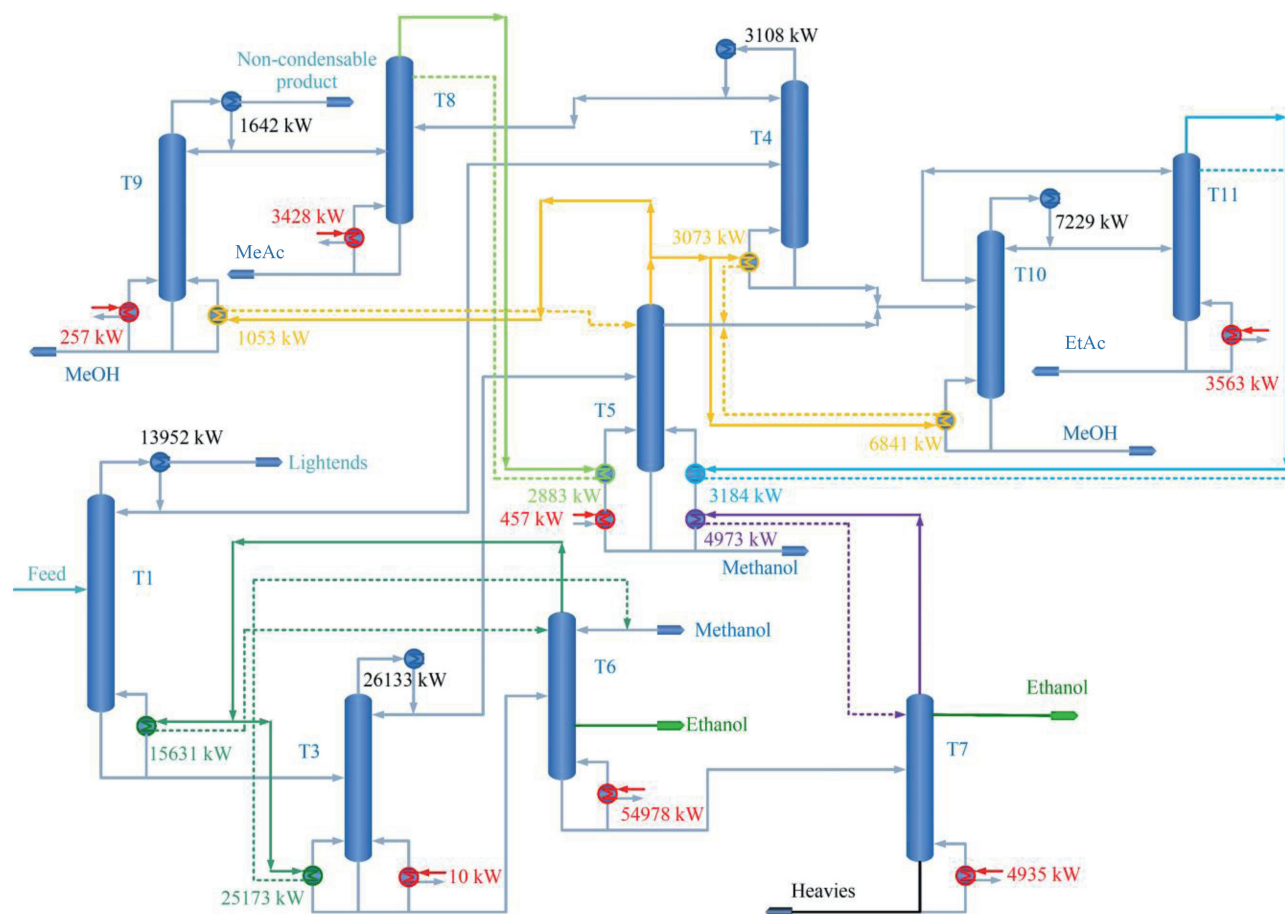


Fig. 12. The heat integration structure diagrams of the optimal solution for the superstructure.

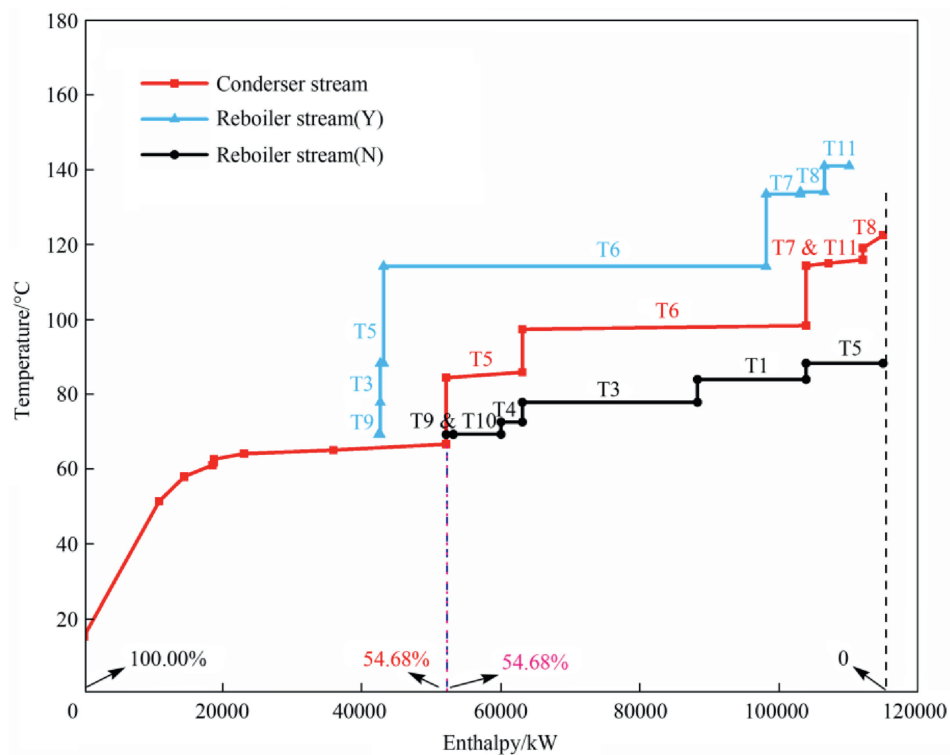


Fig. 13. The temperature–enthalpy diagrams of the optimal solution for the superstructure.

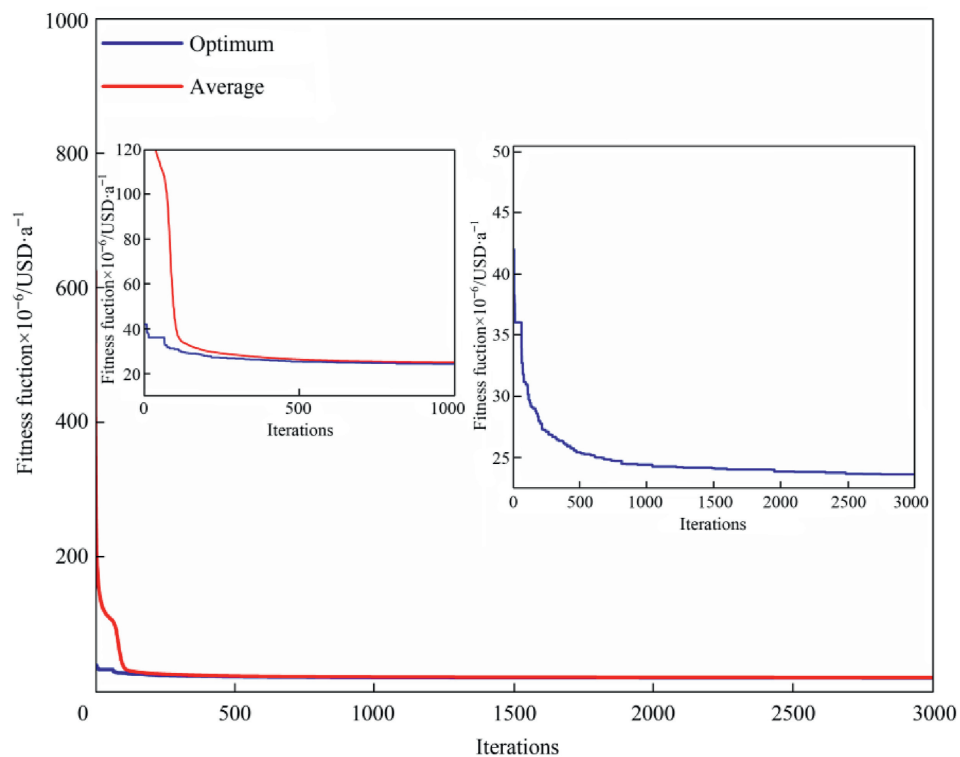


Fig. 14. The optimization progress curves of TAC for sequence '13'.

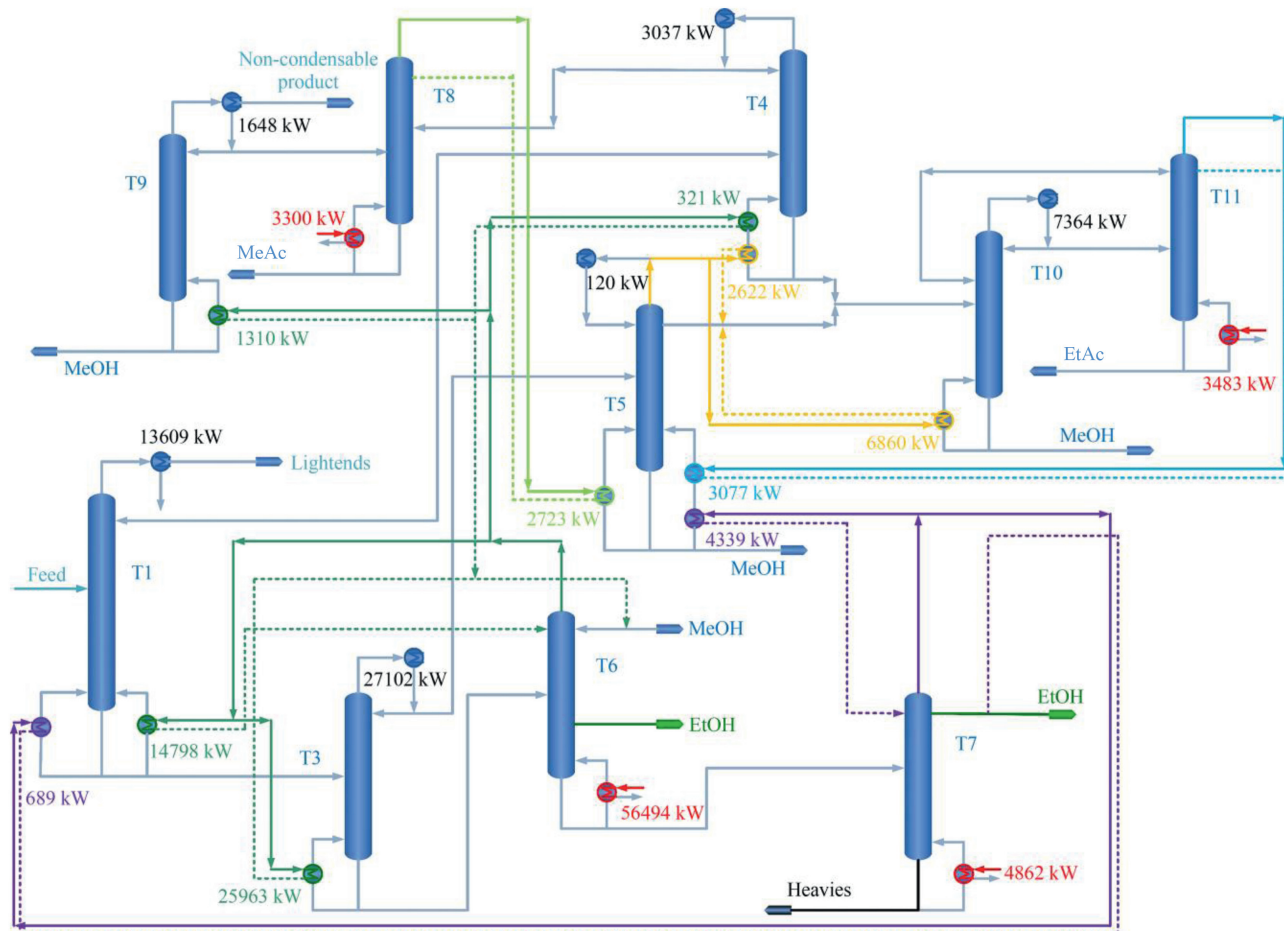


Fig. 15. The heat integration structure diagrams the optimal solution for sequence '13'.

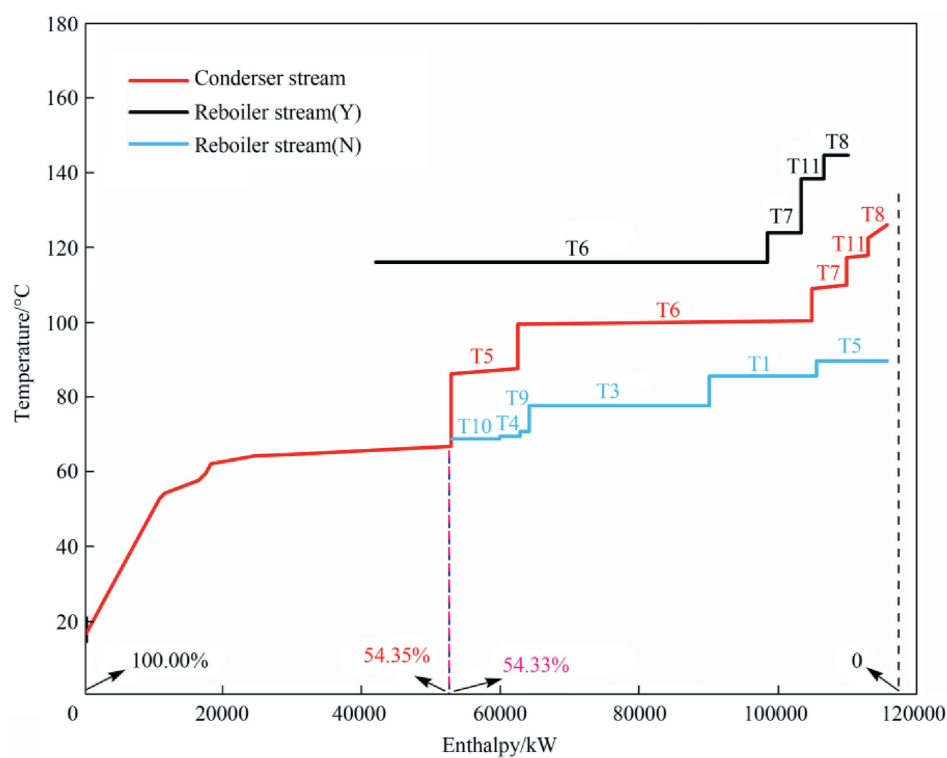


Fig. 16. The temperature–enthalpy diagrams of the optimal solution for sequence '13'.

Table 7

Comparison of optimization results

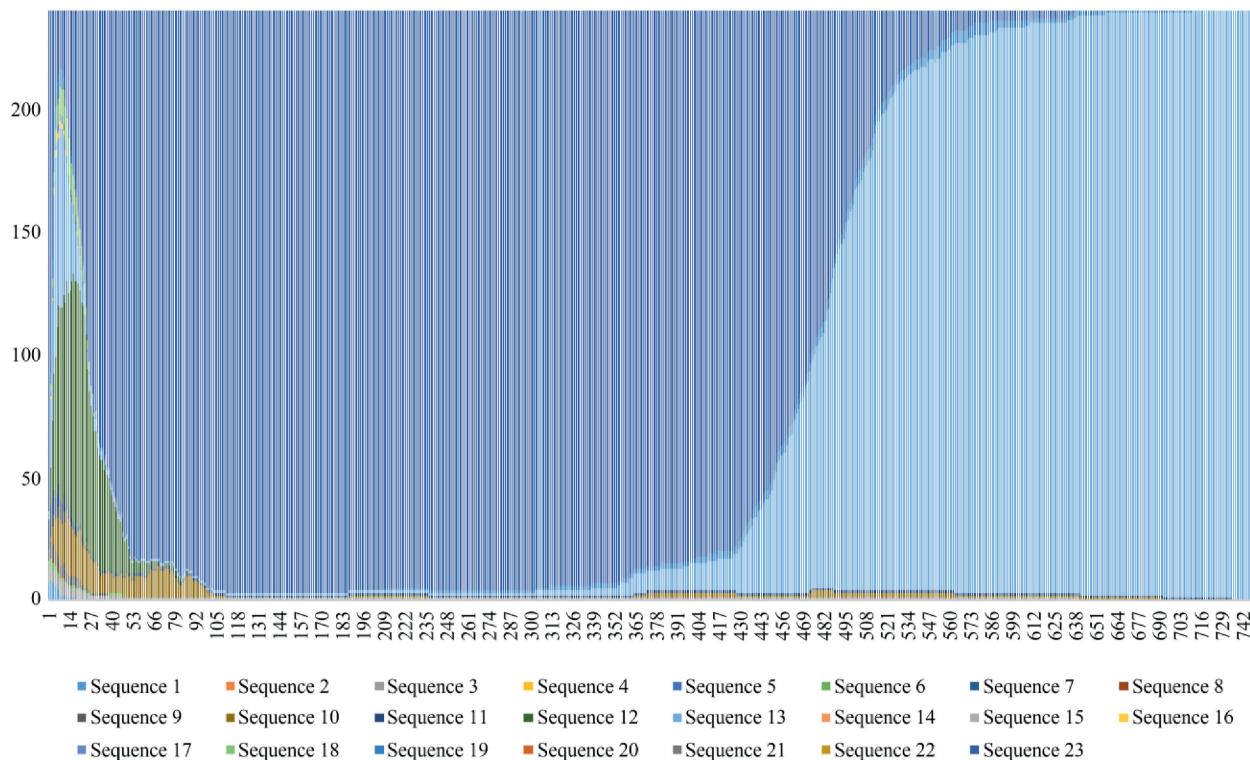
Options	Superstructure optimization	Sequence '13' single process optimization	Benchmark process
Average $\times 10^6/\text{USD}\cdot\text{a}^{-1}$	23.9877	23.9814	33.1843
Optimum $\times 10^6/\text{USD}\cdot\text{a}^{-1}$	23.5746	23.6277	32.8990
Variable dimension	59	48	37
(Steam/ethanol)/ $\text{kg}\cdot\text{kg}^{-1}$	1.8478	1.8618	2.4840
(Low-pressure steam/ethanol)/ $\text{kg}\cdot\text{kg}^{-1}$	1.8478	1.8618	2.3735
(Medium-pressure steam/ethanol)/ $\text{kg}\cdot\text{kg}^{-1}$	—	—	0.1105
CAPEX $\times 10^6/\text{USD}\cdot\text{a}^{-1}$	6.2530	6.2244	4.9284
OPEX $\times 10^6/\text{USD}\cdot\text{a}^{-1}$	17.3216	17.4033	27.9706

condenser stream from T6, T7, T8, and T11 is fully recovered. Recovery of latent heat eliminates the need for additional reboilers in T1, T3, T4, T5, T9, and T10. However, the reboiler in T6 is still necessary as it does not receive sufficient latent heat from the condenser stream to fulfill its whole duty. T7, T8, and T11 have a reboiler stream temperature that is too high to utilize condensing heat exchange and thus require heat utilities. Conversely, the condenser stream of T1, T3, T4, T9, and T10 cannot utilize latent heat and require cold utilities. Additionally, the latent heat of condensation from T5 is not fully used and, therefore, involves the addition of cold utilities. Moreover, it has similar AHER and TMHER after heat integration, 54.33% and 54.35%, respectively, which is extremely near to the superstructure optimization with a difference of just 0.64%.

The results of the superstructure optimization and the single-process optimization can be found in Table 7 to be equivalent, with a difference of only 0.22% in the optimal value and of 0.75% in specific steam consumption (SSC) of ethanol. The benchmark of prior research is used to provide a reference for comparison

[11]. The TAC results of both superstructure and single-process optimization are superior to benchmark configuration by 28.34% and 28.18% in terms of TAC results, respectively. Furthermore, it is a 5.74% improvement compared to the configuration with the best TAC reported in the previous research. However, this process's capital cost (CAPEX) is higher than those of the benchmark and the other configurations previously reported, as expected. Because the previously reported methods are 8- or 9-column configurations, Sequence '13' has 10 columns, leading to increased CAPEX. From the perspective of the SSC of ethanol in Supplementary Material S4, the results show $1.8478 \text{ kg}\cdot\text{kg}^{-1}$ (steam/ethanol), which is much higher than the benchmark but only marginally lower than that in the previous optimized result of $1.8177 \text{ kg}\cdot\text{kg}^{-1}$ (steam/ethanol). In addition, yields, mass flows, and mass fractions for superstructure and sequence 13 can be found in Supplementary Material S3.

The distribution of sequences as the iteration progresses can be found in Fig. 17, where sequence 23 is the infeasible sequence. At the beginning of the optimization, there is a wide variety of

**Fig. 17.** The distribution of sequences with iteration.

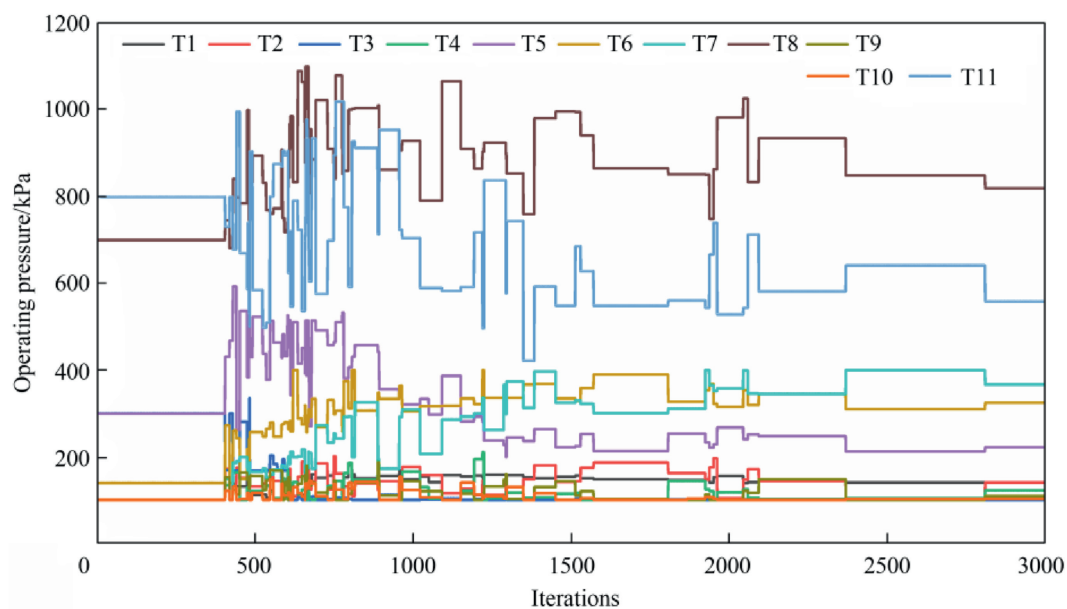


Fig. 18. Variation of operating pressure in best value with iteration.

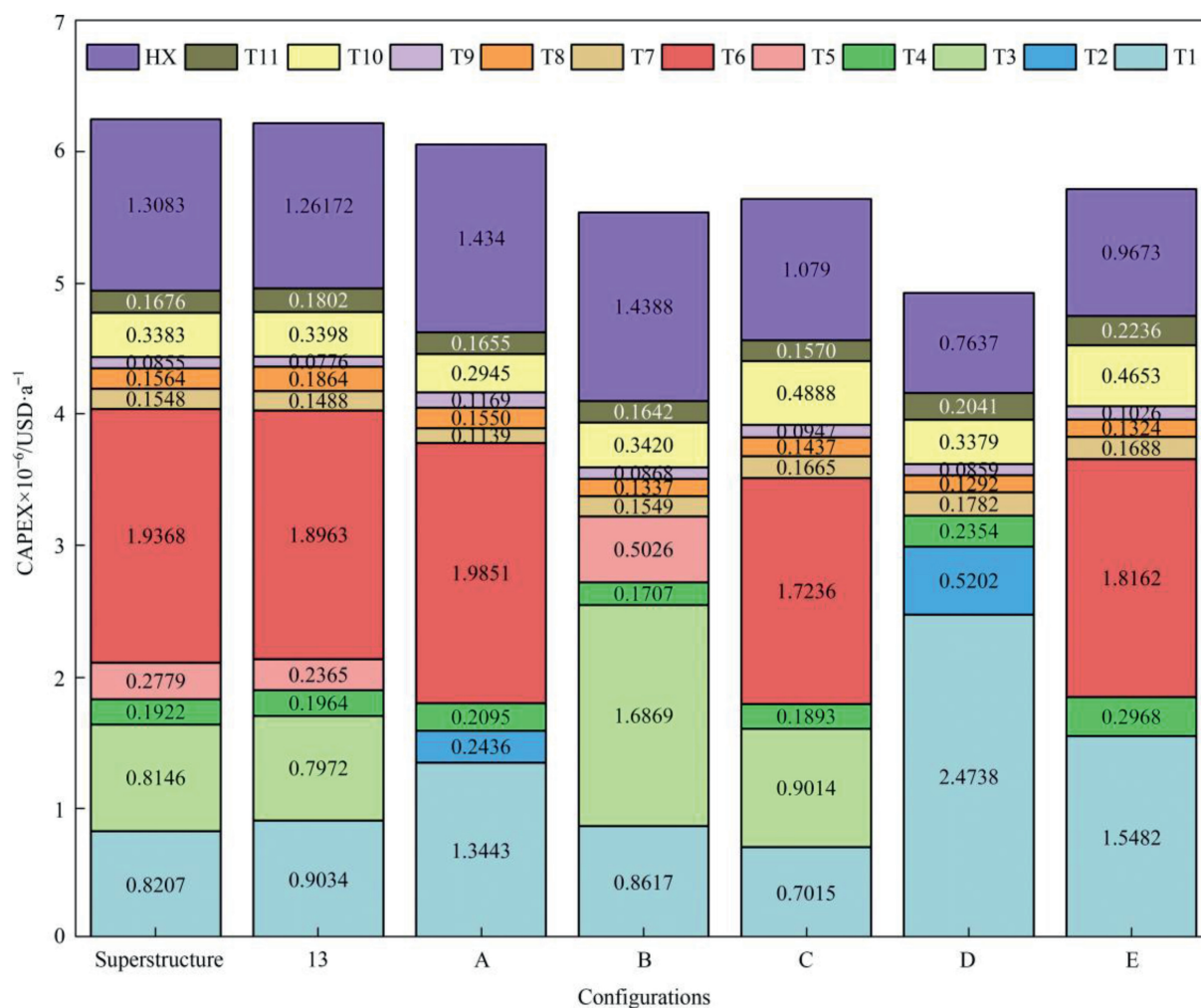


Fig. 19. CAPEX in the superstructure, sequence '13' and five previously reported configurations.

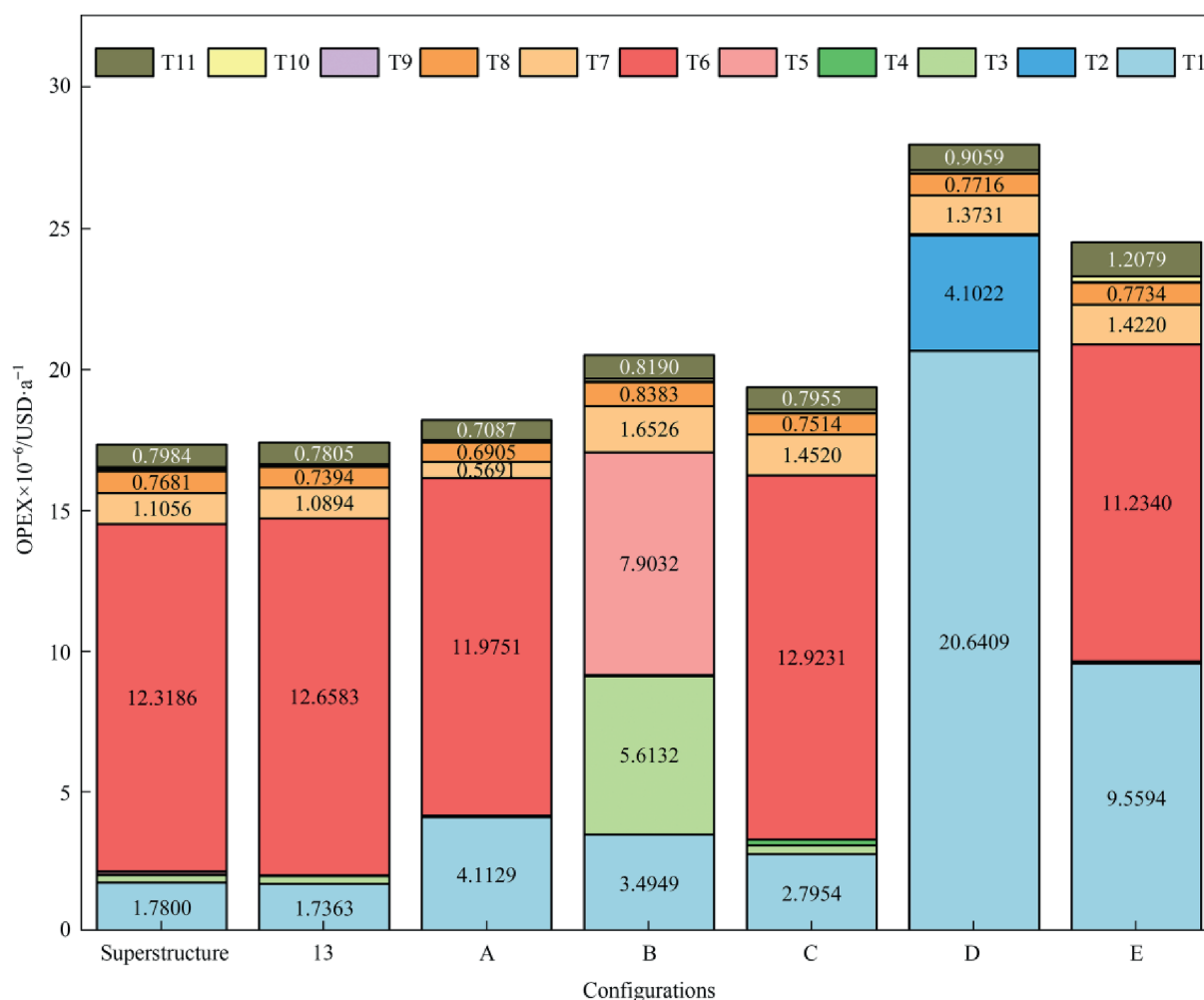


Fig. 20. OPEX in the superstructure, sequence '13' and five previously reported configurations.

sequences, with a large proportion of feasible sequences; however, as the optimization progresses, more and more sequences are excluded. It is not until generation 500 that the ratio of feasible to infeasible solutions begins to reverse, and the optimal sequence 13 stands out from the rest. Corresponding to the trend in Fig. 10, it can be seen that the fastest-changing interval is the interval of sequence screening.

The variation of operating pressure with iteration is illustrated in Fig. 18, with the most pronounced change occurring in the sequence screening timeframe as well. We can also notice from this that as the optimization proceeds, T6, T7, and T8 increase the pressure and T5 and T11 decrease it. From the results in Figs. 13 and 16, it is straightforward to conclude that excellent heat transfer is required for this sequence of systems to satisfy the requirement that T5 can supply heat to the lower-energy stages. Similarly, T5 is capable of being provided with heat by the higher-energy columns. The T5 column is closest to the ethanol separation, which means it is the highest grade of column equipment that can potentially receive the latent heat of condensation. This is because all columns involved in ethanol separation are of high grade and can only be heated by steam unless the operating pressure of the high-pressure azeotropic distillation columns is continually increased, which would add additional steam consumption.

The detailed CAPEX, operating costs (OPEX) and TAC for the superstructure optimization, sequence '13' single-process optimization, and the five previously reported configurations [11] are shown in Figs. 19–21, respectively. In addition, more information about SSC is shown in Supplementary Material S4. Fig. 21 illustrates that T1 has a low-cost share in the more efficient configurations for CTE product purification. The presence of non-condensable light components places T1 in the low energy level of the device for all configurations, where heat cannot be utilized in any way. Therefore, columns with similar low energy levels should perform simpler separation tasks such as non-sharp splitting, which transfers the purification burden to the higher-energy-level columns, where the treatment is more conducive to energy laddering.

Furthermore, the columns responsible for ethanol extraction from the side draw contribute significantly to the system's energy consumption, serving as both a heat source and a heat sink. Therefore, it is necessary to improve its energy taste to maximize the utilization of its latent heat and thus achieve a lower SSC. Finally, considering that OPEX accounts for 73%–85% of TAC, it is fruitful to increase the equipment appropriately to reduce the purification burden and increase the potential heat exchange for the separation of CTE products.

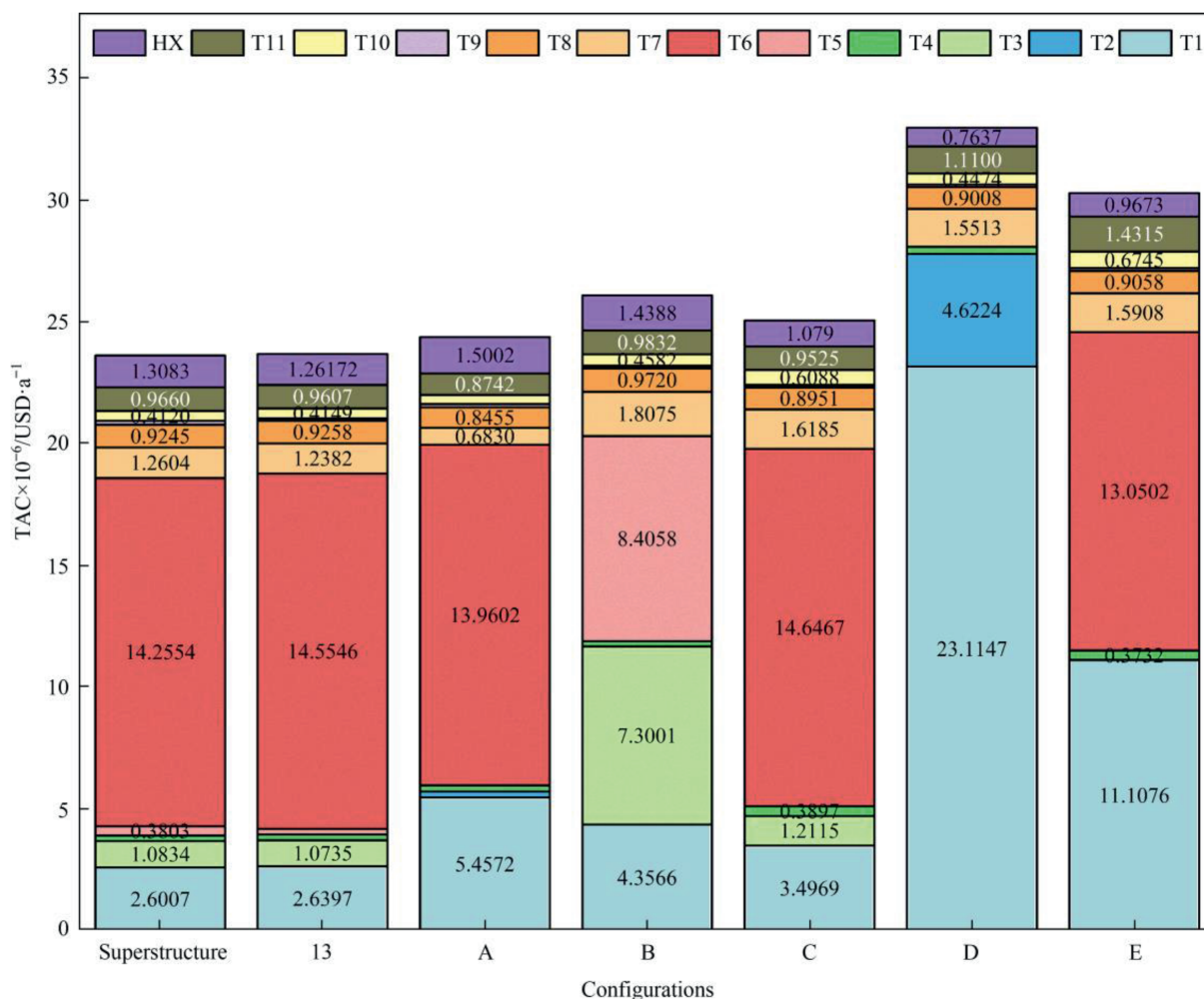


Fig. 21. TAC in the superstructure, sequence '13' and five previously reported configurations.

5. Conclusions

This work further explores the competitive configuration for ethanol distillation of the CTE route by the simulation- and superstructure-based process synthesis. In contrast to empirical-based approaches, the superstructures constructed in this process synthesis are comprehensive, considering all potential sequences, including sharp and non-sharp splits. The solution of the process synthesis problem successfully provides a more economical distillation sequence, of which the TAC is 28.34% lower than the benchmark, making the CTE route more promising as the clean utilization of coal. Meanwhile, the slight difference between optimized results obtained from the superstructure-based optimization and the single-process optimization also indicates that the adaptive strategy to select branches and nodes of SENs is helpful in large-scale process synthesis.

In addition, this work successfully performs a superstructure-based optimization that uses rigorous models with 59 decision variables, the largest rigorous model-based synthesis of a chemical process that has been reported. It demonstrates the simulation-based approach, which is iterated by stochastic algorithms, and capable of dealing with large-scale process optimization and synthesis.

Although this work is based on rigorous modeling, the presence of azeotropic compositions makes it impractical to judge the component boundaries accurately; therefore, the determination of some values relies on empirical means. In addition, combining or

splitting columns can be explored to investigate the optimal separation sequence for CTE product separation comprehensively. Ultimately, it is recommended that the methodology of this paper be used for systems with clear boundaries that are easy to recognize. Otherwise, empirically based values will be needed to determine the component boundaries, introducing uncertainty into the screening of sequences.

Declaration of 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.

Supplementary Material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.cjche.2023.12.019>.

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