

Rigorous Simulation-Based Automatic Identification of the Optimal Separation Sequence

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Separation sequence optimization is an energy-saving strategy for improving the process economy. This work introduces a rigorous simulation-based method to identify the best separation sequence for the separation of N -component mixture, aiming to reduce the total operating cost. Through data interaction between Aspen HYSYS and MATLAB, a separation sequence matrix and an operating cost matrix for the N -component mixture are established. Based on this, the optimal separation sequence with the lowest total operating cost is automatically identified. The separation of 5-component hydrocarbon mixture is studied to illustrate the feasibility of the proposed method. This work also evaluates separation sequences for variant feed compositions, and shows the effects of feed compositions on the best separation sequences. This method is also applicable for separation of the other mixture, giving a new solution for identification of the optimal separation sequence.

1. Introduction

As a thermal separation method, distillation is the most important and widely used separation technology in the chemical processes (Özçelik and Mert, 2016). However, the temperature difference between the reboiler and condenser leads to inevitable degradation of energy. Because of involving evaporation steps, more than half of the process heat distributed to a plant is dedicated to supply heat for the reboilers of distillation columns (Caballero, 2015), which significantly influences the overall plant profitability. If multi-component mixtures are separated into a number of products by distillation, there is an explosive growth of the number of all possible separation sequences along with the number of components. Different distillation columns are related to different separation processes, resulting in discrepant operating costs. Identifying the optimal separation sequence plays a critical role in the reduction of the total operating cost.

The identification of the optimal separation sequence can be formulated as a mixed integer nonlinear programming (MINLP) by considering all the mass and enthalpy balances, and phase equilibrium equations (Grossmann et al., 2005). Kelloway and Daoutidis (2014) developed a biorefinery superstructure and established a MINLP model to get the optimal biorefinery configuration with the highest economic potential or carbon efficiency. Wang et al. (2019) decomposed the MINLP problem into integer programming and nonlinear programming problems to reduce the computation required in both sharp and non-sharp distillation sequence optimization. Raman and Grossmann (1994) proposed generalized disjunctive programming to overcome the difficulties of MINLP model. Due to the mathematical complexity associated with rigorous methods, the optimal determination of column sequences has been generally carried out using shortcut methods (Caballero and Grossmann, 2014). However, the computational intension of the simplified models can still not be reduced when more components are considered. Lv et al. (2019) utilized the partition method to target the optimal distillation sequence of the reaction-distillation-recycle integration system. Yin and Liu (2019) developed a systematic method to integrate the reactor with distillation sequence, and automatically identified the best one on the basis of non-key components and marginal minimum vapor rates. A matrix method is proposed for targeting the optimal distillation sequence and reactor parameters simultaneously by Lv and Liu (2019). These contributions lower the mathematical complexity based on further simplification of

the model. However, the simplified model is too unrealistic, and the proven optimum may not be the best solution in the practice (Aspelund et al., 2010). In order to solve the problem easier and more effective, it is imperative to overcome the great challenge in the distillation sequence optimization with rigorous simulation. This work proposes a rigorous simulation-based method to automatically determine the optimal separation sequence with the lowest total operating cost. The method is accomplished by the data interaction between Aspen HYSYS and MATLAB. First, the separation sequence matrix (S) for the *N*-component mixture is established based on the permutation matrix (P) in MATLAB. Then, Aspen HYSYS is used to conduct rigorous simulation for all possible separation tasks in S matrix, and the operating cost matrix (C) is generated after the data transferred to MATLAB. Subsequently, the multiplication of S matrix and C matrix is performed to identify the optimal separation sequence. The proposed method is used to analyse the separation of 5-component hydrocarbon mixture, and variant feed compositions are evaluated to obtain their effects on the optimal separation sequences.

2. Establishment of the separation sequence matrix

Take a quaternary mixture as an example. Assuming that all distillation columns perform the sharp split, the State Task Network (STN) superstructure (Caballero and Grossmann, 2014) for separating the mixture into nearly pure components is shown in Figure 1. Note that A, B, C and D represent the four components with decreasing volatility.

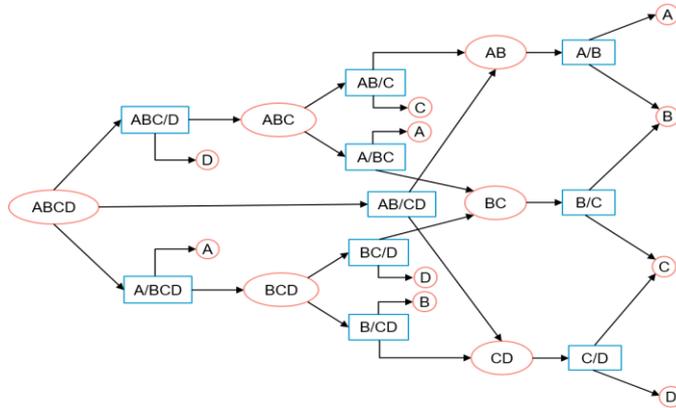


Figure 1: State Task Network Superstructure for a quaternary Mixture

As shown in Figure 1, there are five separation sequences and ten alternative separation tasks for separating a quaternary mixture. Each sequence consists of three separation tasks. A separation sequences matrix (S) is developed to show all the sequences as given in Figure 2. Each row corresponds to a separation sequence, and each column corresponds to a separation task (arranged in order of decreasing partition component numbers and the partition location from left to right). When a separation task is used in a separation sequence, the corresponding column is denoted as “1”. Otherwise, it takes “0”.

	L_4			L_3				L_2		L_1
	A/BCD	AB/CD	ABC/D	A/BC	AB/C	B/CD	BC/D	A/B	B/C	C/D
1	0	0	1	0	1	0	0	1	0	0
2	0	0	1	1	0	0	0	0	1	0
3	0	1	0	0	0	0	0	1	0	1
4	1	0	0	0	0	0	1	0	1	0
5	1	0	0	0	0	1	0	0	0	1

10 separation tasks

Figure 2: Separation sequences matrix for a quaternary mixture

For separating a *N*-component mixture, the number of all separation sequences can be calculated as Eq(1) (Thompson and King, 1972). Eq(2) shows the number of alternative separation tasks in all the separation sequences (Lv and Liu, 2019), where L_i is the number of separation tasks of partitions occurred in *i* components. *N*-1 separation tasks are used in each sequence, so the S matrix for a *N*-component mixture is *m*×*n* dimensional, and *N*-1 columns are denoted as “1” in each row.

$$m = \frac{(2(N-1))!}{N!(N-1)!} \quad (1)$$

$$n = L_N + L_{N-1} + \dots + L_2 = 1 \times (N-1) + 2 \times (N-2) + \dots + (N-1) \times 1 \quad (2)$$

Permutation matrix (P) is introduced to automatically generate the S matrix for a N-component mixture. There are N-1 separation tasks for each separation sequence. In other words, N-1 partitions are required in each sequence. If 1, 2, 3, ... N-1 represent the partition location from left to right, the separation sequence matrix can also be considered as full permutation of the N-1 numbers (Lv and Liu, 2019). The size of P matrix is (N-1)! \times (N-1). It is noted that each row corresponds to the separation sequence, and each column corresponds to the partition location. However, some of the partition order can be inverted, so there are duplicated partition sequences in P matrix. Figure 3 gives the permutation matrix for a quaternary mixture.

Partition location

$$\begin{bmatrix} 3 & 2 & 1 \\ 3 & 1 & 2 \\ 2 & 3 & 1 \\ 2 & 1 & 3 \\ 1 & 3 & 2 \\ 1 & 2 & 3 \end{bmatrix}$$

Figure 3: Permutation matrix for a quaternary mixture

Each row in P matrix corresponds to the same row in S matrix. On this basis, this work proposes a programmable method to automatically build the S matrix according to the following procedure:

(1) For the first column in P matrix, assign the value of $p_{i,1}$ to t , and the element in the t^{th} column of the i^{th} row in S matrix is taken as "1".

(2) For the column $j > 1$ in P matrix:

(i) If the value of $p_{i,j}$ is smaller than the minimum value (U) of the $j-1$ preceding elements in i^{th} row, $t = L_{U+1} + L_{U+2} + \dots + L_N + p_{i,j}$.

(ii) If the value of $p_{i,j}$ is larger than the maximum value (T) of the $j-1$ preceding elements in i^{th} row, $t = L_{N-T+1} + L_{N-T+2} + \dots + L_N + T \times (N-T-1) + p_{i,j} - T$.

(iii) If the value of $p_{i,j}$ lies between the maximum and minimum values of the $j-1$ preceding elements in i^{th} row, find the two neighboring values of the value $p_{i,j}$. Supposing that the greater value is H and the smaller value is V, $t = L_{H-V+1} + L_{H-V+2} + \dots + L_N + V \times (H-V-1) + p_{i,j} - V$.

Likewise, take the t^{th} element of the i^{th} row in S matrix as "1".

(3) Repeat steps 1-2 for each row of P matrix.

(4) Delete the duplicated rows, and the S matrix for a N-component mixture can be determined.

3. Identification of the optimal separation sequence

S matrix lists all the separation sequences for N-component mixtures intuitively. The best sequence is identified by calculating the total operating cost of each sequence.

3.1 Operating cost matrix

Before identifying the optimal separation sequence for a N-component mixture, operating cost matrix (C) is constructed. C matrix is a column matrix, which consists of the operating costs of the n separation tasks in S matrix in the same order. The operating cost of each separation task includes utility costs consumed by condenser, reboiler, and the pump or compressor used to pressurize the feed of the distillation column. The duties of the distillation columns, pumps and compressors are estimated based on the rigorous simulation in Aspen HYSYS. Through data interaction between Aspen HYSYS and MATLAB, the C matrix can be generated as Figure 4.

$$C = \begin{bmatrix} C_1 & C_2 & C_3 & \dots & C_{n-1} & C_n \\ 1 & 2 & 3 & \dots & n-1 & n \end{bmatrix}$$

Separation tasks

Figure 4: Operating cost matrix for a N-component mixture

3.2 Solving procedure

The proposed method is applicable to any number of components. Detailed solving procedure is shown in Figure 5:

- (1) For a given N -component mixture, build P matrix in MATLAB.
- (2) Based on the number of components (N) and P matrix, generate S matrix in MATLAB.
- (3) Conduct rigorous simulation models for all the alternative separation tasks in S matrix using Aspen HYSYS, and the operating cost data are transferred to MATLAB to establish the C matrix.
- (4) Calculate the multiplication of S and C matrices to get the total operating cost of each separation sequence.
- (5) By comparison, accomplish the identification of the optimal separation sequence and output results.

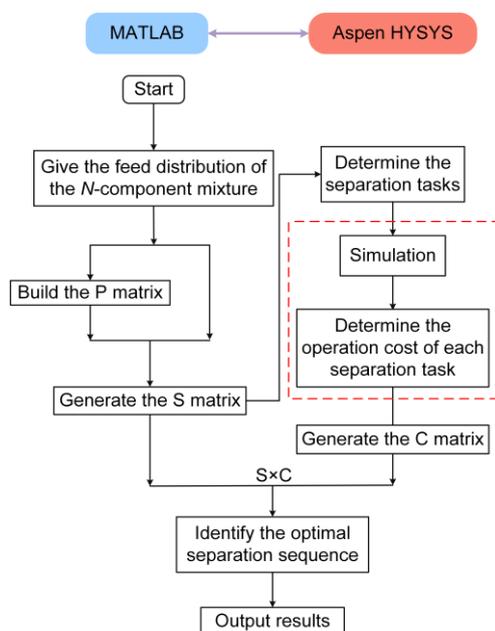


Figure 5: Procedure for automatic identification of the optimal separation sequence

4. Case study

As important organic raw materials, aromatics play a pivotal role in chemical industries, so manufacturing aromatics products from hydrocarbon mixtures has been widely concerned. The case of separating hydrocarbon mixture into non-aromatics, benzene, toluene, xylene and C_9+ aromatics is studied to illustrate the feasibility of the proposed method. Because different feed compositions result in different optimal separation sequences, the effects of feed compositions on the best sequences have been also analyzed in this work.

4.1 Identification of the optimal separation sequence

The feed distribution of the 5-component hydrocarbon mixture is shown in Table 1 (Zhang et al., 2019), and the flow rate takes $40 \text{ t}\cdot\text{h}^{-1}$. According to the solving procedure in Section 3.2, the S matrix (14×20) can be generated in MATLAB R2018a V9.4 (MathWorks, 2018). Aspen HYSYS V10.0 (AspenTech, 2017) is used to conduct rigorous simulation for the 20 separation tasks based on the product specifications (Xu, 2014), and the C matrix ($10^6 \text{ \$}\cdot\text{xy}^{-1}$) is built through data interaction with MATLAB. The multiplication of S and C matrices is then performed to get the total operating cost matrix C' ($10^6 \text{ \$}\cdot\text{xy}^{-1}$), as depicted in Figure 6. The optimal separation sequence is the 6th sequence, and the corresponding sequence is depicted in Figure 7.

Table 1: The feed distribution of the 5-component hydrocarbon mixture (wt%)

Non-aromatics (N_0)	Benzene (B)	Toluene (T)	Xylene (X)	C_9+ aromatics (C_9)
0.2240	0.0574	0.1965	0.4068	0.1153

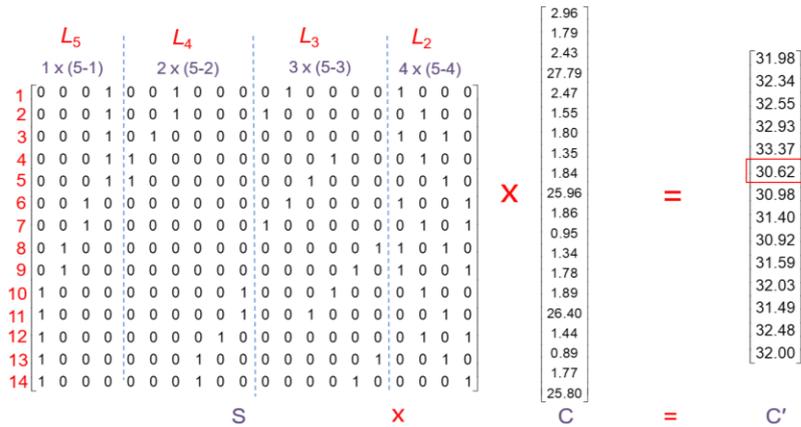


Figure 6: Relevant matrices for separating the 5-component hydrocarbon mixture

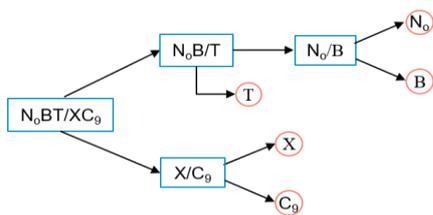


Figure 7: The optimal separation sequence for separating the 5-component hydrocarbon mixture

4.2 The effects of feed compositions on the optimal separation sequences

In this subsection, the optimal separation sequences for variant feed compositions are analyzed. As shown in Figure 8a, each vertex of the regular pentagon represents a certain component of the mixture. For any point A within the pentagon, the distances of point A to the five sides represent the relative flow rates of the diagonal components, denoted as h_1, h_2, h_3, h_4 and h_5 . Any point within the pentagon corresponds to a certain composition of the mixture. The closer the point is to a vertex, the higher is the corresponding component. For the point A, the composition of each component can be calculated by Eq(3).

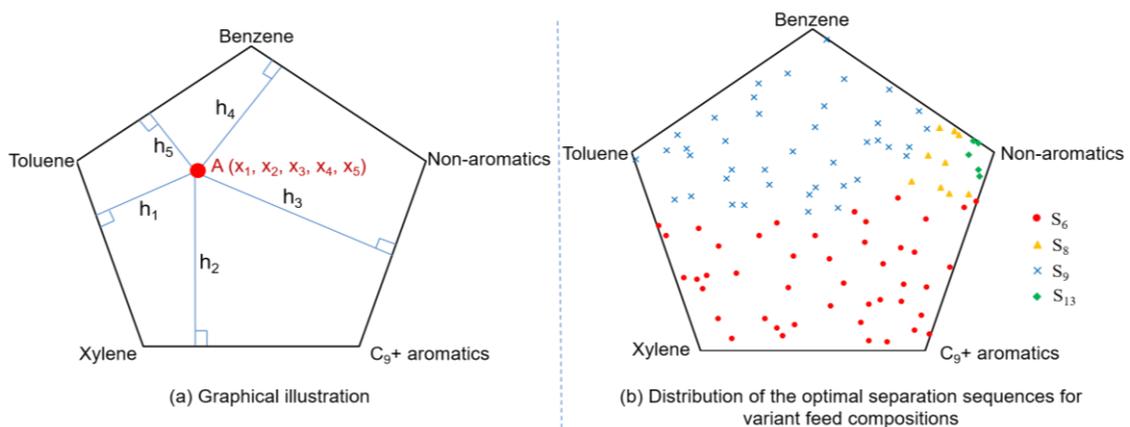


Figure 8: Effects of feed compositions on the optimal separation sequences

$$x_i = \frac{h_i}{\sum_{i=1}^5 h_i} \quad (i=1, 2, 3, 4, 5) \tag{3}$$

MATLAB is utilized to randomly generate 100 raw materials and they are analyzed using the proposed method. Figure 8b gives the distribution of the optimal separation sequences for variant feed compositions. Note that S_i represents the optimal separation sequence is the i^{th} sequence of S matrix. The best separation sequence is the 6th or 9th sequence in most cases. However, when non-aromatics is much more than the other components, the best sequence is altered to the 8th or 13th sequence. For the aromatic components, the optimal separation sequence for the mixture with more benzene tends to the 9th one, and the best one is the 6th sequence as the mixture contains more xylene or C₉₊ aromatics. From a further insight into Figure 8b, the optimal separation sequence prefers separating mixture into two parts with similar flow rates. The other 10 separation sequences do not appear in Figure 8b, when targeting the lowest total operating cost.

5. Conclusions

In this paper, a systematic method based on rigorous simulation was proposed for automatically identifying the optimal separation sequence of multiple components. A 5-component hydrocarbon mixture was studied to illustrate the feasibility of the proposed method, and the results showed that the operating cost of the optimal separation sequence was 3.062×10^7 $\text{\$}\cdot\text{y}^{-1}$, which was 8.24 % lower than the worst one. The investigation on variant feed compositions indicated that the optimal separation sequence preferred separating mixture into two parts with similar flow rates. Because of introducing rigorous simulation, the method would be close to the practice. This method can also be applied to other systems for automatic identifications of the best separation sequences. However, the proposed method is merely aimed at single-objective optimization for the separation system. In the future work, more issues including multiple-objective optimization, flexible system based on different product configurations, simultaneous optimization of reaction-separation system and others will be studied based on the proposed method.

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