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A systematic approach for synthesizing a low-temperature distillation system $\overset{\curvearrowleft}{\sim}$



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A R T I C L E I N F O

ABSTRACT

result is encouraging.

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1. Introduction

Many chemical separation processes, such as gas separation and ethylene production that operate completely or partially in lowtemperature circumstance consume significant quantities of energy. The design and operation of low-temperature distillation processes generally include three sections: the core process (the separation sequence), the heat exchanger network (HEN) and the refrigeration system (RS). The traditional approach in practice for the design of low-temperature processes begins with the design of the separation process; after that, the HEN is designed to recover heat, and finally, the refrigeration system is designed to support the former systems. Because the three parts of the system are interrelated, a separation sequence optimized with the consideration of the heat recovery in the HEN may not necessarily correspond to the minimal energy cost of the global system without considering the RS simultaneously because the energy cost for a low-temperature separation system depends on the shaft work of the compressors. As a result, it is difficult for the traditional method to obtain an optimal design of the entire system with three decoupled parts. Thus, a simultaneous design method is necessary to obtain the optimal total cost for the entire system.

Most of the existing research studies regarding distillation systems in low-temperature processes focused on the optimization of the separation sequences and HEN simultaneously, while the refrigeration system was designed separately [1–4]. Because the refrigeration system accounts for a large proportion of the total energy consumption for a lowtemperature process, the minimum total energy consumption cannot be guaranteed if the refrigeration system is not simultaneously considered in the optimization problem. Wang and Smith [5] attempted to build a mathematic model covering the separation sequence, integration and refrigeration simultaneously, and Tahouni *et al.*[6] improved the model. However, their model is complicated because of the consideration of the design of both HEN and RS, which requires a considerable effort to solve.

In this paper, by combining a stochastic optimization method with a refrigeration shaft work targeting method,

an approach for the synthesis of a heat integrated complex distillation system in a low-temperature process is

presented. The synthesis problem is formulated as a mixed-integer nonlinear programming (MINLP) problem,

which is solved by simulated annealing algorithm under a random procedure to explore the optimal operating

parameters and the distillation sequence structure. The shaft work targeting method is used to evaluate the minimum energy cost of the corresponding separation system during the optimization without any need for a

detailed design for the heat exchanger network (HEN) and the refrigeration system (RS). The method presented

in the paper can dramatically reduce the scale and complexity of the problem. A case study of ethylene cold-end

separation is used to illustrate the application of the approach. Compared with the original industrial scheme, the

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Linnhoff and Dhole [7] proposed a shaft work targeting method by extending the pinch analysis to yield the shaft work targets directly from basic process data. This approach simplifies the identification of the most energy-efficient system for a low-temperature process by not requiring knowledge of the HEN and RS structures. Therefore, in the present paper, by combining stochastic optimization with the shaft work targeting method, an approach for the synthesis of a heat integrated complex distillation system in a low-temperature process is presented. This approach involves the use of an improved simulated annealing algorithm [8] to explore the optimal operating parameters and the distillation sequence structure by a random procedure, a shaft work targeting procedure to evaluate the energy consumption of the separation processes that by-passes the detailed design of HEN and RS, and the shortcut method based on Fenske-Underwood-Gilliland to evaluate the capital cost of the distillation columns. The method presented in this paper can dramatically reduce the problem of the scale and complexity caused by searching a large number of optimal integer variables corresponding to the HEN and RS structures. The optimal distillation separation system that has the minimum total annual cost can be obtained with the consideration of the separation sequence, HEN and RS simultaneously.

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2. Model for the Separation Processes

2.1. Problem statement and assumptions

The problem in this paper can be described as follows.

Given an *N*-component mixture and the available utilities, find the distillation sequence that can separate the mixture into *N* products with a specified separation request and the minimum total annual cost, including the energy cost and the equipment depreciation cost. The separation structure can consist of simple columns, complex columns with side rectifying or side stripping, pre-fractionators and thermally coupled columns.

To simplify the problem, the following assumptions are adopted:

- (1) Constant molar flow inside the distillation columns;
- (2) Constant relative volatility for the components in the mixture;
- (3) No azeotrope exists in the mixture;
- (4) All the streams are saturated;
- (5) Only one middle (distributing) component exists in each non-sharp separator;
- (6) N product streams exist.

Assumptions (1)–(4) are widely used in the synthesis of a distillation sequence to simplify the problem so that the shortcut method for a distillation column design can be used. For non-sharp separation, the complexity of the problem will increase with an increase of the number of distributing components, and thus assumption (5) ensures that the problem is not too complicated.

2.2. Problem formulation

For a certain distillation system, the total annual cost can be the sum of the utility cost required by the separation and the capital costs of all the columns. The costs depend on the variables of the separation sequence (represented by a set of integer variables), the thermal coupling structure (a set of binary variables), the pressure of each column, the reflux ratio of each column and the recovery of the key components in non-sharp separation columns. Therefore, the synthesis problem can be formulated as the following optimization problem:

$$\min \text{COST} = \min C(\{s_i\}, \{\phi_i\}, p, \xi_{\text{LK}}, \xi_{\text{HK}}, r)$$
(1)

 $\{s_i\} \in \mathbf{SP}, \{\phi_i\} \in \boldsymbol{\Phi}, p \in \mathbf{P}$

$$\xi_{LK} \in [0.98, 1], \xi_{HK} \in [0, 0.02]$$

where the integer number series $\{s_i\}$ and $\{\phi_i\}$ represent the separation sequence structure and the thermal coupling scheme, respectively. *SP* and $\boldsymbol{\Phi}$ are the sets of all possible separation sequences of $\{s_i\}$ and thermal coupling structures $\{\phi_i\}$, respectively. *P* is a set of the feasible ranges of operating pressure *p*. The bound ranges of the recovery of light and heavy key components in non-sharp separation column ξ_{LK} and ξ_{HK} , respectively, are assumed based on operation specifications, and the ratio of the actual reflux ratio to minimum reflux ratio $r = R/R_{min}$, is fixed to 1.2 which is assumed as a relatively better value and will not be optimized in the model for simplification.

An improved simulated annealing algorithm presented by An and Yuan [9] is used to solve the problem. The algorithm combines simulated annealing with the simplex method, in which the simplex method is used to search for continuous variables, and the discrete variables are generated and evolve *via* the random method. Because of its characteristic of stochastic nature, the algorithm does not require an explicit definition of the search space through a superstructure, which includes all the potential solutions and the necessary constraint equations, and the convexity that is necessary for a traditional mathematical MINLP approach. Furthermore, because the detailed design of HEN and RS can be by-passed *via* the shaft work target method, there is no need for any other variables to describe HEN and RS. Once the above variables are fixed on a set of feasible values, the temperature of each stream and the heat duty of each condenser or reboiler corresponding to a certain separation sequence will be determined by the shortcut method. The capital cost is estimated using methods recommended by H. Silla [10]. Because the exergy of cold utilities in a low-temperature process comes from the shaftwork of refrigeration compressors, the energy cost of the refrigerant utilities is calculated by electric cost of the refrigeration compressors.

2.3. Representation of the separation sequence and the thermal couple structure

Based on An and Yuan [11], either sharp separation or non-sharp separation of the distillation tasks in a separation sequence can be represented by a set of integer number series. As is shown in Fig. 1, C_1-C_N are *N* components arranged in volatility descending order. An arrow with an odd number that points between two adjacent components indicates a sharp separation task, while an arrow with an even number that points at a component indicates a non-sharp separation task. For example, the arrow with number 2 indicates the non-sharp separation in a pre-fractionator in which the light key component is C_1 , the heavy key component is C_3 , and the distributing component is C_2 . In this way, the integer number series $\{s_i\}(i = 1, 2, ..., T)$ could be a T-dimensional ordered array, which consists of the ordinals of the separation tasks.





As has been assumed before, the feed of each column will either be saturated liquid or be saturated vapor. Thus, a T-dimensional binary array { ϕ_i }($i = 1, 2, ..., T, \phi_i = \{0, 1\}, \phi_1 = 0$) can be used to describe the feed condition of each column, namely, whether the interconnections between the separation tasks are a thermal couple or not, with a value of 0 showing the saturated liquid, and a value of 1 indicating the thermal couple stream.

2.4. Decomposition of the problem

To apply the shortcut method to the distillation column design, a complex distillation column flowsheet is converted into its thermodynamic equivalent simple column flowsheet. For example, in Fig. 2, the flowsheet (b) is the thermodynamic equivalent simple column flowsheet of the complex distillation column configuration (*c*). Each simple column has one stripping section and one rectifying section and finishes a single separation task (*M*). The number of column sections (*S*) and the separation tasks (*M*) in the thermodynamic equivalent simple column flowsheet meet the following inequalities according to An and Yuan [11]:

$$2(N-1) \le S \le 2(2N-3) \tag{2}$$

$$N-1 \le M \le 2N-3. \tag{3}$$

Therefore, when a separation sequence contains only sharp separation to separate *N*-components' mixture, (N - 1) sharp separation tasks with 2(N - 1) column sections are required. The number of separation tasks will increase when non-sharp separation is introduced. Each additional non-sharp separation increases a separation task, and thus, the maximum number of separation tasks (*M*) for *N* components

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