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Original Research Article

Coupling Optimization of the Distillation and Absorption Columns and Its Application in Methanol-to-Olefins Process

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ABSTRACT

Distillation and absorption are techniques used to separate mixtures, utilizing energy and absorbent materials as separation agents. These two processes are often combined for gas mixtures to enhance the recovery of key components while also reducing energy consumption. The operation of the distillation column influences not only its own performance but also that of the absorption column, and vice versa. A simplified model of combined absorption and distillation columns has been developed to analyse their performance concurrently. This model examines the relationship between the intermediate stream and the operating parameters of both units, yielding qualitative rules that can help minimize energy consumption and guide the optimization of the combined process. In the case of the methanol-to-olefins (MTO) process, the columns for demethanation and ethylene recovery have been qualitatively analysed using the proposed method, thus enabling efficient optimization of both columns. The optimal ethylene molar concentration at the top of the distillation column is found to be 0.5. Additionally, the corresponding reflux ratio is 1.6029, while the flow rates for the distillate and absorbent are 902.3 kmol/h and 900 kmol/h, respectively.

KEYWORDS

Distillation, Absorption, Coupling, Methanol-to-Olefins, Optimization.

1. INTRODUCTION

The modern chemical industry is facing an increasing demand for light olefins, which are currently in short supply in the global market [1]. The methanol-to-olefin (MTO) technology, which uses methanol as a raw material to produce ethylene and propylene, is competitive based on the data from Chinese market prices [2]. This competitiveness arises because coal constitutes a significant portion of China's energy structure and can be utilized to produce methanol [3].

For the olefin separation process in methanol-to-olefin (MTO) technology, it is essential to remove methane from ethylene. If methane is not removed, it may appear at the top of the ethylene recovery column in the downstream separation process, negatively impacting the purity and yield of ethylene. Some olefin separation processes utilize cryogenic separation of the cracking gas, where components C_1 and C_2 are separated directly using a distillation column. However, this method consumes a significant amount of energy and requires a high investment cost, as the column must be cooled to temperatures below -100 °C [4]. Currently, the separation

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process is increasingly adopting absorption columns with intermediate cooling to reduce energy consumption and costs. Chen et al. [5] conducted a simulation analysis of three pre-depropane processes and found that the amount of absorbent used in the double-column pre-segmentation, oil-absorption system, and the double-section single-column oil-absorption and stripping system was lower than that in a single demethanizer column. To optimize the separation, the separation sequence was properly adjusted. A solvent recovery column was added after the demethanation column, allowing the absorbent to circulate exclusively between the demethanation and solvent recovery columns. Zhang [6] simulated the pre-cut oil-absorption demethanator and analysed how the absorbent influences separation performance. The study revealed that there is an optimal absorbent flow rate for a given reboiler load in the demethanator. Additionally, it is beneficial to maximize the propane concentration in the absorber to minimize olefin loss. Exergy analysis has been widely used to assess energy consumption in chemical industrial processes [7] and can provide a more accurate evaluation of energy usage. However, performing a rigorous exergy analysis requires complex iterative calculations to solve the coupled models of distillation and absorption columns, allowing for the assessment of overall exergy loss.

Distillation and absorption are techniques used to separate mixtures by employing energy and absorbents as separation agents. Exergy analysis can provide a more comprehensive evaluation of the energy and absorbent consumption involved, yet there have been no reports on this in the open literature. To address this gap, we will develop a simplified optimization model for the coupled distillation and absorption columns, which will be utilized to optimize the columns in the MTO (Methanol-to-Olefins) system. In this study, we will determine the relationship among ethylene content, the amount of absorbent used, energy consumption, and exergy loss in the demethanizer column. Additionally, we will establish a rigorous optimization model aimed at minimizing exergy consumption to validate the results of the simplified calculations.

2. SIMPLIFIED MODEL OF THE SYSTEM

For a mixture containing three components (B, C, and E, arranged from lightest to heaviest), component B has an extremely low boiling point, while component C is the primary product. Coupled distillation and absorption columns can effectively separate the mixture without the need for cryogenic refrigeration, as illustrated in **Figure 1**. In this process, the distillate includes a certain proportion of component C, which is further separated by the absorber to recover component C. G is used as the absorbent, and the absorption liquid is returned to the distillation column as reflux.

Both distillation and absorption rely on energy and absorbents to separate mixtures. The top product of the distillation column serves as the feed for the absorber, meaning that the operational parameters of one affect the separation performance and energy consumption of the other, and vice versa. To minimize the overall exergy loss in the system, both columns must be optimized with their interdependence in mind. The composition and flow rate of the distillation column's top product are critical factors influencing the performance of the distillation-absorption system, as they affect utility consumption and exergy loss. These factors are directly related to the operational parameters of the distillation column.

To optimize the distillation-absorption system, rigorous simulation and iterative calculations are required. To simplify the modeling process, qualitative relationships among energy consumption, exergy loss, and the composition of the intermediate stream will be established, and a model will be built to optimize the system and its operational parameters based on the following assumptions:

1) The distillate contains only components B and C;
2) The absorber operates isothermally;
3) The tail gas at the top of the absorption column does not contain any absorbent.

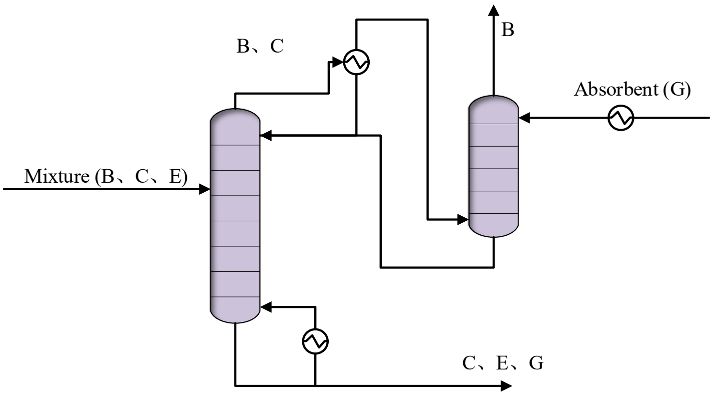

Figure 1. Coupled process of distillation and absorption

2.1 Simplified model of absorption column

The absorber is a column with N theoretical plates, operating under pressure P. The absorbent is introduced at the top stage and makes countercurrent contact with the gas mixture. The absorption factor of component i on tray j is given by eq. (1).

$$A_{i,j} = \frac{l}{m_{i,j} v} \tag{1}$$

where $m_{i,j}$ is the phase equilibrium constant of component i on tray j, which depends on temperature, pressure, and composition; l and v are the flow rates of the liquid and gas on tray j, respectively.

The absorption fraction, absorption factor of any component, and the number of theoretical plates can be related by the Hatton-Franklin equation shown in eq. (2). This equation is theoretical, and it is difficult to solve the relationship among the corresponding factors. eq. (2) can be simplified with the average absorption factor used, instead of calculating the absorption factor at each stage. The modified equation with average absorption factor is shown by eq. (3).

$$\frac{v_{N+1} - v_1}{v_{N+1}} = \frac{A_1 \cdots A_N + A_2 \cdots A_N + \cdots + A_N}{A_1 \cdots A_N + A_2 \cdots A_N + \cdots + A_N + 1} - \frac{l_0}{v_{N+1}} \left(\frac{A_2 \cdots A_N + A_3 \cdots A_N + \cdots + A_N + 1}{A_1 \cdots A_N + A_2 \cdots A_N + \cdots + A_N + 1} \right) \tag{2}$$

$$\frac{A_i^{N+1} - A_i}{A_i^{N+1} - 1} = \phi_i \tag{3}$$

where ϕ_i and A_i are the absorption fraction and average absorption factor of component i.

For a given ϕ_i, A_i can be calculated according to Eq. (3) to obtain the L/V; for a given L/V, ϕ_i can be calculated in the case that N and the average absorption temperature are already known.

2.2 Simplified model of distillation column

The minimum number of theoretical plates can be calculated according to the Fenske equation (eq. (4)).

$$N_{\min} = \frac{\lg\left[\left(x_L / x_H \right)_D / \left(x_L / x_H \right)_W \right]}{\lg \alpha_{LH,av}} \tag{4}$$

where x denotes the component's concentration; subscripts L and H represent the light and heavy key components, while D and W represent the top and bottom products of the column; $\alpha_{LH,av}$ indicates the average relative volatility of light and heavy key components.

In a distillation column with multiple feeds, as shown in **Figure 2**, each feed is associated with a specific minimum reflux ratio. The overall minimum reflux ratio for the entire column is determined by the highest value among these individual ratios, as shown in eq. (5).

$$R_{\min} = \max(R_{\min,1}, R_{\min,2}, \dots, R_{\min,k}) \tag{5}$$

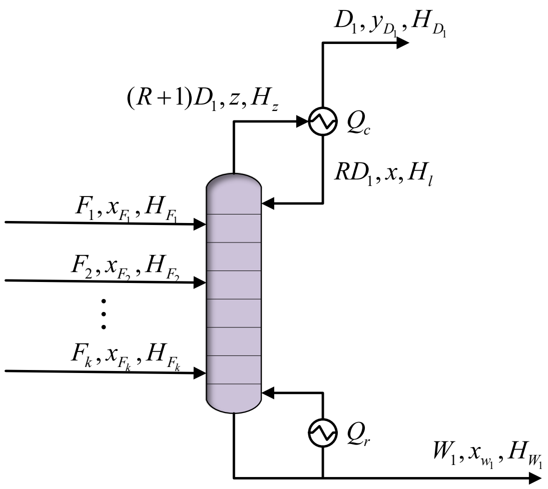

Figure 2. Multi-components distillation column with multiple feeds

The minimum reflux ratio for each feed is determined using a modified Underwood equation [8], as illustrated in eq. (6) and eq. (7).

$$\sum_{i=1}^{c} \frac{\bar{\alpha}_i x_{i,F_j}}{\bar{\alpha} - \theta_j} = 1 - q_j \quad j = 1, 2, \dots, k \tag{6}$$

$$R_{\min,j} + 1 - \sum_{j=1}^{i-1} \left(1 - q_j\right) \frac{F_j}{D} = \sum_{i=1}^{c} \frac{\bar{\alpha}_i}{\bar{\alpha}_i - \theta_j} \left(x_{i,D} - \sum_{j=1}^{i-1} \left(\frac{F_j x_{i,F_j}}{D} \right) \right) \quad j = 1, 2, \dots, k \tag{7}$$

where c is the number of components; $\bar{\alpha}_i$ is the average relative volatility of component i; F_j and q_j are the flow rate and thermal state parameter of j-th feed; x_{i,F_j} and $x_{i,D}$ are the fraction of component i in the j-th feed and distillate product; θ_j is the intermediate parameter.

The actual reflux ratio R can be determined by the minimum reflux ratio. Eq. (8) and eq. (9) are empirical correlations among N, $N_{\min}$, R, and $R_{\min}$ [9].

$$Y = \frac{N - N_{\min}}{N + 1}, \qquad X = \frac{R - R_{\min}}{R + 1} \tag{8}$$

$$Y = 0.75 - 0.75 X^{0.5668} \tag{9}$$

The temperature of the condenser is related to the component of the distillate and can be determined according to the dew point equation. For a partial condenser, the reflux liquid is in equilibrium with the distillate product, and its component and flow rate can be determined according to the phase equilibrium and material balance. Eq. (10) and eq. (11) show the heat balance of the condenser and full column.

$$Q_c + D_1(R+1)H_Z = D_1 H_{D_1} + RD_1 H_l \tag{10}$$

$$Q_c + Q_r + \sum_{j=1}^{k} F_j H_{F_j} = D_1 H_{D_1} + W_1 H_{W_1} \tag{11}$$

where, H_Z, H_{D_1}, and H_l represent the enthalpy of stream in and out (distillate and reflux) of the partial condenser, kJ/kmol; H_{F_j} and H_{W_1} represent the enthalpy of stream in and out of the column, kJ/kmol.

2.3 Exergy analysis

Exergy is the maximum amount of useful work produced by a process from the current state to the environment state. A stream's exergy can be calculated by eq. (12) [10].

$$E_x = (H - H_0) - T_0(S - S_0) \tag{12}$$

where, H denotes molar enthalpy, kJ/kmol; S represents molar entropy, kJ/kmol/K; T_0 indicates the environmental temperature, K; H_0, S_0 denote enthalpy and entropy in the environmental state; E_x represents exergy, kJ/kmol.

The minimum work required by the separation equals to the difference between the exergy of the output streams and that of the input stream, as shown by eq. (13).

$$W_{\min} = \sum\nolimits_{out} fE_x - \sum\nolimits_{in} fE_x \tag{13}$$

where, f denotes the molar flow rate of feed or product stream, kmol/h; $W_{\min}$ indicates the minimum amount of work, kW.

For distillation column, heat (Q_r, T_r) is input into the reboiler at the bottom and removed (Q_c, T_c) through a condenser at the top. The exergy of utility can be calculated based on eq. (14).

$$E_{xQ} = Q\left(1 - T_0 / T\right) \tag{14}$$

where, E_{xQ} denotes the exergy of a heat source, kW; Q represents the heat duty, kW; T indicates the temperature of the heat source, K.

For a completely reversible separation process, the minimum work required by the separation equals the work input from the utilities. As for the actual distillation and absorption process, exergy loss is inevitable. The overall exergy loss can be calculated by eq. (15).

$$\Delta E_x = Q_r\left(1 - T_0 / T_r\right) + Q_c\left(1 - T_0 / T_c\right) + Q_{cooler}\left(1 - T_0 / T_{cooler}\right) - W_{\min} \tag{15}$$

where Q_r, Q_c, and Q_{cooler} are the heat duties of the reboiler, condenser, and cooler, respectively, kW; T_r, T_c, and T_{cooler} are the utility temperatures of the reboiler, condenser, and cooler, respectively, K.

Figure 3 shows that the calculation procedure of the exergy loss based on simplified models. According to this procedure, the operating parameters of the distillation column, the amount of absorbent, and the duty of each heat exchanger can be obtained when the light component impurities in the feed ($\sum F_B$) are completely removed from the top of the absorption column.

To calculate energy consumption and exergy loss, the flowrate and composition of products must be known. In this calculation procedure, the key is to obtain the absorption fraction of component B based on the simplified absorption mechanism and material balance, and obtain the gas phase product flowrate at the top of the distillation column. Then, the flow rate and composition of each stream can be obtained based on the material balance of these two columns, respectively.

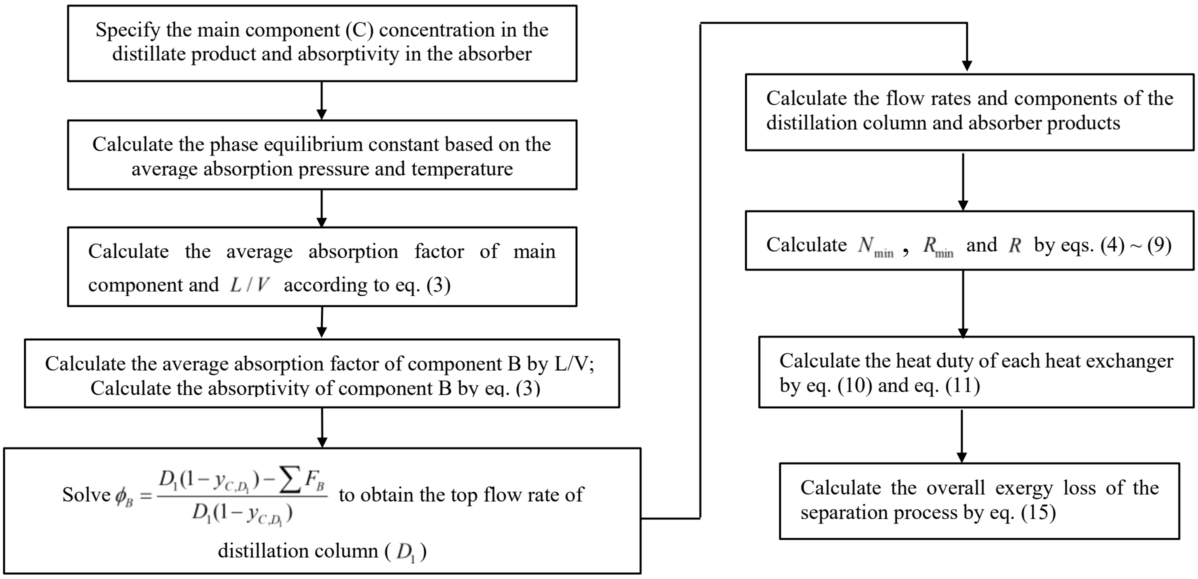

Figure 3. Flowchart for simplified calculation

Using this simplified model, the influence of key parameters in adjacent distillation and absorption columns can be analysed based on the specified parameters of each column, allowing for the determination of optimal operating parameters and suitable utilities.

3. CASE STUDY

In the olefin separation process of an MTO plant, the feedstock contains methane, ethylene, ethane, propylene, propane, butene, dimethyl ether (DME), and other components. It is divided into gas and liquid products, which are sent to the third and fifth plates of the demethanizer, respectively, to remove methane. The distillate of the demethanizer contains some ethylene and is further separated in the absorption column. The propane absorbent is inlet into the absorption column after cooling to -37 °. The ethylene-rich absorption liquid circulates into the demethanizer, and methane leaves the absorption column from the top. The number of trays, feed tray positions, and operating pressures of the distillation and absorption columns are shown in **Table 1**. The compositions and flow rates of feed streams are shown in **Table 2**. The performance of the demethanation column directly affects the content of light components in the final ethylene product.

Table 1. Parameters of distillation and absorption columns

Column	Number of trays	Feed tray	Operating pressure [MPa]
Demethanizer	16	1/3/5	2.9
Absorber	10	1/10	2.75

Table 2. Feed compositions and flow rates of distillation column

Components	Methane	Ethylene	Ethane	Propylene	Propane	DME	Flow rate [kmol/h]
Molar fraction of gas	0.6558	0.3035	0.0044	0.034	0.0023	0	0
Molar fraction of liquid	0.0551	0.5231	0.0116	0.379	0.0307	0.0002	0.0003

The methane in the bottom product of the demethanizer will be sent to the top of the ethylene distillation column in the subsequent separation process, and the methane content in the bottom product of the demethanizer should be strictly controlled. For the ethylene distillation column, the top temperature and pressure are -37 °C and 1.6 MPa, respectively. The ethylene product is extracted from the sideline; methane and part of the ethylene are emitted from the top. The higher the methane content in its feed, the more ethylene is lost from the top. The amount of methane in the feed is related to the ethylene recovery rate. In the top product, the molar fraction of methane (y_{CH_4}) is 0.0542, and the methane content in the bottom product can be determined by Eq. (16).

$$n_{CH_4} = F_{C_2H_4}(1-\varphi_{C_2H_4})y_{CH_4} / (1-y_{CH_4}) \tag{16}$$

where $\varphi_{C_2H_4}$ is the ethylene recovery rate in the ethylene rectifying column; $F_{C_2H_4}$ is the ethylene flow rate in the ethylene distillation column feed. For the absorption column, the ethylene loss rate in the tail gas is specified as D_{2,C_2H_4}=2 kmol/h.

The demethanizer's exergy loss and the absorbent flowrate change along with the ethylene concentration in the intermediate stream. And the ethylene concentration in the distillate is related to the reflux ratio and distillate flow rate. The simplified procedure mentioned above can be used to analyse their relation, and the result can be used as the initial value of the rigorous model.

With the ethylene loss flow rate in the tail gas of the absorber specified, the ethylene absorptivity varies with the ethylene content of the absorber's gas feed. The corresponding absorptivity can be obtained based on the procedure shown in **Figure 3**. The phase equilibrium constant is calculated based on the average temperature of the absorption column, considering the ethylene concentration and the feed temperature at the bottom. The phase equilibrium constant used in the calculation and the absorptivity obtained are shown in **Table 3**.

Table 3. Phase equilibrium constant and absorptivity

Ethylene concentration	Top temperature of the demethanizer [°C]	Phase equilibrium constant		Absorptivity	
		Methane	Ethylene	Methane	Ethylene
0.30	-57.5	3.288	0.702	0.291	0.988
0.35	-53.3	3.353	0.725	0.305	0.990
0.40	-49.5	3.409	0.748	0.318	0.992
0.45	-45.9	3.459	0.770	0.332	0.994
0.50	-42.5	3.502	0.792	0.347	0.995
0.55	-39.3	3.540	0.814	0.362	0.996
0.60	-36.2	3.574	0.835	0.378	0.997

With the environmental temperature taken as 25 °C and the temperature differences of the heater and the cooler taken as 5 °C and 3 °C, respectively, the distillation-absorption columns are analysed based on the procedure shown in **Figure 3**. The result is shown in **Figure 4**. The results show that the flow rate of the distillate at the top of the demethanizer increases with the ethylene concentration at the top. A greater distillate flow rate of demethanizer is beneficial, as the methane content at the bottom of the distillation column and the corresponding reflux ratio will be reduced. However, due to the increase in temperature and ethylene concentration at the top of the demethanation column, the amount of absorbent will increase.

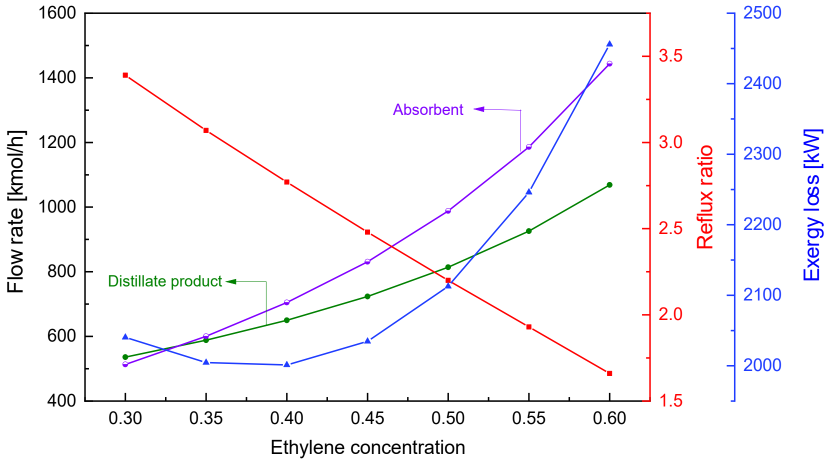

Figure 4. The result of the simplified models

Based on MESH equations and SRK equations, rigorous models of distillation and absorption columns are established on MATLAB software [11]. Figure 5 shows the calculation procedure for the rigorous models. When the rigorous model is used, the molar concentration of ethylene in the gas product at the top of the demethanizer cannot be set easily; only the reflux ratio and the flowrate of the distillate can be specified. In order to analyse the variations of energy consumption, the amount of absorbent and the minimum overall exergy loss along with the ethylene concentration, the reflux ratio and flow rate of the distillate product are taken as decision variables, and the requirement on ethylene concentration is taken as the constraint, the results corresponding to different ethylene concentrations is obtained based on the proposed model. In this process, reflux ratio and distillate flowrate in simplified calculation results are taken as initial values. The result is shown in **Figure 6** and **Figure 7**.

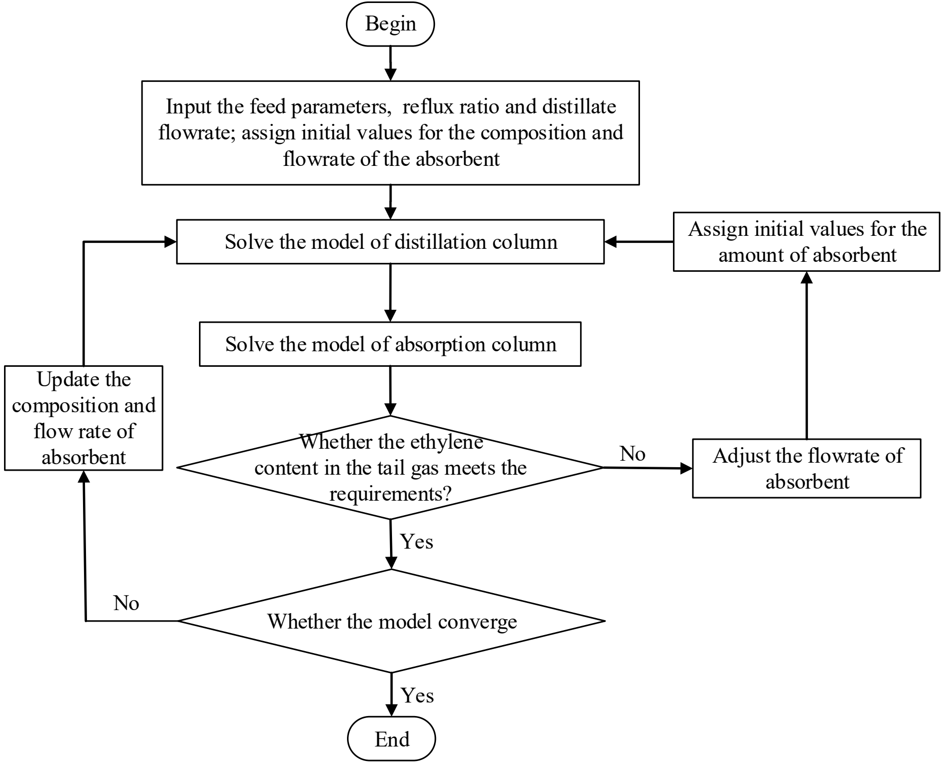

Figure 5. Flowchart for rigorous calculation

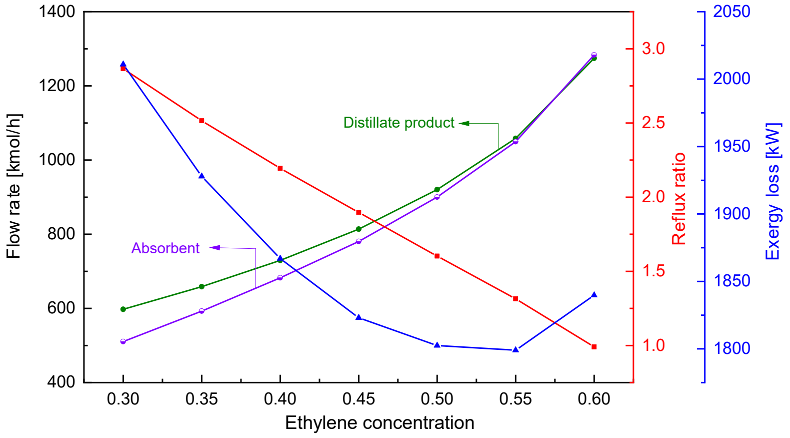

Figure 6. The result of the rigorous models

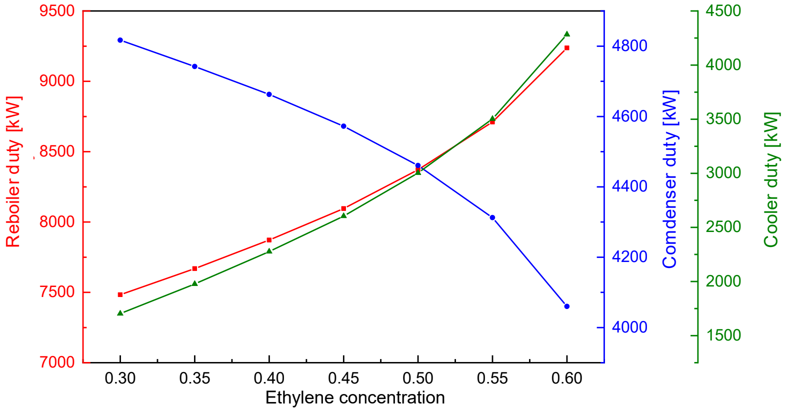

Figure 7. Energy consumption of each heat exchanger

In **Figure 4** and **Figure 6**, the exergy loss curves have the same trend and show that, along with the increment of ethylene concentration at the top of the demethanizer, exergy loss decreases first and then increases. This indicates an optimal ethylene concentration at the top of the demethanizer. The reason for the decrease in exergy loss is that the top temperature of the demethanizer rises along the ethylene concentration and results in the increment of the refrigerant temperature. The exergy loss increases again due to the high ethylene concentration leading to a sharp increase in absorbent and the increment in the condenser and reboiler's duty. The exergy loss is almost the same when the ethylene concentration at the top is 0.5 and 0.55. As shown in **Figure 7**, the higher the concentration, the more the overall energy consumption. Thus the optimal value of 0.5 is appropriate, and the corresponding reflux ratio and distillate flow rate are 1.6029 and 902.3 kmol/h, respectively.

4. CONCLUSIONS

In this paper, a simplified model of combined absorption and distillation columns is presented to analyse their performance simultaneously. This model can determine the operating parameters and energy consumption of both devices without requiring a complicated iterative process. For the demethanizer and the ethylene recovery column in the MTO process, the qualitative relationship between energy consumption and the composition of intermediate streams is identified using the proposed method.

Furthermore, the rigorous model, with the parameters calculated by the simplified model taken as the initial value, converges efficiently. The ethylene molar concentration at the top of the distillation column was optimized to minimize exergy loss, finding the optimal concentration to be 0.5. The corresponding reflux ratio and flow rate of the distillate are 1.6029 and 902.3 kmol/h, respectively. The condenser temperature is -34.6 °C, with propylene refrigerant employed to ensure the regular operation of the demethanizer. The heat duties are as follows: reboiler at 8,371.5 kW, condenser at -4,460.5 kW, and absorbent cooler at -3,003.0 kW.

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NOMENCLATURE

Symbols

A_i	average absorption factor of component i	
c	number of components	
E_x	exergy	[kJ/kmol]
E_{xQ}	exergy of a heat source	[kW]
$F_{C_2H_4}$	ethylene flow rate in the ethylene distillation column feed	[kmol/h]
F_j	the flow rate of j-th feed	[kmol/h]
f	molar flow rate	[kmol/h]
H	molar enthalpy	[kJ/kmol]
H_0	enthalpy in the environmental state	[kJ/kmol]
H_{D_1}	the enthalpy of diatillate	[kJ/kmol]
H_{F_j}	the enthalpy of stream inlet the column	[kJ/kmol]
H_l	the enthalpy of reflux	[kJ/kmol]
H_{W_1}	the enthalpy of stream outlet the column	[kJ/kmol]
H_Z	the enthalpy of stream inlet the partial condenser	[kJ/kmol]
l	flow rate of the liquid on tray j	[kmol/h]
$m_{i,j}$	phase equilibrium constant of component i on tray j	
Q	heat duty	kW
q_j	thermal state parameter of j-th feed	
S	molar entropy	[kJ/kmol/K]
S_0	entropy in the environmental state.	[kJ/kmol/K]
T	temperature of heat source	[K]
T_0	environmental temperature	[K]
$W_{\min}$	minimum amount of work	[kW]
x	component's concentration	
$x_{i,D}$	fraction of component i in the distillate product	
x_{i,F_j}	fraction of component i in the j-th feed	

Greek letters

$\bar{\alpha}_i$	average relative volatility of component i	
$\alpha_{LH,av}$	average relative volatility of light to heavy key components	
θ_j	the intermediate parameter	
v	flow rate of the gas on tray j	[kmol/h]
ϕ_i	absorption fraction of component i .	
$\varphi_{C_2H_4}$	ethylene recovery rate in ethylene rectifying column	

Subscripts and superscripts

D	top product of the column
H	heavy key component
L	light key component
W	bottom product of the column
c	condenser
r	reboiler
$cooler$	cooler

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