



Separation Science and Engineering

Performance Prediction of Structured Packing Column for Cryogenic Air Separation with Hybrid Model[☆]Xiaobin Zhang, Jiakai Zhu, Zhao Wu, Wei Xiong, Xuejun Zhang, Limin Qiu^{*}

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ARTICLE INFO

Article history:

Received 29 May 2013

Received in revised form 24 July 2013

Accepted 10 December 2013

Available online 17 June 2014

Keywords:

Distillation

Cryogenic air separation

Structured packings

Hybrid model

Aspen

ABSTRACT

A detailed investigation of a thermodynamic process in a structured packing distillation column is of great importance in prediction of process efficiency. In order to keep the simplicity of an equilibrium stage model and the accuracy of a non-equilibrium stage model, a hybrid model is developed to predict the structured packing column in cryogenic air separation. A general solution process for the equilibrium stage model is developed to solve the set of equations of the hybrid model, in which a separation efficiency function is introduced to obtain the resulting tri-diagonal matrix and its solution by the Thomas algorithm. As an example, the algorithm is applied to analyze an upper column of a cryogenic air separation plant with the capacity of 17000 m³ h⁻¹. Rigorous simulations are conducted using Aspen RATEFRAC module to validate the approach. The temperature and composition distributions are in a good agreement with the two methods. The effects of inlet/outlet position and flow rate on the temperature and composition distributions in the column are analyzed. The results demonstrate that the hybrid model and the solution algorithms are effective in analyzing the distillation process for a cryogenic structured packing column.

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1. Introduction

Cryogenic air separation is now the most economical approach to separate oxygen (O₂) and nitrogen (N₂) from the air on a large scale, which can provide customized products of different purity by changing the process [1]. Column is a key unit for the cryogenic air separation with high power consumption. Compared with traditional random-packing columns, structured-packing column (SPC) has the merits of greater capacity, smaller pressure drop and higher separation efficiency [2,3]. In order to improve the efficiency of structured packings and reduce the initial investment, the design of the distillation process needs to be optimized.

Two strategies are usually used to model the distillation in a column, equilibrium-stage (EQ) model and non-equilibrium stage (NEQ) model [4,5]. The EQ model is widely used, which assumes thermodynamic equilibrium between bulk phases, with an empirical efficiency correlation, such as the Murphree efficiency, to offset the difference between calculations and practical conditions. The accuracy of the correlations for different stages under different thermodynamic conditions is questionable [6]. Comparably, the NEQ model is more accurate since it treats the separation process as a mass-transfer-rate-governed one that it really is [7]. Instead of the assumption of thermodynamic equilibrium between bulk phases, the thermodynamic equilibrium is only assumed in a thin interfacial zone. The heat and mass transfer between a bulk phase

and interfacial zone is determined by empirical correlations [8,9]. Seader and Henley have pointed out that the NEQ model and its solution lead to a new era in separation equipment design and simulation [10]. However, since the NEQ model has much more non-linear equations, it is difficult to obtain converged solutions. As a result, its application to large-scale industrial distillation processes is limited. Additionally, calculations of empirical heat and mass transfer coefficients significantly increase the uncertainty of the solution. Good initialization is necessary to obtain final converged solutions [11,12]. In fact, the turbulence-intensified heat exchange between phases is much faster than the mass transfer, so it is reasonable to neglect the thermal non-equilibrium in the NEQ model. The reduced NEQ model is called hybrid model [12,13]. Tang and Wu [12] have evaluated the hybrid model by modeling a separation process for methanol/ethanol/*n*-propanol ternary mixture in SPC. The application of the model to cryogenic SPC for air separation has not been reported.

This study develops a hybrid model to obtain the solution of the distillation process in SPC for cryogenic air separation. We consider the pressure drop and mass transfer resistance in the liquid phase. The model equations are turned into a tri-diagonal matrix by introducing a separation efficiency function and solved by the Thomas algorithm, instead of the complex Newton iteration method used in [12], so that the calculations of partial derivatives of thermodynamic functions are not needed. A cryogenic upper column of an air separation plant with the capacity of 17000 m³ h⁻¹ is considered, with O₂ and N₂ components only in the air mixture. The model is validated by the temperature and component distributions with the rigorous simulations using the Aspen RATEFRAC module. Finally, the effects of inlet/outlet positions and flow rates on the distillation process are analyzed.

[☆] Supported by the Major State Basic Research Development Program of China (2011CB706501) and the National Natural Science Foundation of China (51276157).

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2. Model and Algorithm

Commercially available SPCs are comprised of identical n layers of structured packing. Each packing layer is rotated 90° with respect to the previous one. Each layer is an ensemble of a large number of corrugated sheets as shown in Fig. 1, forming a triangular flow channel with dimensions of height (h), side (s), and base (b), as well as the corrugation angle α with respect to the horizontal. Two adjacent corrugated sheets are superimposed so that the opposite corrugations form a cross-type pattern with the crests of the corrugations nearly in contact.

Each layer can be considered as one stage as in traditional plate columns, where the bulk phases are homogeneous. The column has n stages. On each stage, it is assumed that $T_j^L = T_j^I = T_j^V$, where T is the temperature, superscripts L, I, and V represent the liquid phase, interface and vapor phase, respectively, and subscript j stands for the j th stage of total n . The following assumptions are also made for the calculations: (a) pressure in equilibrium, $P_j^L = P_j^I = P_j^V$; (b) no component and temperature gradient in the radial direction; and (c) phase equilibrium only in the gas/liquid interfacial zone. Fig. 2 shows the model on the j th stage, which involves gas-phase feed rate F_j^V , liquid-phase feed rate F_j^L , molar flow rate L , molar fractions of component i in the liquid phase x_i and in the gas phase y_i , specific enthalpy h per mole, temperature T , exhausting gas S_j^V and liquid S_j^L , mass flux $N_{i,j}$ and heat flux $e_{i,j}$ between phases, here $e_{i,j} = 0$ for our calculations.

Based on the above assumptions, the set of control equations is exactly the same as that of the conventional NEQ model [10] without the heat transfer equations.

Material balance equations for the component:

$$M_{i,j}^V \equiv (V_j + S_j^V)y_{i,j} - V_{j-1}y_{i,j-1} - F_j^V z_{i,j}^V + N_{i,j}^V = 0, \quad i = 1, 2, \dots, c \quad (1)$$

$$M_{i,j}^L \equiv (L_j + S_j^L)x_{i,j} - L_{j+1}x_{i,j+1} - F_j^L z_{i,j}^L - N_{i,j}^L = 0, \quad i = 1, 2, \dots, c. \quad (2)$$

Material balance equation at the phase interface:

$$M_{i,j}^I \equiv N_{i,j}^V - N_{i,j}^L = 0, \quad i = 1, 2, \dots, c. \quad (3)$$

Energy balance equations without the heat lost:

$$E_j^V \equiv (V_j + S_j^V)H_j^V - V_{j-1}H_{j-1}^V - F_j^V H_{F,j}^V = 0 \quad (4)$$

$$E_j^L \equiv (L_j + S_j^L)H_j^L - L_{j+1}H_{j+1}^L - F_j^L H_{F,j}^L = 0. \quad (5)$$

The equations for the molar-fraction summation for each phase are applied at the vapor–liquid interphase:

$$\sum_{i=1}^c x_{i,j}^L - 1 = 0, \quad \sum_{i=1}^c y_{i,j}^V - 1 = 0. \quad (6)$$

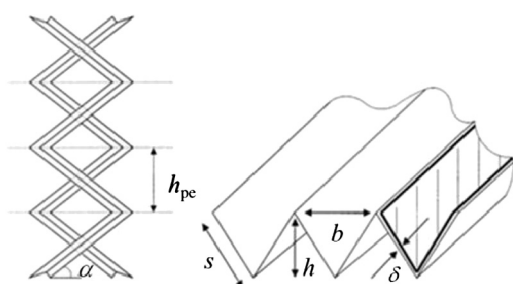


Fig. 1. Basic geometry of the packing.

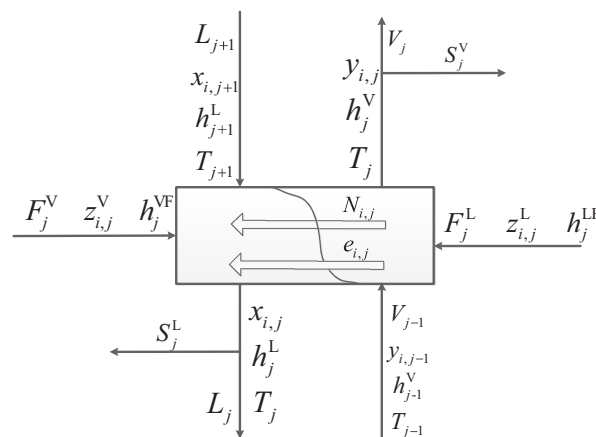


Fig. 2. Schematic diagram of stage j .

The hydraulic equation for stage pressure drop is given by

$$P_{j+1} - P_j - \Delta P_j = 0 \quad (7)$$

where ΔP_j is the pressure drop at stage j .

Phase equilibrium for each component is assumed to exist only at the interphase:

$$y_{i,j}^I - K_{i,j}x_{i,j}^I = 0 \quad (8)$$

where $K_{i,j}$ is the phase equilibrium constant. The general forms for mass-transfer rates of component i across vapor and liquid films on a stage are as follows

$$N_{i,j}^V \equiv k_{i,j}^V a_j v_j (y_{i,j} - y_{i,j}^I) + y_{i,j} N_{t,j} \quad (9)$$

$$N_{i,j}^L \equiv k_{i,j}^L a_j v_j (x_{i,j}^I - x_{i,j}) + x_{i,j} N_{t,j} \quad (10)$$

where $k_{i,j}^V$ and $k_{i,j}^L$ are the mass transfer coefficients ($\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$), a_j is the effective mass transfer area at stage j ($\text{m}^2 \cdot \text{m}^{-3}$), and v_j is the volume of stage j (m^3).

Combining Eqs. (8) and (3) with Eqs. (9) and (10), we have

$$x_{i,j}^L = \frac{k_{i,j}^L a_j x_{i,j} + k_{i,j}^V a_j y_{i,j} + (y_{i,j} - x_{i,j}) \sum_{i=1}^c N_{i,j}^V}{k_{i,j}^L a_j + k_{i,j}^V a_j K_{i,j}}. \quad (11)$$

Substituting Eq. (11) into Eqs. (8) and (9), and relating it with Eq. (1), the 'separation efficiency function', which shows the relationship between vapor and liquid compositions, we have

$$x_{i,j} = \alpha_{i,j} y_{i,j} - \beta_{i,j} y_{i,j+1} - \gamma_{i,j} \quad (12)$$

$$\alpha_{i,j} = \frac{k_{i,j}^L k_{i,j}^V a_j + k_{i,j}^L \sum_{i=1}^c N_{i,j}^V + (k_{i,j}^V K_{i,j} + k_{i,j}^L) (V_j + S_j^V)}{k_{i,j}^L k_{i,j}^V a_j K_{i,j} - k_{i,j}^V K_{i,j} \sum_{i=1}^c N_{i,j}^V} \quad (13)$$

$$\beta_{i,j} = \frac{(k_{i,j}^V K_{i,j} + k_{i,j}^L) V_{j+1}}{k_{i,j}^L k_{i,j}^V a_j K_{i,j} - k_{i,j}^V K_{i,j} \sum_{i=1}^c N_{i,j}^V} \quad (14)$$

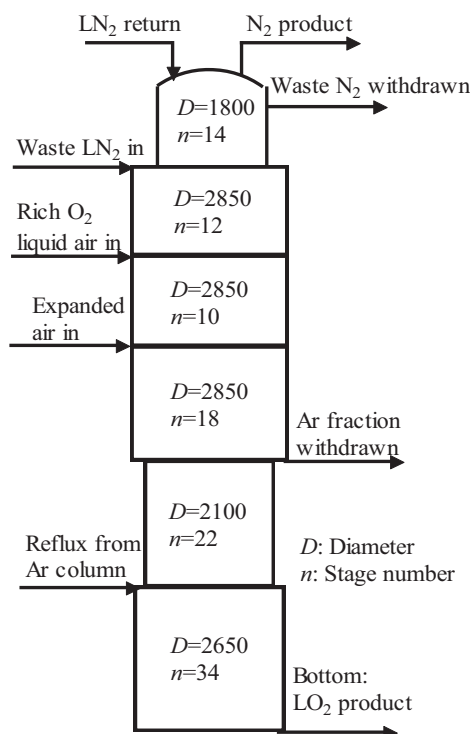


Fig. 3. Schematic diagram of the upper column of capacity of 17000 m³h⁻¹.

$$\gamma_{i,j} = \frac{(k_{i,j}^V K_{i,j} + k_{i,j}^L) F_j^V z_{i,j}^V}{k_{i,j}^L k_{i,j}^V a_j K_{i,j} - k_{i,j}^V K_{i,j} \sum_{i=1}^c N_{i,j}^V} \quad (15)$$

Substituting Eqs. (12)–(14) into Eqs. (1) and (2) leads to

$$A_{i,j-1} y_{i,j-1} - A_{i,j} y_{i,j} + A_{i,j+1} y_{i,j+1} = L_{j-1} \gamma_{i,j-1} - F_j^V z_{i,j}^V - F_j^L z_{i,j}^L - (L_j + S_j^L) \gamma_{i,j} \quad (16)$$

where

$$\begin{aligned} A_{i,j-1} &= L_{j-1} \alpha_{i,j-1}, \quad A_{i,j} = L_{j-1} \beta_{i,j-1} + (V_j + S_j^V) + (L_j + S_j^L) \alpha_{i,j}, \\ A_{i,j+1} &= V_{j+1} + (L_j + S_j^L) \beta_{i,j}. \end{aligned} \quad (17)$$

Eq. (16) is a tri-diagonal matrix of vapor composition and can be solved with the Thomas algorithm.

3. Comparison and Validation

To validate the proposed algorithms for the distillation process, a practical upper column for cryogenic air separation with the capacity of 17000 m³h⁻¹ is taken as an example. The upper column has seven imports and four exports, with different flow rates of gas and liquid phases in different segments. The column is divided into six segments with different inner diameters, as shown in Fig. 3. The upper column is coupled to the lower column through the evaporator–condenser between them. Table 1 gives the parameters of these exports and imports. For simplicity, the air is considered as a binary mixture of O₂ and N₂, and the argon (Ar) fraction is added to the oxygen fraction, so the molar fractions of oxygen and nitrogen are 21.9% and 78.1%, respectively. The column uses Mellapak 750 structured packing, whose geometrical parameters are shown in Table 2.

The set of equations is solved numerically using the FORTRAN compiler. The software Refprop8.0 [14] is called as subroutines to calculate the physical properties as well as the phase equilibrium, which is based on the Helmholtz free energy concept. The values of $k_{i,j}^V$, $k_{i,j}^L$, a_j , and pressure drop ΔP for the structured packing are calculated through the empirical correlations in literature [15–17].

The numerical method is validated by comparing the results with the rigorous calculations of the software Aspen plus. Both the equilibrium and non-equilibrium models are used with the RATEFRAC module. The Peng–Robinson equation of state is chosen for the calculation of thermodynamic quantities, and the pressure drop is determined by the expression developed by Sultz Corp. For calculations of $k_{i,j}^V$, $k_{i,j}^L$ and a_j , the model developed by Hanley is adopted, which correlates experiments especially for the application of Mellapak serial SPC [18]. The heat transfer coefficient is determined by the model from the Chilton–Colburn analogy by Taylor and Krishna [19].

Fig. 4 shows that the distributions of N₂ and O₂ calculated with the two methods are in good agreement. The results from the hybrid model are between the values from Aspen EQ and NEQ models, implying that the proposed algorithm is effective in solving the equations. The temperature profiles of the hybrid model and that by the Aspen also match well, as shown in Fig. 5. The Aspen simulation shows little difference between the temperatures of vapor and interface, because of the large heat exchange coefficient, validating the assumption of thermal equilibrium. The temperature distribution with Aspen NEQ is smoother than that with the hybrid method, which is similar in the composition distribution as shown in Fig. 4.

Table 1
Boundary conditions of the upper distillation column

	Position (n)	Phase	x_{O_2}	x_{N_2}	Flow rate /mol·s ⁻¹
LN ₂ return	1	L	0.000158	0.9998420	275.107
Waste N ₂ withdrawn	3	G	0.013593	0.986407	583.495
Waste LN ₂ in	15	L	0.186878	0.813122	215.302
Rich O ₂ liquid air in	27	L	0.414313	0.585687	64.591
Expanded air in	36	G	0.21825	0.78175	112.396
Liquid air from below column	37	L	0.385288	0.614712	276.791
Ar fraction withdrawn	55	G	0.99983	0.00017	233.056
Reflux from Ar column	77	L	0.999954	0.000046	224.345
Bottom		L	1	0	681.46

Table 2
Dimensions of Mellapak 750

Specific area a_p /m ² ·m ⁻³	Corrugation angle α / (°)	Void fraction ε	Crimp height h /mm	Base b /mm	Side s /mm	Element height h_{pe} /mm
750	45	0.925	4	7.2	5.4	212

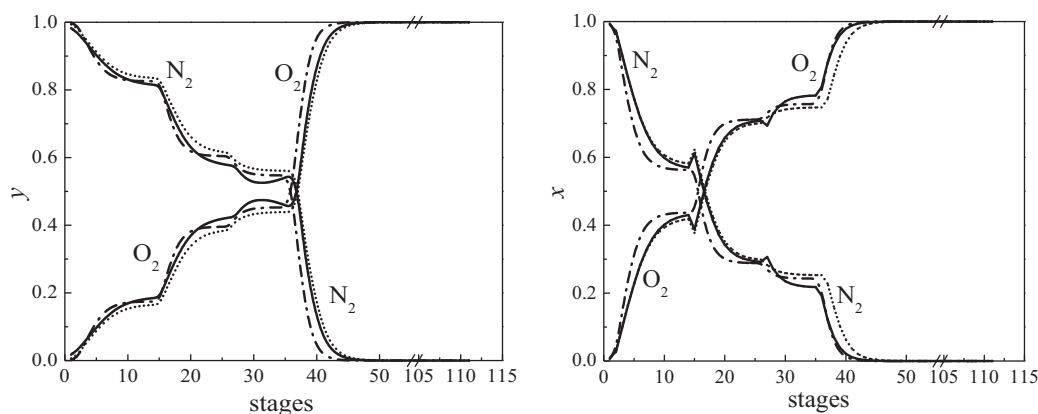


Fig. 4. Composition profiles in vapor and liquid phases using Aspen NEQ, EQ and hybrid models. Aspen NEQ model; - - - Aspen EQ model; — hybrid model.

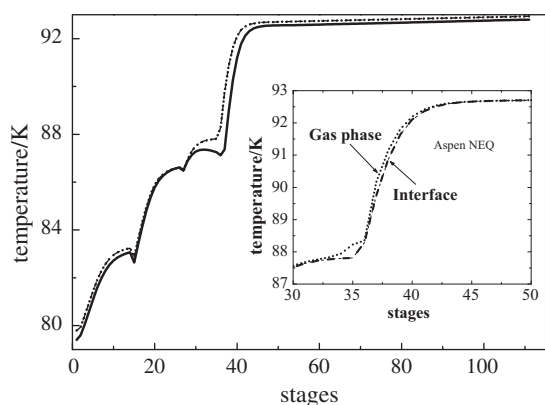


Fig. 5. Temperature distributions using Aspen NEQ and hybrid models. Aspen NEQ model; — hybrid model.

The calculated distributions of components and temperature are almost unchanged when the stage number is larger than about 40, which means that the distillation process is very weak in these stages. The reason is attributed to the binary assumption, where O_2 and Ar are considered as one component, while their separation occurs on these stages. It is confirmed by the calculation of Aspen NEQ for the O_2 – N_2 –Ar ternary mixture, while keeping other boundary conditions unchanged, as shown in Fig. 6. For $n < 55$, the component distributions are almost the same as that with binary mixture calculations. For $n > 55$, N_2 fraction is almost unchanged and O_2 fraction increases, while Ar fraction increases first and then decreases.

4. Analysis and Discussion

Figs. 5 and 6 show that the curves of component and temperature present three “steps” at corresponding feed locations. Table 3 compares the given values of N_2 fraction at the inlets/outlets with those calculated on the corresponding stages, and obvious deviations are observed. The mismatches will induce large exergy loss and energy consumption because of the large difference in concentration and temperature. Therefore, it is significant to validate the effects of inlet/outlet flow rates and positions on the composition and temperature distributions.

4.1. Effects of inlet/outlet flow rates

Figs. 7 and 8 give the typical distributions of temperature and gas molar fraction with $\pm 5\%$ variations in flow rates of LN_2 , waste GN_2 withdrawn, rich O_2 liquid air in, and expanded air in. The curves of the base flow rates are always between those with higher and lower flow rates, implying that the solutions of the present model are robust and reasonable. The effects of variations in Ar fraction withdrawn and reflux flow rates are not investigated due to the assumption of binary mixture. The temperature distribution has different sensibility to the rate change of inlets/outlets. LN_2 reflux rate, waste LN_2 in, waste LN_2 withdrawn, and liquid air reflux rate bring about large temperature changes, especially for the stages they are connected. The reasons are as follows. Firstly, the base flow rates of these inlets/outlets are large, so the absolute values of $\pm 5\%$ are also large. Secondly, N_2 fraction is relatively large at these inlets/outlets, whose boiling point is the lowest compared with O_2 component, so it is more sensitive to temperature. It is also found that the N_2 fraction

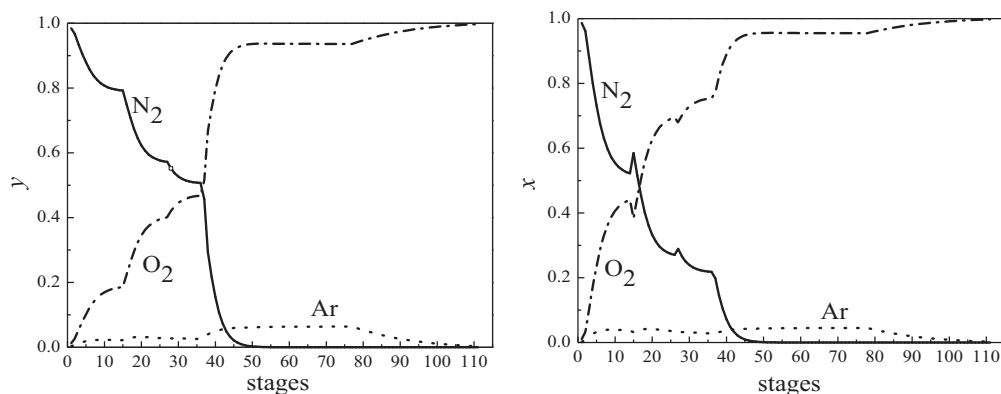


Fig. 6. Calculated composition profiles in vapor and liquid phases with O_2 – N_2 –Ar ternary mixture using Aspen NEQ.

Table 3
N₂ concentration at inlet/outlet conditions and at the same stages

	1	3	15	27	36	37	55	77
Calculations	0.991461	0.957356	0.611987	0.306753	0.461123	0.121792	1.60×10^{-6}	5.90×10^{-7}
Given values	0.999842	0.986407	0.813122	0.585687	0.78175	0.614712	0.00017	0.000046

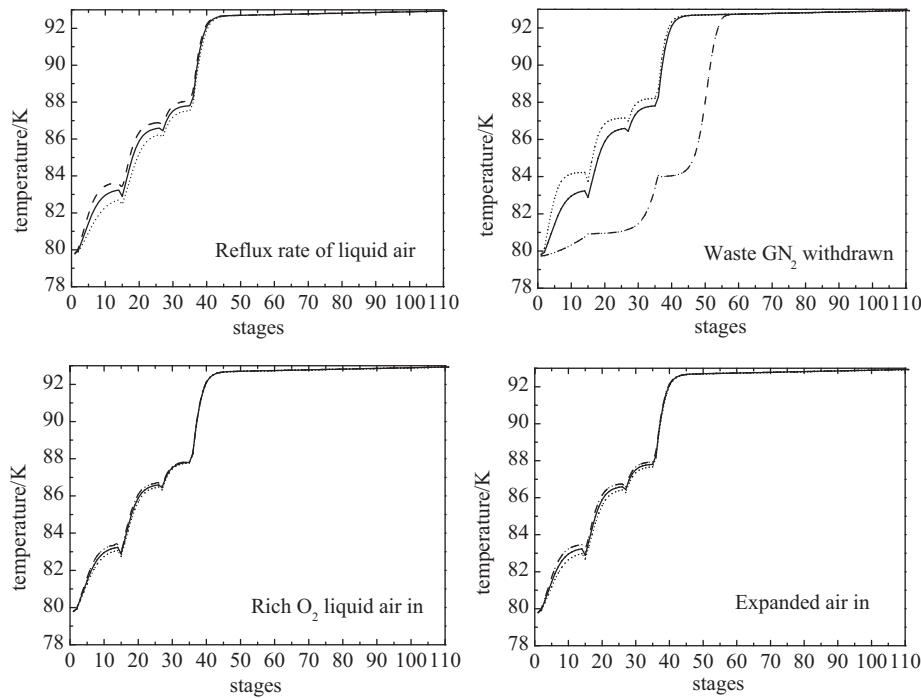


Fig. 7. Effects of variances in inlet/outlet flow rates on temperature distributions. — base flow; +5%; --- -5%.

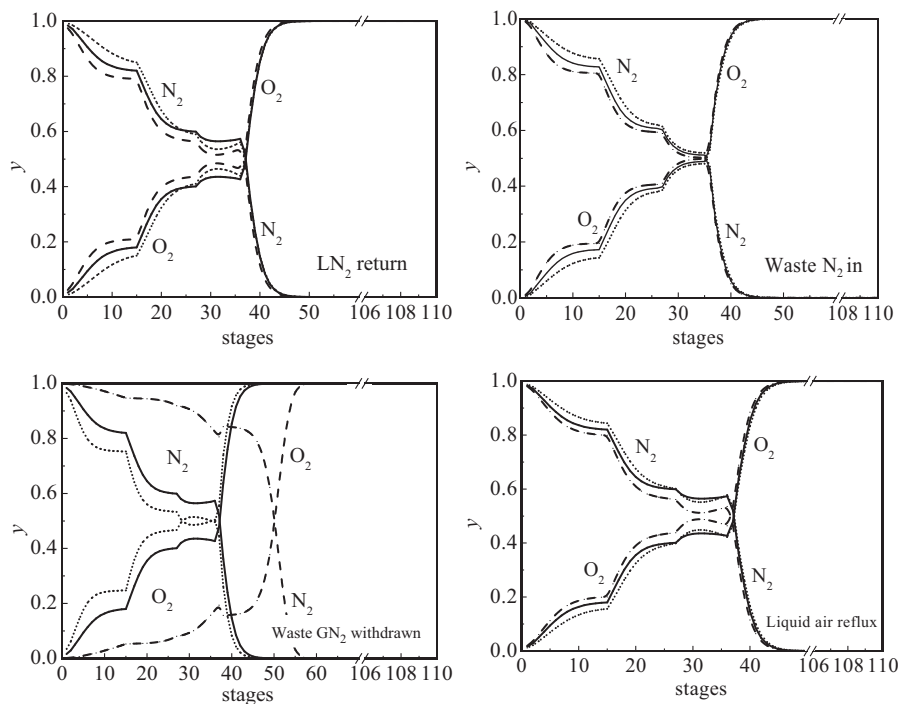


Fig. 8. Effects of variances in inlet/outlet flow rates on the component distributions in gas phase. — base flow; +5%; --- -5%.

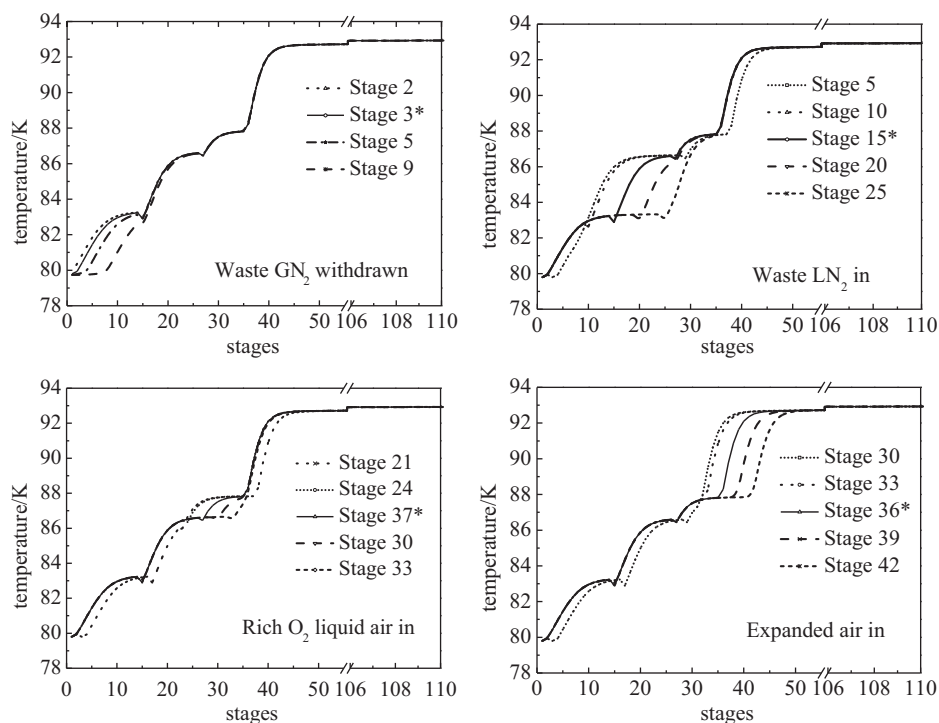


Fig. 9. Effects of variances in inlet/outlet positions on the temperature distribution.

of GN_2 products withdrawn at the top of the column will increase with the increases of flow rate of LN_2 , waste LN_2 in and reflux rate of liquid air, while it decreases with the increases of flow rate of the withdrawn waste GN_2 .

4.2. Effects of inlet/outlet positions

Fig. 9 shows the effects of positions of waste GN_2 withdrawn, waste LN_2 in, rich O_2 liquid air in and expanded air in on the temperature distribution, in which the symbol * means the original inlet/outlet position listed in Table 1. The curves of the base position are always between those with more and less stages. The effect is more significant near the position and is negligible far away from it. The horizontal segments of the curves indicate no energy exchanges on these stages. Therefore, the mass transfer is also restricted, which is not suitable for practical operations of SPC because these stages do nothing for air separation. From this viewpoint, the original inlet/outlet positions listed in Table 1 seem to be acceptable since the length of the horizontal segments of temperature curves is shorter compared with other cases.

For the effects of positions on the component distribution, a typical relation is given in Fig. 10. As the stage number of rich O_2 liquid air in

increases (moves downward), N_2 fraction in both phases at the same stage decreases and then changes slowly. The inlet/outlet position is adjustable in a range in which N_2 fraction changes little. The effects of positions of waste GN_2 withdrawn and waste LN_2 in on the N_2 fraction are also investigated, and the curves are similar to that in Fig. 10. The inlet/outlet positions listed in Table 1 are near the appropriate range, so they are acceptable.

5. Conclusions

Based on the operating characteristics of structured packing, a hybrid model was developed and solved by the Thomas algorithm to analyze a column of a cryogenic air separation plant with the capacity of $17000 \text{ m}^3/\text{h}^{-1}$. Rigorous simulations with the software Aspen plus RATEFRAC module were also performed to validate the solution. The small temperature difference between liquid and vapor phases calculated with the Aspen proves the thermal equilibrium assumption on the stage. The composition and temperature profiles with these two methods are in good agreement.

The effects of inlet/outlet positions and flow rates on the temperature and composition distributions in the SPC were analyzed. The results

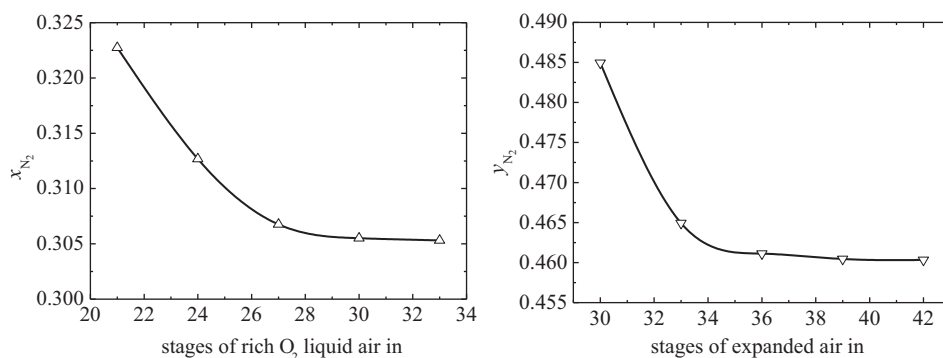


Fig. 10. Molar fraction of nitrogen component at the stage of rich O_2 liquid air in.

show that the inlet/outlet position with large nitrogen fraction has significant effects on the temperature and composition distributions. The effect of inlet/outlet position on the temperature distribution is limited to a small range near the position. There is a lower limit of stage to affect the distillation process for waste GN_2 withdrawn, rich O_2 liquid air in and waste LN_2 in.

Nomenclature

a	effective mass transfer area per unit volume, m^3
e	heat flux, $\text{J} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
F	feed rate, $\text{mol} \cdot \text{s}^{-1}$
H	enthalpy, J
h	specific enthalpy, $\text{J} \cdot \text{mol}^{-1}$
K	phase equilibrium constant
K	mass transfer coefficient, $\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
L	flow rate of liquid phase, $\text{mol} \cdot \text{s}^{-1}$
N	mass flux, $\text{mol} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$
P	pressure, Pa
V	flow rate of gas phase, $\text{mol} \cdot \text{s}^{-1}$
v	volume of the stage, m^3
x	molar fraction in liquid phase
y	molar fraction in gas phase
z	molar fraction in the feed or exhausting flow

Superscripts

I	interface
L	liquid phase
V	gas phase

Subscripts

i	component
j	stage number

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