

## Design method for multi-component distillation system based on quasi-binary method\*

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**Abstract**—A novel design method for a multi-component distillation system based on the quasi-binary model is presented. The quasi-binary method, which converts the multi-component system to a quasi-binary system, could simplify the design process of multi-component distillation. Subsequently the software integration method is introduced to the distillation design and an automatic calculation program is developed by using Visual C++ language. The design of multi-component distillation, in which the minimum reflux ratio  $R_{min}$  or liquid-vapor ratio  $(L/V)_{min}$  and the minimum numbers of stage  $N_{min}$  is determined easily and quickly, is automatically performed by the technology of software integration. Three examples are solved to demonstrate the feasibility and effectiveness of the presented method for the multi-component distillation.

Key words: Distillation Design, Quasi-Binary Equilibrium, Multi-Component, Software Integration

### INTRODUCTION

Distillation is a widely used separation process that dominates all separations in the chemical and petroleum industries, and it is the largest energy consumer among process unit operations. Thus, the task of optimal design and synthesis of distillation processes is an important and challenging issue, especially for multi-component separation processes.

The design techniques of distillation have attracted more and more attention in order to reduce the energy demand. Shortcut design and rigorous design methods are discussed in detail in the literature for different cases. The most popular and easy to use shortcut technique for designing binary system distillation is the McCabe-Thiele method. This simple method allows one not only to quantitatively predict minimum reflux conditions and minimum number of stages, but also to get a clear illustration and a pictorial understanding of these and other concepts, such as that of pinch conditions. Nothing of the kind is available for multi-component distillation, where the need for a multi-variant description of vapor-liquid equilibrium makes it very difficult to visualize the column design procedure. Hence, stage-by-stage models are used, which have a mathematically simple structure but also a serious drawback. To overcome this limitation, classical stage equations are analyzed [1] through a dynamical systems approach and the distillation column is regarded as a continuous countercurrent contactor with negligible axial dispersion. These are more complex than the stage-by-stage equations, but small in number and therefore can be solved by using convenient mathematical tools which allow one to reach a deep insight into both steady state and dynamical column behavior. The Underwood method [2] has been available for a long time as a suitable technique for the minimum energy demand calculation of ideal mixtures. In recent years, a shortcut method has been used for non-ideal mixtures by

the modified Underwood equation [3-8]. Although the short-cut method and its extensions have shown satisfactory design results, it is possible that such solutions are not accurate enough in the design of a distillation process because of the existence of several assumptions, such as constant molar overflow, constant relative volatility.

A rigorous design method [9-11] is adopted to reduce the energy consumption or the capital cost for the heat integration distillation process. However, due to complexity, most past research [12,13] on complex distillation configurations was restricted to three component mixtures, and only a few promising flowsheets were constructed for ternary mixtures. The design method of four or more component mixtures has been proposed by some scholars [14-17], but the corresponding applicability is also limited due to the combinatorial problem of the possible configurations for multi-component separations. Moreover, there is a lack of quick and simple design procedure as well as modeling and synthesis methods for these types of distillation schemes. In consequence, the optimal design of multi-component distillation processes is usually sought in a search space, which excludes considerations of complex distillation flowsheets due to the lack of experience and available knowledge.

In this paper, a novel design method of multi-component distillation process, which is based on the quasi-binary model, is presented. First, the quasi-binary equilibrium, which converts the multi-component system to quasi-binary system using congregated component, is introduced to simplify the multi-component distillation process. Second, the design process is automatically performed by the technology of software integration, of which the minimum reflux ratio  $R_{min}$  or liquid-vapor ratio  $(L/V)_{min}$  and the minimum numbers of stage  $N_{min}$  is determined easily and quickly. Three examples of the multi-component distillation for ideal and non-ideal mixtures are given as well.

### PROBLEM STATEMENT

The problem addressed in this paper can be stated as follows: Given are a multi-component feed composition, flow rate and specified product. The relative volatility of all components is not con-

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stant and varies along the column. The feeds, side streams and heat flows are located at the fitful point. The problem then consists of determining the minimum number of stages and the minimum liquid-vapor ratio (or reflux ratio).

### QUASI-BINARY EQUILIBRIUM OF MULTI-COMPONENT (QBEMC)

The vapor-liquid equilibrium relationship is the foundation for the design of a distillation column. For a simple binary system, there are four variables and two degrees of freedom in the system based on the phase rule. If one variable is fixed, only one variable among the others can be changed independently. But the multi-component situation is more complex than that of binary system. There are  $N$  variables that can be changed independently for the  $N$  components case. The equilibrium diagram of multi-component is very difficult to formulate accurately, which is the main difficulty and has not been solved satisfactorily so far. To solve the problem, the QBEMC is put forward and described as follows.

#### 1. The Concept of Congregated Component

Unlike a binary system, the product composition of the bottom and distillate are not determined directly for multi-component distillation. Instead, the composition of one component is generally subject to a low value in the distillate, and the composition of another component is subject to a low value in the bottom as well. The other components are not specified except the specified components which are called key components. One of the key components, for which the value of volatile is high, is called the light key component; the other is called heavy key component.

In this paper, the congregated component on the basis of the key components is presented to describe the multi-component system. The congregated component could be attained by the following steps: firstly the components are ranked according to their relative volatility by a descending order, that is, for the sequence ABCDE of the feed mixture, A and E represent the most volatile component and the least volatile component, respectively. Secondly, according to the separation task, the two key components are determined. For example, the task is to separate the components C and D. The component C is called the light key component, while the component D is called heavy key component. Thirdly, the components (A, B, C) are congregated to one component - the light congregated component (LCC); in turn the other components (D, E) are called the heavy congregated components (HCC). Then the multi-component system is converted to a quasi-binary system. The design method of a distillation column for a binary system could be applied to the quasi-binary system.

Since the determination of light congregated component and heavy congregated component is based on the separation specification not on the volatility, the volatility difference could not affect the design method, but it could affect calculation time. If the volatility difference is large in the light congregated component or heavy congregated component, the design of the column, which follows the separation column of light congregated component and heavy congregated component, would be easy or considered as the ideal mixture. Then the calculation would require short time. On the contrary, a long time would be required for the calculation.

#### 2. Phase Equilibrium of QBEMC

For a quasi-binary system, the equilibrium relationship of congregated component could be derived from that of the individual component, which could be formulated as follows:

For the  $j$  stage:

$$y_{j1} = K_{j1} x_{j1} \quad (1)$$

$$y_{j2} = K_{j2} x_{j2} \quad (2)$$

$$\dots \quad \dots$$

$$y_{ji} = K_{ji} x_{ji} \quad (3)$$

$$X_j = \sum_{i=1}^k x_{ji} = x_{j1} + x_{j2} + \Lambda + x_{jk} \quad (4)$$

$$Y_j = \sum_{i=1}^k K_{ji} x_{ji} = y_{j1} + y_{j2} + \Lambda + y_{jk} \quad (5)$$

$$Y_j = K_j X_j \quad (6)$$

$$K_j = \frac{\sum_{i=1}^k K_{ji} x_{ji}}{\sum_{i=1}^k x_{ji}} \quad (7)$$

Here,  $Y_j$ ,  $X_j$  are vapor and liquid composition of the congregated component, respectively. The  $K_j$ , equilibrium constant of the quasi-binary system, is a function of temperature, pressure, and composition and varies along the column. Eqs. (4) and (5) show that there are numerous combinations of  $x_{ji}$  ( $y_{ji}$ ), which could be summed equal to the same value of  $X_j$  ( $Y_j$ ). So the real  $K_j$  along the stages must be determined according as the separation task, which is important to the design process and very difficult in multi-component distillation.

Here the simulation software, such as PROII, HYSYS, and ASPENPLUS could be employed to simulate multi-component distillation processes. In this paper, ASPENPLUS is adopted to obtain the equilibrium of QBEMC. For a given value of reflux ratio and number of stages, the composition of the individual component on each stage would be obtained subsequently. Then the  $Y_j$ - $X_j$  relationship along the column would be described in a two-dimensional diagram easily or as an equation by curve-fitting. But the relationship of  $Y_j$ - $X_j$  would be limited because it corresponds to a certain stage number and reflux ratio. Therefore, a series of simulation processes for different stage number and reflux ratio are performed (in Fig. 1,

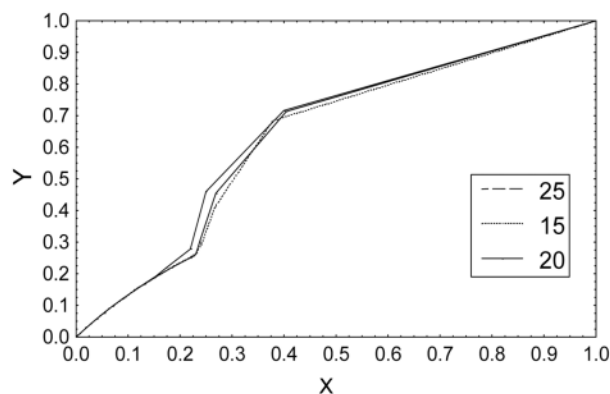


Fig. 1. Quasi-binary equilibrium of cyclohexane-water-isopropanol system.

the three profiles represent the number of stages, 15, 20 and 25, respectively). The result shows that the profiles of  $Y_j-X_j$  are very similar and the difference between them can almost be neglected. Thus, the localization of  $Y_j-X_j$  equilibrium could be overcome by using the average of a series of results and then the  $Y_j-X_j$  equilibrium relationship could serve for the design of distillation.

### QUASI-BINARY METHOD (QBM) FOR COLUMN DESIGN

With multi-component treated as a quasi-binary system, a column separating a multi-component mixture can be treated as a column separating LCC and HCC mixtures. According to the separation task, quasi-binary equilibrium is attained by using the simulation software. The minimum reflux ratio is calculated by the Lagrange quadratic interpolation, and the minimum stage number is calculated stage by stage. The total design process is automatically performed by the technology of software integration. The program schematic is shown as Fig. 2. The detailed design steps are described in the following sections.

#### 1. Determining of Phase Equilibrium

The value of the liquid-vapor ratio and the number of stages of the distillation separation process is affected deeply by the equilibrium relationship of the system. So the quasi-binary equilibrium of a multi-component system should be described before one calculates the value of the liquid-vapor ratio and the number of stages.

The reflux ratio  $R$  and stage number  $N$  must be initialized according to the property of the mixture before being simulated by calling the software. The Fenske and Underwood equations can be used to calculate the  $R_{min}$  and  $N_{min}$  when the relative volatilities are constant or do not change significantly along the column. Then the  $R_{min}$  and  $N_{min}$  are multiplied by a constant and the  $R$  and  $N$  are attained. For example,  $R=3R_{min}$ ,  $N=4N_{min}$ . Subsequently, the  $R$  and  $N$  are input to simulation software as the initialization. While the relative volatility is not treated as a constant, the  $R$  and  $N$  could be set at a large number according to experience and are input to the simulation software as the initialization. Then the composition of each component

of liquid and vapor along the column would be received by rigorous simulation. According to the separation task, the LCC and HCC must be determined. Sequentially, the composition of LCC and HCC would be attained along the column by Eqs. (1)-(5). Afterward, the quasi-binary equilibrium equation could be obtained by using Eqs. (6) and (8).

#### 2. Calculation of Minimum Reflux Ratio $R_{min}$ or Liquid-vapor Ratio $(L/V)_{min}$

When the composition data of each congregated component along the column has been obtained, the minimum reflux ratio can be calculated as follows.

First, the composition of LCC and HCC for the feed point is calculated. For example, four components with the molar composition are set as  $x_{j1}$ ,  $x_{j2}$ ,  $x_{j3}$ ,  $x_{j4}$ , respectively. The target is to separate the second and third components. That is, the first and second components are the light congregated components and the third and fourth components are height congregated components. Then the  $X_F$  liquid composition of LCC, equals the sum of  $x_{j1}$  and  $x_{j2}$ . The  $Y_F$  could be calculated by using the Lagrange quadratic interpolation method. For example, among the stochastic three points  $(X_0, Y_0)$ ,  $(X_1, Y_1)$ ,  $(X_2, Y_2)$  on the equilibrium line, the interpolation method is described as the following interpolation function equations.

$$Y_F = Y_0 l_0(X_F) + Y_1 l_1(X_F) + Y_2 l_2(X_F) \quad (8)$$

$$l_0(X_F) = \frac{(X_F - X_1)(X_F - X_2)}{(X_0 - X_1)(X_0 - X_2)} \quad (9)$$

$$l_1(X_F) = \frac{(X_F - X_0)(X_F - X_2)}{(X_1 - X_0)(X_1 - X_2)} \quad (10)$$

$$l_2(X_F) = \frac{(X_F - X_0)(X_F - X_1)}{(X_2 - X_0)(X_2 - X_1)} \quad (11)$$

Then the composition of liquid and vapor for each stage along the column is received.

Second, it is important to estimate if there are concave points among the composition points. The slope method is adopted to accomplish this work. The composition of the top point and bottom point is specified,  $(X_d, X_b)$  and  $(X_q, X_q)$ , respectively. Connecting the top point (or bottom point) and feed point would draw a line which slope given as  $a = (X_d - Y_F)/(X_d - X_F)$ . The equation of the line is described as  $Y' = aX + b$ . For each liquid composition  $X_j$ , the  $Y_j'$  could be calculated. If  $Y_j' > Y_j$ , there is concave point among the composition points. Otherwise, the concave point is not subsistent.

Third, the minimum reflux ratio could be calculated, respectively, for a concave situation and non-concave situation. For a non-concave situation, the minimum reflux ratio could be calculated by the slope of the line which connects the top point (or bottom point) and feed point. For the concave situation, the Lagrange quadratic interpolation method and golden section method are adopted to find the point of tangency which exists between the feed point and top point. Then drawing a line from the point of tangency to feed point or top point of column, the slope of the line could be used to calculate the minimum reflux ratio  $R_{min}$ .

#### 3. Calculation of Minimum Number of Stages

The stage to stage calculation method is adopted to calculate the minimum number of stages. The operation line is  $Y=X$  for infinite reflux. The top point of the column  $(X_d, X_q)$  is set as the beginning

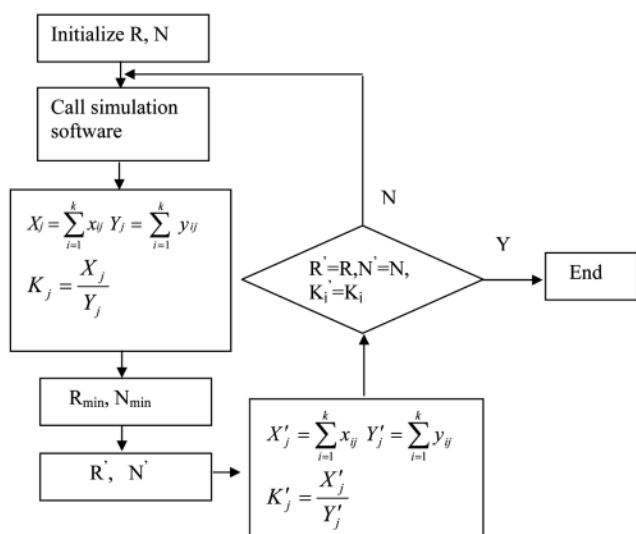


Fig. 2. The program schematic.

point of the operation line. Then  $Y_1 = X_d$ . The  $X_1$  is calculated by the chord position method. Then  $Y_2 = X_1$ ; the rest may be deduced by analogy until  $X_n < X_q$ . The minimum number of stages is  $n$ .

The iterative equation is described as follows:

$$X' = X_{j-1} - \frac{f(X_{j-1})}{f(X_{j-1}) - f(X_{j+1})}(X_{j-1} - X_{j+1}) \quad j=1, 2, \dots, k \quad (12)$$

Here

$$f(X_{j-1}) = g(X_{j-1}) - Y_{j-1} \quad j=1, 2, \dots, k \quad (13)$$

$$f(X_{j+1}) = g(X_{j+1}) - Y_{j+1} \quad j=1, 2, \dots, k \quad (14)$$

The  $g(X)$  is the polynomial of Lagrange quadratic interpolation, which is given above by Eqs. (8)-(11).

$$f(X') - g(X') - Y_{j-1} \quad j=1, 2, \dots, k \quad (15)$$

The iterative calculation goes on with the following steps:

- (1) Given composition point:  $(X_{j-1}, Y_{j-1})$ ,  $(X_j, Y_j)$ ,  $(X_{j+1}, Y_{j+1})$
- (2) Calculating the  $g(X_{j-1})$ ,  $g(X_{j+1})$ ,  $f(X_{j-1})$ ,  $f(X_{j+1})$ ,  $X'$  and  $f(X')$
- (3) If  $|f(X_{j-1})| < \varepsilon$ , then  $X = X'$ , End. Otherwise go step (4).
- (4)  $X_{j-1} = X_{j+1}$ ,  $X_{j+1} = X'$ , go step (2).

#### 4. Calculation of the Operation Reflux Ratio $R$ and Number of Stages $N$

Once the minimum reflux ratio and minimum numbers of stages of a column separating the multi-component mixture have been determined, the following two methods could be used to calculate the operation reflux ratio and number of stages. In the first method, the operation reflux ratio is set times the minimum reflux ratio. Then the number of stages is calculated using stage to stage. In another method, the operation number of stages could be obtained by multiplying a factor of the minimum number of stages. Then the actual reflux ratio is calculated in turn. In this paper, the former method is adopted. The ratio factor between the operation reflux ratio and minimum reflux ratio is set to 1.2 ( $R = 1.2R_{min}$ ) in order to compare the result with that of the reference [18]. But the factor is not limited only to 1.2. Other values, such as 1.3 or 1.5, could be adopted to accomplish the multi-component distillation by QBM. The vapor and liquid composition of feed could be calculated, which is mentioned above in section 5.2. The number of stages is calculated from the distillate point and bottom point to the feed point, respectively, by the chord position method, as mentioned above. The total number of stages is the sum of the number of stages of rectifying section and stripping section. At the same time, the feed location is determined.

#### 5. Verification of the $R_{min}$ and $N_{min}$

The calculation results of  $R_{min}$  and  $N_{min}$  are verified in this section to demonstrate the design method presented in this paper. The number of stages is set to a large number ( $N=300$ ) to rigorously simulate. If the specified separation is not satisfied by adjusting the reflux ratio and the feed location, the value of  $R_{min}$  calculated by the above method would be accurate and the design method is feasible. Similarly, the  $N_{min}$  could be verified. The following examples would give further explanation.

### EXAMPLES

Example 1: The feed of methanol, ethanol and 1-propanol with

**Table 1. Comparison of shortcut design and QBM**

	Shortcut design	QBM
$X_F$ (mole fraction: methanol/ethanol/1-propanol)	0.3/0.1/0.6	0.3/0.1/0.6
$\alpha_{methanol}/\alpha_{ethanol}/\alpha_{1-propanol}$	1.805/1/0.488	1.805/1/0.488
$N_{min}$	21	17
$R_{min}/(L/V)_{min}$	2.78/0.735	2.93/0.741
$N$	40	40
$R$	3.48	3.52
Feed stage	21	22

mole fraction of 0.3, 0.1 and 0.6 is considered. The desired separation is methanol from the other two components. The relative volatilities are nearly constant and are 1.805, 1 and 0.488, respectively. The product specifications were 1% recovery of the heavy and light keys component in the distillate and bottoms. The results are summarized in Table 1.

From Table 1, it can be seen that the results obtained by the two methods of column design have little error for the prediction of the minimum stage number and minimum reflux ratio ( $R_{min}$ ) or liquid-vapor ratio ( $(L/V)_{min}$ ) at the same feed composition and product specification. It illustrates the validity of the QBM for column design. Noticeably,  $N_{min}$ ,  $N$ ,  $R$ ,  $R_{min}$  and the feed location are automatically calculated by using QBM, which is extremely quick and simple because iteration is avoided, especially compared to the other available methods, such as the boundary value method [19] or the use of manifolds [20].

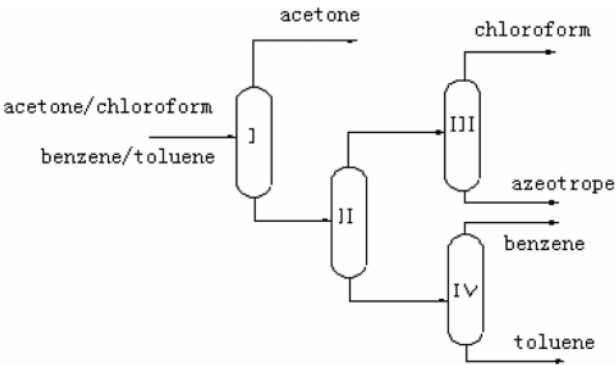
Example 2: Consider the four-column distillation sequence separating the quaternary mixture with azeotropic point of acetone, chloroform, benzene and toluene. The feed data for this example are given in Table 2 [18] and the separation sequence is shown in Fig. 3. The results are summarized in Table 2.

From Table 2, it can be seen that there exists larger error among the FUG HYSYS and the QBM in the calculation result of  $N_{min}$  and  $R_{min}$  or  $(L/V)_{min}$ . The reason is that there are several assumptions in the FUG method for prediction of  $N_{min}$  and  $R_{min}$  or  $(L/V)_{min}$ . So it is not accurate to column design especially for a non-ideal mixture. Numerous other examples studied gave the same general results. The reason for the error between HYSYS and the QBM could be the proportion factor, which is that the ratio of  $R$  and  $R_{min}$  is different. For the HYSYS method, the proportion factors adopted in the four columns are different. But the same proportion factor is set to the four columns for the QBM. To illustrate the results of QBM method presented in this paper, a rigorous simulation using ASPEN-PLUS is adopted.

The specification of distillate and bottom could be reached in the following two conditions: first, the stage number is set to a smaller value than  $N_{min}$  given by FUG or HYSYS; the production specification could be attained while the  $R$  or  $(L/V)$  increases larger. In the same manner, the  $R_{min}$  or  $(L/V)_{min}$  is set to a smaller value than that of FUG or HYSYS; the production specification also could be obtained while the stage number becomes larger. But if the stage number (or reflux ratio) is smaller than  $N_{min}$  (or  $R_{min}$ ) given by QBM, the production specification always cannot be satisfied. Thus, the result of QBM is accurate and the QBM is effective for the multi-

**Table 2. Comparison of the QBM and the FUG shortcut design**

	Splits			
	Column I	Column II	Column III	Column IV
$X_F$	0.26/0.32/0.21/0.21	0.09/0.39/0.26/0.26	0.16/0.83/0.01/0	0/0.02/0.48/0.50
$X_D$	0.93/0.04/0.03/0	0.16/0.83/0.01/0	0/1.0/0/0	0/0.02/0.98/0
$X_B$	0.09/0.39/0.26/0.26	0/0.02/0.48/0.50	0.31/0.67/0.02/0	0/0/0/1.0
D/F	0.21	0.47	0.41	0.5
Shortcut method (FUG)				
$R_{min}$	5.12	1.95	5.35	2.29
$N_{min}$	25	26	23	11
R	6.14	2.34	6.42	2.75
N	53	58	48	25
HYSYS				
$R_{min}$	6.06	2.5	4.5	2.12
$N_{min}$	28	20	20	9
R	6.3	3.1	7.26	2.96
N	53	58	48	25
QBM				
$(L/V)_{min}/R_{min}$	0.787/3.72	0.775/3.44	0.814/4.38	0.57/1.324
$N_{min}$	9	21	23	19
$(L/V)/R$	0.816/4.45	0.805/4.13	0.84/5.26	0.614/1.59
N	29	59	47	38
Feed stage	17	10	25	18

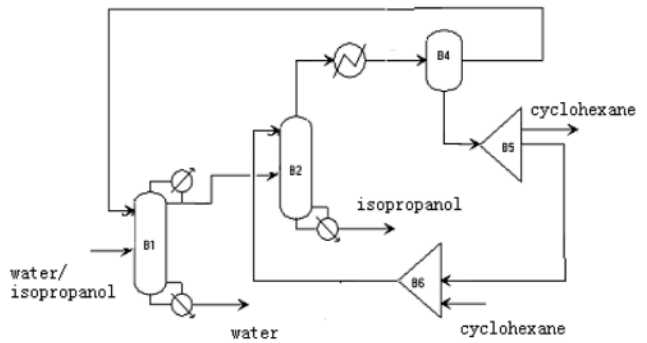


**Fig. 3. Four column distillation sequence separating the quaternary mixture of acetone, chloroform, benzene and toluene.**

component non-ideal mixture too, which could accurately predict the  $N_{min}$  and  $R_{min}$  or  $(L/V)_{min}$ .

Example 3: In this example the azeotropic distillation for separating the mixture isopropanol/water is analyzed. The mole fraction of feed mixture is 0.41/0.59 and the cyclohexane is employed as entrainer. The separation flowsheet is shown in Fig. 4, which comprises two distillation columns—concentrating column and dehydration column. The concentrating column is a general column which is designed easily. The dehydration column is a complex azeotropic distillation column and will be discussed in detail. The product specifications were 99.5% recovery of isopropanol at the bottom and less than 0.01% residual quantity of isopropanol at the distillate.

Because the relative volatilities of the azeotropic mixture vary significantly along the column, the initial values of R and N would



**Fig. 4. The flowsheet of azeotropic distillation for separating the mixture isopropanol/water.**

**Table 3. Design results of the azeotropic distillation of isopropanol-water system**

	Column I	Column II
Nmin	13	14
$(L/V)_{min}$	0.96	1.07
N	15	15
L/V	1.15	1.28

be given a relatively large number according to the experience of calling the simulation software. Subsequently, the automatic calculation program would be performed to accomplish the process design. The results are shown in Table 3. The liquid-vapor ratio is adopted to replace the reflux ratio as there is not a condenser for the dehydration column. Afterward, the obtained L/V and N are input to AS-

PENPLUS software to simulate the separation process. The product specification is satisfied. It indicates that the QBM is feasible for distillation design of non-ideal mixtures and complex systems.

## CONCLUSION

The design of distillation for multi-component separation has been studied. The concept of congregated components, which converts a multi-component system to quasi-binary system using light congregated components and heavy congregated components, has been put forward to simplify multi-component distillation processes. Then the design method based on the quasi-binary, which is automatically performed by the technology of software integration, is presented to complete the design of multi-component distillation. The results of three examples are shown: the proposed method is valid for multi-component distillation processes, which is not only applied to ideal mixtures but also non-ideal mixtures.

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## NOMENCLATURE

A, B, C, D, E : pure component of mixture  
 K : phase balance constant of integrated component  
 k : phase balance constant of pure component  
 L : mole flow rate of liquid  
 V : mole flow rate of vapor  
 y : mole fraction of pure component in the vapor phase  
 x : mole fraction of pure component in the liquid phase  
 N : number of stages in a column  
 R : reflux ratio  
 X : mole fraction of integrated component in liquid phase  
 Y : mole fraction of integrated component in vapor phase  
 $\alpha$  : relative volatility of component

## Subscripts

F : feed of the column  
 D, d : distillate product  
 B, b : bottom product  
 Min : minimum  
 i : the i-th component

n : the n-th stage

## Acronyms

FUG : Fenske-Underwood-Gilliland method  
 MCS : Model of Column Section  
 VLE : Vapor-Liquid Equilibrium  
 LCC : Light congregated component  
 HCC : Heavy congregated component  
 QBM : Quasi-Binary Method

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