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RESEARCH ARTICLE

Liquid mixtures separation and heat consumption in the process of distillation

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Abstract

Objectives. *The aim of this study is to investigate different distillation modes of a binary ideal mixture and determine how various factors affect heat consumption in the column boilers. In addition, it intends to assess the difficulty of separating mixtures. Our research is based on analyzing the characteristics of vapor-liquid equilibrium.*
Methods. *To conduct our study, we used a graphic-analytical tool to calculate the distillation process of a binary mixture and mathematical models based on the Aspen Plus software package along with DSTWU, RadFrac, and the Sensitivity module. We also used the Peng-Robinson equation (PENG-ROB) to determine the liquid-vapor equilibrium.*
Results. *We employed the graphical method and mathematical models to obtain the operation parameters of two column variants for the distillation of binary ideal benzene-toluene mixtures. In each variant the initial mixture contained the same amount of the low- or high-boiling component. The number of plates in the column sections, reflux ratio, energy consumption, and indicators of internal energy saving were determined.*
Conclusions. *Study results show that using the coefficient of the component distribution between the vapor and liquid phases is a promising method for preliminary assessments of the separation difficulty and measurements of the expected heat consumption in the boilers of columns. Comparison studies showed that the heat consumption in the boiler decreases as the internal energy saving in the columns increases.*

Keywords: *distillation, binary mixtures, relative volatility, reflux ratio, distribution coefficient, internal energy saving in distillation*

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НАУЧНАЯ СТАТЬЯ

Разделение жидких смесей и затраты теплоты при ректификации

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Аннотация

Цели. *Работа посвящена изучению различных режимов ректификации бинарной идеальной смеси и установлению влияния различных факторов на затраты теплоты в кипятильниках колонн, а также оценке трудности разделения смесей на основе анализа характеристик парожидкостного равновесия.*
Методы. *В качестве методов исследования выбран графоаналитический расчет процесса ректификации бинарной смеси и математическое моделирование с использованием программного комплекса Aspen Plus с последовательным применением методов DSTWU, RadFrac и модуля «Sensitivity». Равновесие жидкость-пар было определено по уравнению Пенга-Робинсона (PENG-ROB).*
Результаты. *С использованием графического метода и метода математического моделирования получены параметры работы колонн двух вариантов ректификации бинарных идеальных смесей бензол–толуол, в каждом из которых исходная смесь содержит одинаковое количество низко(высоко)кипящего компонента. Определены числа тарелок в секциях колонны, флегмовые числа, энергозатраты и показатели внутреннего энергосбережения.*
Выводы. *Анализ полученных результатов показывает перспективность использования коэффициента распределения компонентов между паровой и жидкой фазами для предварительной оценки трудности разделения и ожидаемых затрат теплоты в кипятильниках колонн. Сравнение рассмотренных вариантов показало, что при увеличении внутреннего энергосбережения в колоннах затраты теплоты в кипятильнике уменьшаются.*

Ключевые слова: *ректификация, бинарные смеси, относительная летучесть, флегмовое число, коэффициент распределения, внутреннее энергосбережение при ректификации*

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INTRODUCTION

Heat consumption in the process of separating liquid mixtures by distillation results from the need of evaporating the liquid to form a vapor flow in the column. It is determined by the difficulty of separating a particular initial mixture. It is known that the still residue rather than the original mixture is evaporated in a complete distillation column. Therefore, the correct assessment of heat consumption in the boiler of a distillation column is a subject of considerable interest. This paper analyzes the main factors affecting heat consumption in the distillation process with an ideal benzene–toluene mixture as an example.

THEORY

The conventional method [1, 2] of assessing the difficulty of separating a mixture is to use the relative volatility coefficient of the starting components. For ideal systems, the coefficient is determined by the ratio of saturated vapor pressures of the pure components (P_A and P_B) at a certain temperature t: $\alpha = P_A/P_B$.

Note that even in the case of ideal binary mixtures, the value of α is not constant but rather depends on the low-boiling component (LBC) concentration in the mixture. When changing the mixture composition, its boiling temperature changes, and as a result, the value of P_A/P_B, alters. This is conditional upon by the non-linearity of saturated vapor pressure dependence on temperature. Thus, for the benzene–toluene mixture at low LBC concentrations in the mixture (less than 5%), the coefficient of relative volatility is equal to 2.30, and at high LBC concentrations (more than 80%), the coefficient is equal to 2.55. The concentration difference is about 10%. For comparison, in the benzene–*m*-xylene system, the concentration difference exceeds 20%.

Assuming that the coefficient of relative volatility is constant [1, 2], the equilibrium compositions of liquid x and vapor y for ideal binary mixtures are determined by the following relationship:

$$y = \frac{\alpha x}{\alpha x + 1 - x}. \tag{1}$$

When α is close to 1, one can take

$$y = \alpha x. \tag{2}$$

The LBC concentration in the vapor phase as compared to its concentration in the liquid is equal to

$$y - x = \alpha x - x = (\alpha - 1)x. \tag{3}$$

The value (α – 1) is called the enrichment factor [3]. It is clear from (3) that the enrichment factor is the relative increase in LBC concentration in the vapor phase compared to its concentration in the liquid phase. A wide range of changes in the values of α (from 1 to ∞) makes it impossible to express analytically the dependence of heat consumption and determine the influence of α and $(\alpha - 1)$ values on heat consumption in the distillation process.

To facilitate assessment of the mixture separation ability by distillation is to determine the value of P. We called it the mixture "separability," and is determined as follows [4]:

$$P = \frac{\alpha - 1}{\alpha + 1}. \tag{4}$$

The value of P varies from 0 (at $\alpha = 1$) to 1 (at $\alpha = \infty$). From (4) it follows:

$$\alpha = \frac{1 + P}{1 - P}. \tag{5}$$

Substituting the expression of α into (1) we obtain the equilibrium dependence of the vapor composition y on the liquid composition x for mixtures obeying the Raoult's law:

$$y = \frac{(1 + P)x}{1 - P + 2Px}. \tag{6}$$

A value comparison of the above estimates is presented in Table 1.

For any LBC concentration values in the liquid, the coefficient of relative volatility is represented as the ratio of the phase equilibrium coefficients of the components of the mixture being separated:

$$\alpha = \frac{K_A}{K_B}, \tag{7}$$

where $K_A = P_A/P_{total}$ and $K_B = P_B/P_{total}$; P_{total} is the total pressure in the column.

Table 1. Comparison of various estimates of the ability of liquid mixtures to be separated by distillation

α	**P**	**α – 1**	**α**	**P**	**α – 1**
1	0	0	3	0.5	2
1.01	0.0050	0.01	4	0.6	3
1.05	0.0244	0.05	5.6	0.7	4.6
1.1	0.0476	0.1	9	0.8	8
1.2	0.0909	0.2	19	0.9	18
1.3	0.1304	0.3	50	0.961	49
1.4	0.1667	0.4	100	0.98	99
1.5	0.2	0.5	1000	0.998	999
1.857	0.3	0.857	∞	1	∞
2.3	0.4	1.3	–	–	–

The values of the phase equilibrium constant of the components K_A and K_B at P_{total} depend on the component concentration, as the vapor pressure of the pure components is a function of their boiling point t, and the latter depends on the mixture composition. At low LBC concentrations, the constant K_B is close to 1. In this case, we assume that $\alpha = K_A$. For medium concentrations, the coefficient of relative volatility α is determined by the K_A/K_B ratio (7). This means that the coefficient α takes into account both the LBC concentration increase in the vapor phase compared to the liquid phase ($K_A = y/x$) and the high-boiling component (HBC) increase in the liquid phase compared to the vapor phase ($1/K_B = 1-x/1-y$), i.e., it takes into account the same effect twice.

When the LBC concentration tends to 1, the coefficient of relative volatility becomes $\boldsymbol{\alpha} = 1/K_B$. In this case, the LBC concentration in the vapor phase (y) slightly exceeds its concentration in the liquid (x). Let us take for example $x = 0.9$ and $y = 0.95$. Then

$$\alpha = \frac{1}{K_B} = \frac{1-x}{1-y} = \frac{1-0.9}{1-0.95} = \frac{0.1}{0.05} = 2 \tag{8}$$

characterizes only the increase in the HBC concentration in the liquid phase compared to the vapor phase. In this case, the value of α equal to 2 indicates a fairly the good ability of this liquid binary mixture to be separated by distillation. However, a small relative increase in the LBC concentration

$$\frac{y-x}{x} = \frac{0.95-0.90}{0.90} = 0.0556$$

testifies otherwise.

In literature [1, 2], the ratio of phase equilibrium concentrations of any component is called the distribution coefficient

$$m = \left(\frac{y}{x}\right)_{eq}. \tag{9}$$

This coefficient is the tangent of the inclination angle of the secant drawn from the coordinate origin to the point with equilibrium concentrations y, x. The subscript *eq* in formula (9) indicates the equilibrium LBC concentrations in the vapor and liquid phases.

It should be emphasized that m is the tangent of the inclination angle of the secant, since m is often interpreted in a number of publications [2] as the tangent of the inclination angle of the tangent to the equilibrium line at points with equilibrium concentrations y, x. Only at low LBC concentrations (up to 1–3%) in the mixture, the tangent of the tangent inclination angle differs slightly from the tangent of the secant inclination angle. In other cases, this difference becomes larger (Fig. 1), even for binary mixtures close to ideal. Moreover, this difference is larger in the case of real mixtures.

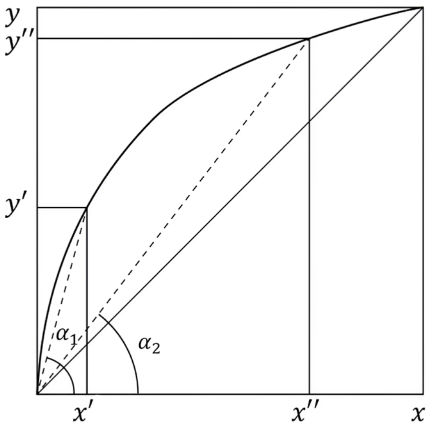

Fig. 1. Graphical determination of the components' distribution coefficient.

Figure 1 shows that the value of the distribution coefficient m at low LBC concentrations is much higher (x',y' point) than at high LBC concentrations (x'',y'' point): $\mathrm{tg}\alpha_1 > \mathrm{tg}\alpha_2$. When $x \to 1$, the coefficient m tends to 1, indicating a decrease in the vapor phase enrichment with the LBC compared to the liquid one. Thus, in our opinion, using the distribution coefficient, which is a measure of the absolute component volatility, is more correct when analyzing mixture separation by distillation. Yet, the relative volatility coefficient is still widely used [5–13] for assessing the ability to separate mixtures. This is partly due to the fact that, when developing energy-efficient distillation schemes for mixtures with a constant relative volatility coefficient, one of the most common methods, the Underwood method, is used to calculate the minimum reflux ratio [14]. However, the latter does not provide acceptable accuracy values. Therefore, the work [15] suggests a method for calculating the minimum reflux ratio based on the phase equilibrium constants of mixture components (distribution coefficients).

RESULTS AND DISCUSSION

We conducted a computational experiment to reveal how the distribution coefficient m affects heat consumption during distillation. We used a benzene–toluene mixture with phase behavior close to ideal. We considered two options of separating the initial mixtures containing 0.74 mole fractions of one of the components (benzene–option 1, toluene–option 2, see Table 2). The task was to obtain a product (distillate for option 1 or still residue for option 2) containing 0.98 mole fractions of the target component and obtain the second product of separation with the equimolar composition. The column productivity L_1 with respect to the initial mixture was taken as 0.01 [kmol/s] in both cases. The excess reflux ratio σ was also assumed to be the same and equal to 1.1.

The calculation of the number of theoretical plates in the column performed graphically in the *y*–*x* diagram is qualitatively schematized in Fig. 2 for both separation options.

The values of the number of plates in the columns and the heat consumption in the boilers are listed in Table 2. In parallel, we carried out a design analysis using the Aspen Plus software along with DSTWU, RadFrac methods and the Sensitivity module. We determined the liquid–vapor equilibrium by using the Peng-Robinson equation (PENG-ROB), as this equation is the most appropriate for modeling the phase equilibrium of a mixture consisting of hydrocarbons [16–18]. The calculation results are listed in Table 2.

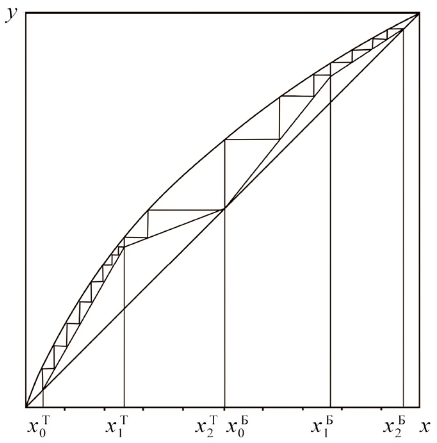

Fig. 2. Graphical determination of the number of plates in the *y*–*x* diagram.

The DSTWU model allows for an approximate column design (minimum reflux ratio, minimum number of plates). The operating reflux ratio was calculated taking into account the excess reflux ratio, which is equal to 1.1. Then the RadFrac model was used for refined calculations. After selecting the total number of plates in the column closest to the previously calculated reflux ratio values the Sensitivity module was used, which allowed determine the feed plate providing the minimum heat consumption in the boiler.

When estimating the possibility of separating mixtures via a relative volatility of components we assumed that if this value is constant, it is equally

Table 2. Comparison of calculation results for two options of benzene–toluene mixture separation

Parameters	Calculation in the *y*–*x* diagram						Computer calculation in Aspen Plus software					
	Option 1			Option 2			Option 1			Option 2		
	Composition, $\left[\frac{\text{kmol LBC}}{\text{kmol mxtr}}\right]$											
	x_0	x_1	x_2	x_0	x_1	x_2	x_0	x_1	x_2	x_0	x_1	x_2
	0.50	0.74	0.98	0.02	0.26	0.50	0.50	0.74	0.98	0.02	0.26	0.50
Theoretical number of plates in the rectifying column, n_r	12.9			4.1			9			3		
Theoretical number of plates in the stripping column, n_s	3.2			15.2			3			16		
Reflux ratio, *R*	0.857			0.208			0.848			0.218		
Heat duty in the boiler Q_b, [kW]	283			210			284			200		
Internal energy saving, *ES*	0.569			0.824			0.591			0.801		
Distribution coefficient, *m*	1.18			1.8			1.18			1.8		

difficult to separate the mixtures. Furthermore, we expected equal heat consumptions in the boilers in the discussed distillation embodiments. However, the results in Table 2 indicate a higher heat consumption (by 26%) in the first separation option. This fully conforms to the proportionality of heat consumption to the product $D(R+1)$, where D is the distillate flow. Since the sampling ratios of the upper product in both options are equal to 0.5, the large heat consumption is due to the high reflux ratio in the first option. The question which arises is why the mixture in option 1 is more difficult to separate. The answer lies in the value of the distribution coefficient m between the vapor and liquid phases (in the range of working concentrations). In option 1, it is much lower (1.18) than in option 2 (1.80) (see Table 2). Therefore, we carried out a preliminary estimation of the possibilities of separating mixtures and of the expected heat consumption based on coefficient analyses of the component distributions between the phases in the initial mixture.

The obtained results illustrate the contribution of the internal energy saving to the heat consumption in the column boiler [19]. The internal energy saving is determined not only by the reflux ratio in the rectifying section, but also by the ratio of the number of theoretical plates in the rectifying and stripping sections of the column. When the initial mixture is fed into the column at the boiling point, the internal energy saving in the distillation column is calculated by the following formula [19]:

$$ES = \frac{R}{R+1} \times \frac{n_\mathrm{r}}{n_\mathrm{r} + n_\mathrm{s}} + \frac{n_\mathrm{s}}{n_\mathrm{r} + n_\mathrm{s}}. \qquad (10)$$

An example of calculation by formula (10) for option 1 is presented below. Let us calculate the value of internal energy saving at operating reflux ratio 0.857; the number of theoretical plates in the rectifying section of the column 12.9; the number of theoretical plates in the stripping section of the column 3.2:

$$ES = \frac{0.857}{0.857+1} \times \frac{12.9}{12.9+3.2} + \frac{3.2}{12.9+3.2} = 0.370 + 0.1988 = 0.569.$$

When carrying out computer calculations, the values of liquid and vapor flows along the column height become known. Then we can use a more accurate formula for calculating energy saving. The formula takes into account the counterflows of the liquid L and vapor V on each plate of the rectifying part of the column [20]:

$$ES = \frac{\sum_{n=2}^{n_\mathrm{y}} \frac{L_{n-1}}{V_{n+1}} + n_\mathrm{o}}{n_\mathrm{y} + n_\mathrm{o}}. \qquad (11)$$

To perform the calculation according to formula (11)

Table 3. Flows of liquid and vapor in a distillation column during separation of a benzene–toluene mixture (option 1)

Plate number	Temperature, °C	Liquid flow, kmol/h	Vapor flow, kmol/h	$\frac{L_{n-1}}{V_{n+1}}$
1	80.1	33.27	0	–
2	80.8	15.18	33.27	0.4602
3	81.5	15.08	33.18	0.4588
4	82.2	14.99	33.08	0.4572
5	83.0	14.90	32.99	0.4556
6	83.7	14.82	32.90	0.4540
7	84.3	14.75	32.82	0.4525
8	84.8	14.69	32.75	0.4512
9	85.3	14.65	32.69	0.4500
10	85.6	50.62	32.65	0.4491
11	85.9	50.54	32.62	–
12	86.6	50.33	32.54	–
13	88.3	49.87	32.33	–
14	92.2	18.00	31.87	–

we will use the data from Table 3. It should also be considered that the first plate is a condenser, in which vapor is condensed to obtain a liquid of the same composition. Therefore, the calculation starts at the second plate and ends at the feed plate. Then:

$$ES = \frac{4.0886 + 3}{3 + 9} = 0.591.$$

The number 4.0886 in the calculation formula is the sum of the ratios of the liquid and vapor flows on the plates of the column rectifying section.

Minor discrepancies in the energy saving values calculated in the *y*–*x* diagram and in the Aspen Plus program stem from the refined computer calculation of the liquid and vapor flows through the column, and when calculating in the *y*–*x* diagram—due to the possibility of counting the fractional part of the theoretical plate.

CONCLUSIONS

The analysis of the distillation of ideal binary benzene–toluene mixtures of different composition showed that the coefficient of the distribution of components between the vapor and liquid phases is a more accurate characteristic of the mixture, which can be used for preliminary assessments of separation difficulty and the expected heat consumption in the boilers of the columns. Using the coefficient of relative volatility α for these purposes, whose value is assumed constant for ideal mixtures, does not always yield correct results. Comparison of the considered options was carried out according to the criterion of internal energy saving. It was shown that the heat consumption in the boiler decreases with increasing internal energy saving in the columns.

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Authors' contribution

M.K. Zakharov – developing the scientific work concept, offering consultations on methodology and research;

A.V. Egorov – collecting and processing the material, writing the text of the article;

A.A. Podmetenny – collecting and processing the material, writing the text of the article.

The authors declare no conflicts of interest.

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