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Mathematical Modeling of the Cyclic Distillation of Binary Mixtures with a Continuous Supply of Streams to the Column

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Abstract—A mathematical model for cyclic binary distillation in a sectioned column with a continuous supply of streams has been proposed. A method has been developed for organizing cyclic modes in a sectioned apparatus with the operation of one of the sections in the liquid flow mode and the other sections in the vapor flow mode. The effect of the parameters of the proposed model on separation in the column has been studied.

Keywords: cyclic distillation, controlled phase movement, ideal displacement, perfect mixing, mass transfer, column separation efficiency, thermodynamic criterion, Murphree tray efficiency

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INTRODUCTION

Growth in the chemical and petrochemical industries requires the development of more effective methods for implementing the process. One of the methods of increasing the efficiency of mass-transfer processes is to use periodic (cyclic) phase movement modes in apparatuses. Cyclic distillation is based on the alternation of two modes [1], i.e., the vapor flow period, in which the vapor stream moves upward through the column, and the liquid flow period, in which the liquid stream moves from each tray to the below-located tray. The feed mixture and the reflux stream are only supplied to the column in the liquid flow period. This operation mode of the apparatus is also known as separate phase movement [2].

The first studies into the intensification of mass transfer processes due to periodic changes in operating parameters were conducted in the 1960s. An increase in the efficiency of cyclic distillation to 200% compared with conventional continuous distillation was shown in [2–10]. The maximum efficiency is achieved under the conditions of complete replacement of the fluid on trays without mixing in the liquid flow period, which is impossible when conventional contact devices are used. The problem of the plug flow of the liquid is successfully solved using trays proposed by Maleta [11, 12].

The industrial implementation of cyclic distillation is limited by the following factors: (a) hydrodynamic impact during the supply of the vapor to the apparatus; (b) the need to install reliable high-speed valves; (c) the difficulty of regulating the flow rates of the vapor, feed, and reflux during the short period of supply; and (d) the periodicity of the operation of a reboiler and a reflux condenser.

The specified disadvantages can be partially avoided using two columns with a common bottom and reflux condenser so that, when the vapor is supplied to one of the columns, the liquid is supplied to the other column [13]. However, in this case, it is impossible to select the optimal ratio of the liquid and vapor flow periods. In addition, the problem of hydrodynamic impact remains.

Organizing the cyclic process without interrupting the supply of the liquid and vapor to the column is of greater interest. This can be achieved by using trays without downcomers with the regulated active tray area [14]. A disadvantage of this method that the liquid mixes during flow, which decreases the efficiency of separation. Another solution is to use a column with a movable perforated cylinder [15]. Trays with downcomers are located in the movable cylinder, and the liquid flows through special pockets in an external motionless cylinder. The use of this design is only justified in apparatuses with small diameters, since an increase in dimensions leads to an increase in energy expenditures on the movement of the cylinder. Another solution is to use an apparatus with sieve trays with downcomers [16]. Here, a tray consists of two semicircular perforated plates that are folded via rotation about the horizontal axis for the liquid flow. The specific feature of this method is the organization of the liquid flow in a tray-by-tray manner beginning

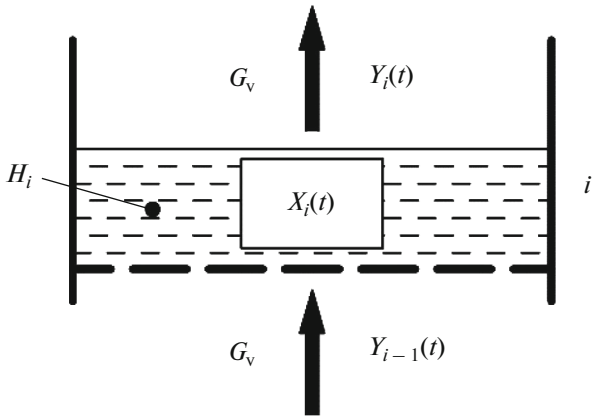

Fig. 1. Schematic representation of a tray in the vapor flow period.

from the bottom of the column, rather than simultaneously along the entire column. The disadvantage of the method is that it is necessary to install equipment for folding each tray, which complicates both the design itself and a control system for this process.

The purpose of this study is to investigate the cyclic distillation of binary mixtures with the continuous supply of streams to a sectioned column using the mathematical model of the process. To achieve this goal, the following objectives are to be achieved: (1) to develop a method for organizing the continuous supply of material streams to the sectioned column of cyclic distillation; (2) to develop a mathematical model for cyclic distillation with the continuous supply of streams to the sectioned column; (3) to study the effect of the parameters of the mathematical model on the efficiency of separation; and (4) to verify the adequacy of the proposed mathematical model.

MATHEMATICAL MODEL

To organize the continuous supply of material streams to a cyclic distillation column, it is proposed to use a vertically sectioned column with the bottom and a reflux condenser that are common to all sections. In each section, the vapor and liquid flow periods alternate so that, at any point in time, the flow of the liquid only takes place in one of the sections. Thus, the proportion of the vapor flow period in the entire column depends on the number of sections. A detailed description of a method for the continuous supply of streams to a column is presented in Method for the Continuous Supply of Streams to a Column.

The presented mathematical model, which takes into account the mixing of the liquid on trays in the liquid flow period, has been obtained by improving the model of binary cyclic distillation that we proposed in [17, 18] based on studies [19, 20]. According to the model, a multisectional column is calculated as individual independent columns with the common bottom and reflux condenser. For each section, the vapor flow period and the liquid flow period are calculated alternately and independently using initial data on the feed mixture.

In the mathematical model of cyclic binary distillation, the following assumptions were made: (a) a change in the concentration of the more volatile component (also called the light key) in the vapor that leaves a tray instantaneously follows a change in the concentration of the liquid on the tray; (b) equimolar mass transfer takes place; (c) the holding capacity of a tray with respect to the liquid is the same at all of the stages of the enriching and stripping sections; (d) the Murphree tray efficiency is constant at the given separation stage; (e) the original mixture is supplied to the column at the initial boiling point; (f) the entrainment of the liquid is not taken into account; (g) in the vapor flow period, there is perfect mixing on all trays; (h) the condenser of the column is a total condenser; (i) the vapor that leaves the bottom is in equilibrium with the liquid; (j) within the stripping and enriching sections of the column, the same amount of the liquid flows from each tray of the section for the liquid flow period; and (k) there is no mass transfer in the liquid flow period. The initial data for calculating the conventional distillation process were as follows: $N, f, R, E_M, n, X_F, G_F, G_V, P, H_W$, and H_D. The initial data for calculating the cyclic mode were as follows: τ, γ, β, and η.

Vapor Flow Period

A schematic representation of the movement of streams on a tray in the vapor flow period is shown in Fig. 1. In the vapor flow period, the variations in the fraction of the light key on the ith tray is described by the following equation:

$$\frac{dX_i(t)}{dt} = \left[Y_{i-1}(t) - Y_i^*(X_i)\right]\frac{G_V E_M}{H_i}. \quad (1)$$

A system of equations written for each tray based on Eq. (1) requires simultaneous solving, since, in order to calculate the variation in the fraction of a component on a tray, it is necessary to know the fraction of this component in the vapor that enters from the previous tray.

For an ideal mixture, the equilibrium concentration of the light key in the vapor on a tray is described by the well-known equation

$$Y_i^* = K_i(T_i, P_i)X_i. \quad (2)$$

At a temperature that corresponds to an equilibrium state, the following condition should be satisfied:

$$\sum_{j=1}^{n} K_{i,j}(T_i, P_{i,j})X_{i,j} = 1. \quad (3)$$

The vapor–liquid distribution ratio for an ideal mixture is calculated as the ratio of the saturated vapor

pressure of the *j*th component over its pure solution to the total pressure in the system as follows:

$$K_{i,j}(T_i, P_{i,j}) = \frac{P_{i,j}(T_i)}{P_i}. \quad (4)$$

The vapor pressure of the *j*th component over a pure solvent can be calculated using the extended Riedel equation [21]

$$\ln P_{i,j}(T_i) = A_j + \frac{B_j}{T_i} + C_j \ln T_i + D_j T_i^2 + \frac{E_j}{T_i^2}. \quad (5)$$

Thus, to calculate the fraction of a component in the equilibrium vapor (2), it is necessary to determine the temperature at which condition (3) is satisfied.

Liquid Flow Period

According to the model, in the liquid flow period within the transport delay time, the liquid flow on trays takes place in the ideal displacement (plug flow) mode. After the transport delay time, the interaction between the tray fluid and the downward-flowing liquid occurs in the perfect mixing mode. The transport delay time is calculated using the following equation:

$$t_{\text{tr.d}} = \frac{1-\beta}{\eta}(1-\gamma)\tau, \quad (6)$$

where β is the degree of mixing between trays, which characterizes the proportion of the duration of the liquid flow in the perfect mixing mode.

If the transport delay time is higher than or equal to the liquid flow time, the downward-flowing liquid replaces the tray fluid according to the ideal displacement (plug flow) model. In this case, to calculate the fraction of the liquid on the tray by the end of the liquid flow period, the following algebraic equation is used:

$$X_i(\tau) = \eta X_{i+1}(\gamma\tau) + (1-\eta) X_i(\gamma\tau). \quad (7)$$

The fraction of the liquid on the feed tray by the end of the liquid flow period is calculated using the equation

$$X_f(\tau) = \frac{H_{f+1}}{H_f}\eta X_{f+1}(\gamma\tau) + (1-\eta) X_f(\gamma\tau) + \frac{G_F(1-\gamma)\tau}{H_f\gamma} X_F. \quad (8)$$

If the transport delay time is lower than the liquid flow time, the following equation is used to calculate the fraction of the liquid on the tray by the end of the liquid flow period:

$$X_i(\tau) = \beta Z_i(\tau) + \frac{\eta}{(1-\gamma)\tau}\int_{\tau - t_{\text{tr.d}}}^{\tau} Z_{i+1}(t)\mathrm{d}t. \quad (9)$$

The concentration of the light key in the liquid on the tray by the end of the liquid flow period depends on the concentration of the component in the mixing unit by the end of the liquid flow period and the integral that describes the variation of concentration in the mixing unit on the above-located tray for the time interval from $\tau - t_{\text{tr.d}}$ to τ. By *mixing unit,* we mean the portion of liquid on the tray in which there is no variation in the concentration of the light key during the plug flow of the liquid. Within the transport delay time, the fraction of the light key in the liquid that flows from the *i*th tray is equal to the fraction of the light key in the mixing unit of the given tray Z_i.

The fraction of the liquid on the feed tray by the end of the liquid flow period is calculated using the following equation:

$$X_f(\tau) = \beta Z_f(\tau) + \frac{H_{f+1}}{H_f}\frac{\eta}{(1-\gamma)\tau} \times \int_{\tau - t_{\text{tr.d}}}^{\tau} Z_{f+1}(t)\mathrm{d}t + \frac{G_F t_{\text{tr.d}}}{H_f\gamma} X_F. \quad (10)$$

The variations in the fraction of the light key in the mixing unit of the *i*th tray is described by the equation

$$\frac{\mathrm{d}Z_i(t)}{\mathrm{d}t} = \frac{\eta}{(1-\gamma)\tau\beta}\{U(t - t_{\text{tr.d}} - \gamma\tau) X_{i+1}(\gamma\tau) + [1 - U(t - t_{\text{tr.d}} - \gamma\tau)] X_i(\gamma\tau) - Z_i(t)\}, \quad (11)$$

where $U(t - t_{\text{tr.d}} - \gamma\tau)$ is the unit step function that takes on the value 0 when $t - t_{\text{tr.d}} - \gamma\tau \le 0$ and the value 1 when $t - t_{\text{tr.d}} - \gamma\tau > 0$. The fraction of the light key in the mixing unit on the *i*th tray at the beginning of the liquid flow period is equal to the fraction of the component in the liquid on the same tray and remains unchanged during the transport delay time as follows:

$$Z_i(\gamma\tau) = Z_i(\gamma\tau + t_{\text{tr.d}}) = X_i(\gamma\tau). \quad (12)$$

The variations in the fraction of the light key in the mixing unit of the feed tray is described by the following differential equation:

$$\frac{\mathrm{d}Z_f(t)}{\mathrm{d}t} = \frac{\eta}{(1-\gamma)\tau\beta}\left\{U(t - t_{\text{tr.d}} - \gamma\tau) \times \left[\frac{H_{f+1}}{H_f} X_{f+1}(\gamma\tau) + \frac{G_F(1-\gamma)\tau}{H_f\gamma} X_F\right] + [1 - U(t - t_{\text{tr.d}} - \gamma\tau)] X_f(\gamma\tau) - Z_f(t)\right\}. \quad (13)$$

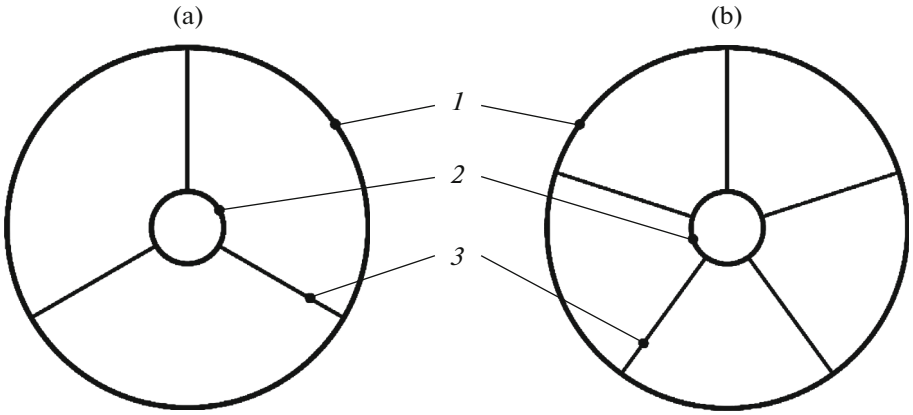

Fig. 2. Division of a column into sections: (a) three sections and (b) five sections; (*1*) external shell, (*2*) internal shell, and (*3*) partitions.

Thus, if the transport delay time is greater than or equal to the liquid flow time, the calculation of the liquid flow is performed using algebraic equations (7) and (8). If the transport delay time is lower than the liquid flow time, the mole fraction of the component in the liquid on the tray by the end of the liquid flow period is calculated by Eqs. (9) and (10) by solving differential equations (11) and (13).

The Bottom of the Column and the Reflux Condenser

Variations in the fraction at the bottom of the column at a constant liquid level is described by the following equation:

$$H_{\mathrm{W}}\frac{\mathrm{d}X_{\mathrm{W}}(t)}{\mathrm{d}t} = \eta H_1 X_1(t) - G_{\mathrm{V}}Y_{\mathrm{W}}(X_{\mathrm{W}}) - \left[G_{\mathrm{F}} - G_{\mathrm{V}}\frac{R-1}{R}\right]X_{\mathrm{W}}(t). \quad (14)$$

Here, $X_1(t)$ is the fraction of the light key in the liquid that flows from the bottom tray of the section operated in the liquid flow period.

The variations in the fraction of the light key in a reflux condenser at a constant liquid level are described by the following equation:

$$H_{\mathrm{D}}\frac{\mathrm{d}X_{\mathrm{D}}(t)}{\mathrm{d}t} = G_{\mathrm{V}}\left[Y_{\mathrm{N}}(t) - X_{\mathrm{D}}(t)\right]. \quad (15)$$

Here, $Y_{\mathrm{N}}(t)$ is the average fraction of the light key in the vapor that enters from the column sections operated in the vapor flow period.

METHOD OF CONTINUOUS SUPPLYING STREAMS TO A COLUMN

To organize the continuous supply of material streams to a cyclic distillation column, it is proposed to divide the column into sections in the vertical direction. Sectioning is performed by dividing the internal space of the column by vertical partitions into the given number of sections (Fig. 2). Contact devices (trays) are located along the height of each section. Each of the sections is connected to the common bottom and the common reflux condenser.

As contact devices, it is proposed to use trays developed by Maleta [12, 13], which ensure the single downward flow of the liquid without mixing. The design feature of Maleta's trays is that there are special chambers to which the liquid is supplied in the liquid flow period (Fig. 3). In the subsequent supply of the vapor, the liquid flows to the tray located below without mixing.

It is proposed to distribute the vapor stream among the sections of the column using a rotating cutoff plate that blocks the supply of the vapor to one of the sections (Fig. 4). Mass transfer takes place in the sections open for the supply of the vapor. The flow of the liquid occurs in the closed section. The rotation of the cutoff plate ensures the alternation of the vapor flow period and the liquid flow period in sections.

It is proposed to distribute the feed stream and the reflux stream among the sections of the column using buffers (Fig. 5). A buffer consists of a rotating cylindrical chamber (Fig. 5a) and a motionless section with connecting pipes (Fig. 5b). The distributed liquid is continuously supplied to the chamber through the lower opening *2* via the connecting pipe *4*. The liquid is supplied to one of the column sections through the upper opening *1* of the cylindrical chamber via the connecting pipes *3*.

The distributing cutoff plate and the rotating chambers of buffers are fastened to a movable shaft that passes through the center of the column. The mechanical connection of these devices into a unified rigid system simplifies the regulation process and excludes a mismatch between the supply of the vapor and the liquid to the column.

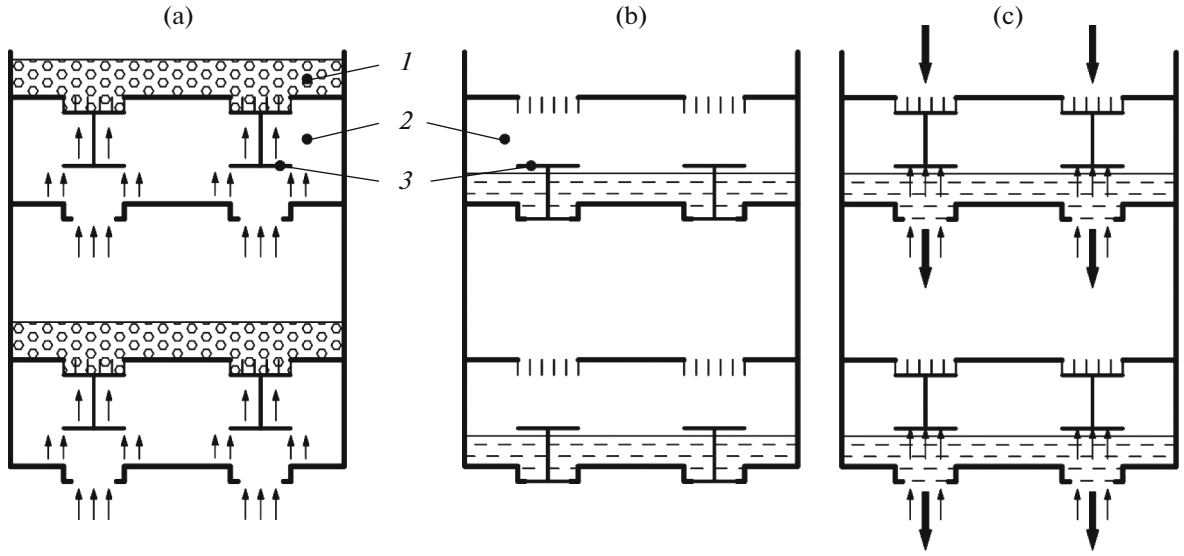

Fig. 3. Operating principle of trays proposed by Maleta: (a) vapor flow, (b) liquid flow into the sluice chamber, and (c) liquid flow to the below-located tray; (*1*) working zone of mass transfer, (*2*) sluice chamber, and (*3*) valve.

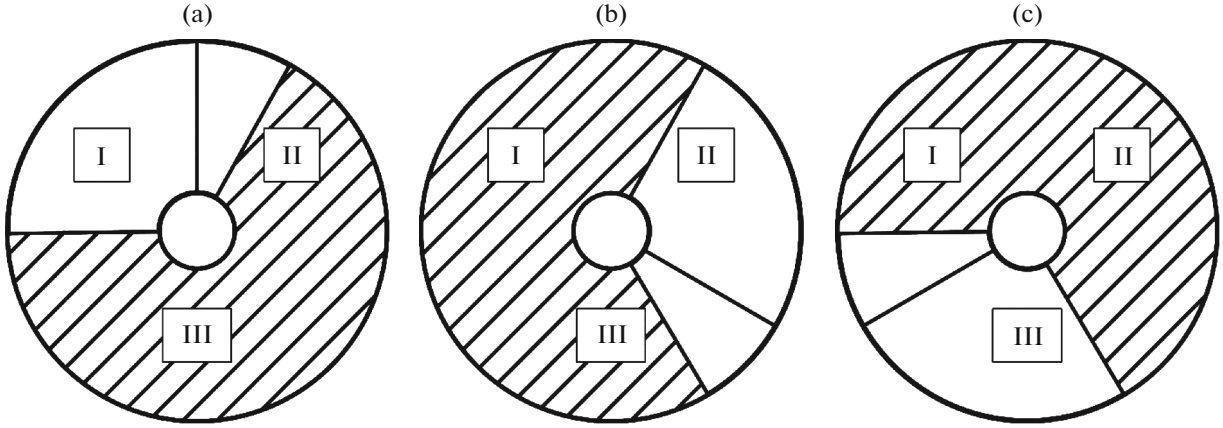

Fig. 4. Operating principle of a distributing cutoff plate in a three-section column: (a) supply of the vapor to sections I and II, (b) supply of the vapor to sections II and III, and (c) supply of the vapor to sections I and III.

CALCULATING THE CYCLIC DISTILLATION PROCESS

A column for separating an ideal model toluene–*ortho*-xylene mixture was chosen as the object of investigation with the following basic parameters: $X_F =$ 0.5 mol/mol, $N = 5$, $f = 3$, $E_M = 0.5$, $P_W = 101.3$ kPa, $P_D = 101.3$ kPa, $G_F = 100$ mol/h, $G_D = 50$ mol/h, $H_W = 5$ mol, and $H_D = 5$ mol.

The operating efficiency of the distillation column was estimated as follows using the thermodynamic criterion of the column separation efficiency ξ [22]:

$$\xi = \frac{\sum_{j=1}^{2} X_{F,j} \ln X_{F,j} - \varepsilon \sum_{j=1}^{2} X_{D,j} \ln X_{D,j} - (1-\varepsilon) \sum_{j=1}^{2} X_{W,j} \ln X_{W,j}}{\sum_{j=1}^{2} X_{F,j} \ln X_{F,j}}. \quad (16)$$

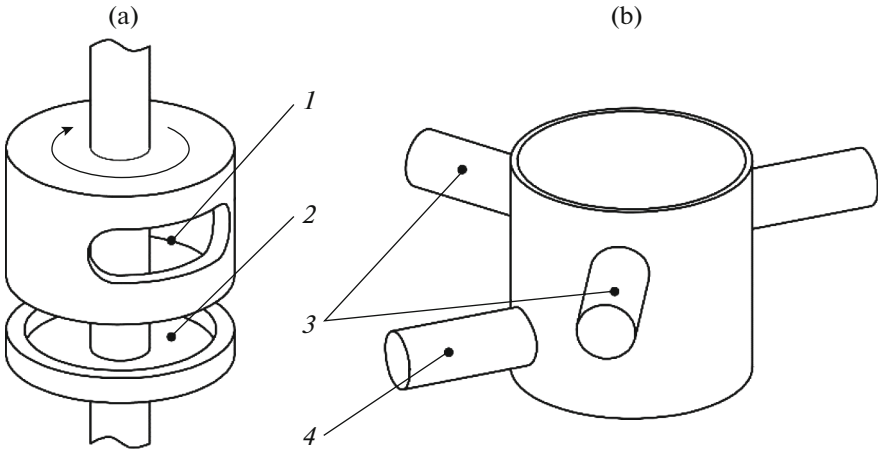

Fig. 5. Schematic representation of a buffer: (a) rotating chamber and (b) motionless section; (*1*) opening for the liquid flow from the chamber, (*2*) opening for the liquid flow into the chamber, (*3*) connecting pipe of the distribution device for the supply of the liquid to the column section, and (*4*) connecting pipe of the distribution device for the introduction of the liquid into the chamber.

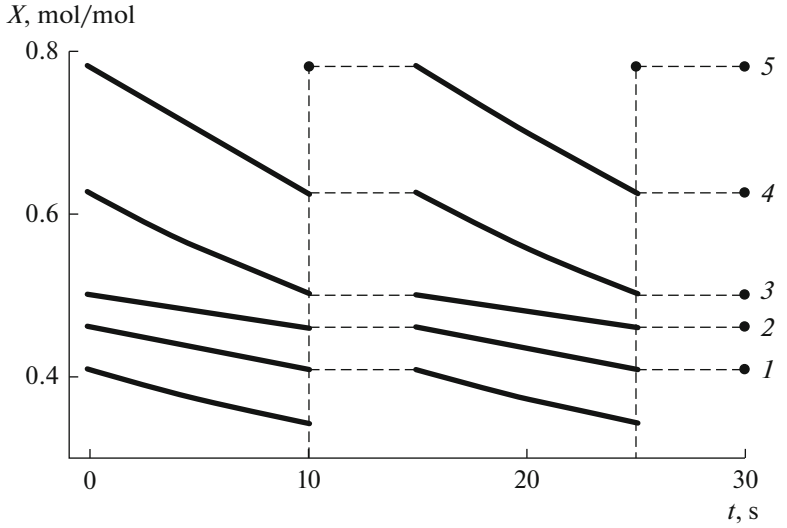

Fig. 6. Fraction of the light key in the liquid on trays as a function of time for section I in a three-section column ($\tau = 15$ s, $\gamma = 0.667$, $\beta = 0$, and $\eta = 1$): (*1*) tray 1, (*2*) tray 2, (*3*) tray 3, (*4*) tray 4, and (*5*) tray 5.

The variations in the fraction of toluene on the trays of section I in the operating mode for a three-section column are presented in Figs. 6 and 7. The flow of the vapor takes place over the time interval of 0–10 s, and the flow of the liquid occurs over the time interval of 10–15 s. The cycle is then repeated. Under the conditions of single replacement of the liquid in the ideal displacement mode (Fig. 6) in the vapor flow period, the fraction of toluene on the tray decreases to the fraction of toluene on the tray located below at the beginning of the vapor flow period as follows:

$$X_i(10) = X_{i+1}(0).$$

This regularity is disturbed at an insufficient fraction of the replaced fluid on the tray (Fig. 7). In this case, a decrease in the efficiency of separation is observed.

In the operating mode, variations in the fraction of toluene in the liquid on the tray in each of the sections are the same (Fig. 8).

In the mathematical model under consideration, the duration of the period τ and the proportion of the vapor flow period γ have no effect on the results of calculations. According to Eq. (1), in the vapor flow period, the variations in the fraction of the light key in the liquid on the tray is in inverse proportion to its volume on the tray. Since there is no flow of the liquid in the vapor flow period, the holding capacity of the tray with respect to the liquid is determined by the flow rates of the feed mixture and the reflux. The volume of

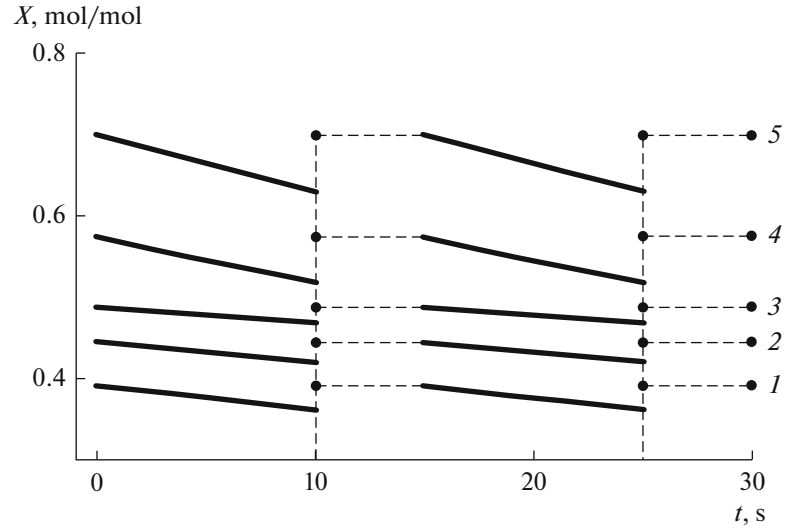

Fig. 7. Fraction of the light key in the liquid on trays as a function of time for section I in a three-section column ($\tau = 15$ s, $\gamma = 0.667$, $\beta = 0$, and $\eta = 0.5$): (*1*) tray 1, (*2*) tray 2, (*3*) tray 3, (*4*) tray 4, and (*5*) tray 5.

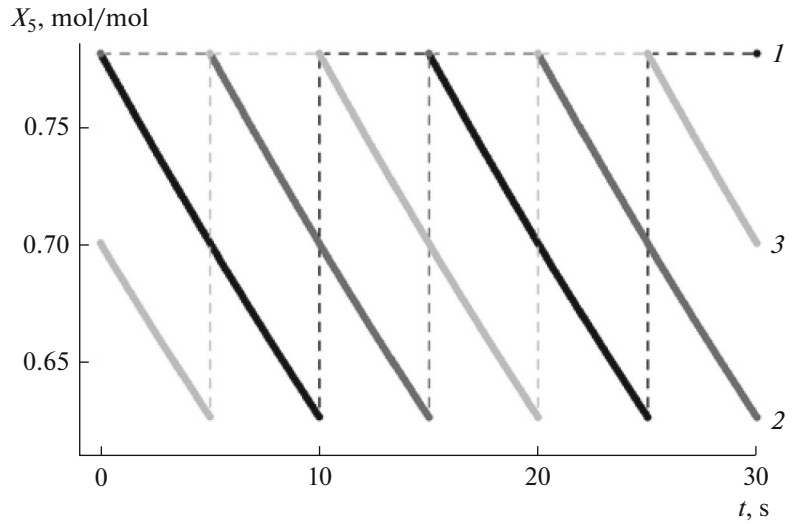

Fig. 8. Fraction of the light key in the liquid on the fifth tray as a function of time for a three-section column: (*1*) section I, (*2*) section II, and (*3*) section III.

the liquid on the tray increases in proportion to the duration of the vapor flow period. Therefore, by the end of the vapor flow period, the concentration of the light key is the same regardless of the cycle time, with other parameters of the process being unchanged. For the liquid flow period, the movement of the liquid depends on the quantity of liquid that flows from the tray, but it does not depend on the duration of the liquid flow period.

The effect of the fraction of the replaced fluid on the tray and the degree of mixing between trays is presented in Fig. 9. The maximum efficiency of separation is achieved under the conditions of single replacement of the fluid without mixing, which is in agreement with literature data [6–8]. When hydrodynamic conditions are different from the ideal displacement mode, it is inexpedient to replace the entire volume of the fluid on the tray. The maximum separation of components is achieved when the condition $\beta + \eta = 1$ is satisfied.

A model calculated in the Unisim Design software (Honeywell) was used to compare the efficiencies of cyclic distillation and conventional continuous distillation. The Peng–Robinson model was used as the model of the vapor–liquid equilibrium for continuous distillation [23].

It can be seen from Fig. 10 that the efficiency of separation in the cyclic mode is higher than the effi-

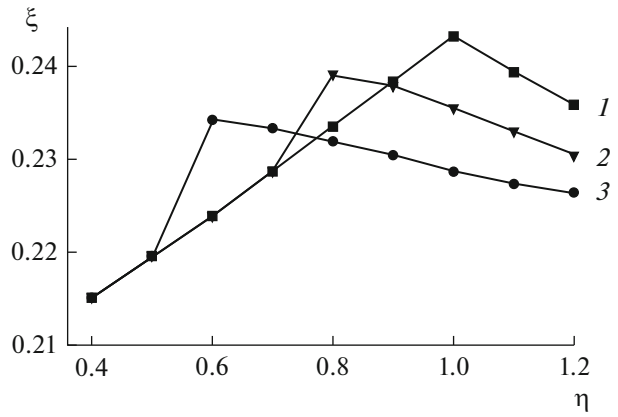

Fig. 9. Thermodynamic criterion of the column separation efficiency as a function of the fraction of the replaced fluid on the tray at $E = 0.5$ for $\beta =$ (*1*) 0, (*2*) 0.25, and (*3*) 0.5.

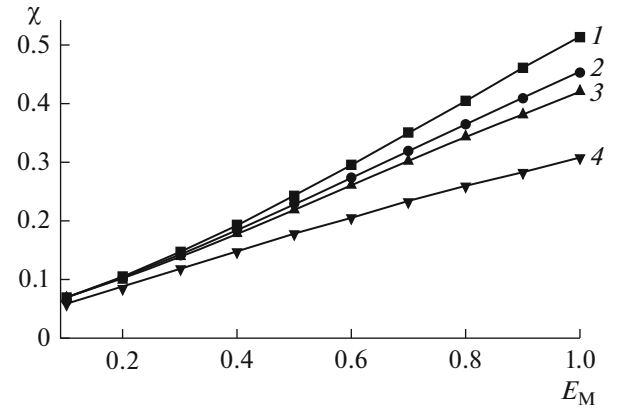

Fig. 10. Thermodynamic criterion of the column separation efficiency as a function of the Murphree tray efficiency: (*1*) cyclic distillation ($\beta = 0$ and $\eta = 1$), (*2*) cyclic distillation ($\beta = 0.5$ and $\eta = 1$), (*3*) cyclic distillation ($\beta = 0$ and $\eta = 0.5$), and (*4*) continuous distillation.

ciency of continuous distillation. The highest efficiency of cyclic distillation relative to continuous distillation is 166%, which is achieved at $E_M = 1$, $\beta = 0$, and $\eta = 1$.

VERIFICATION OF THE MODEL

The verification of the mathematical model of cyclic distillation was performed by comparing the results of calculations and experimental data on cyclic distillation presented in [7]. To verify the adequacy of the model, we used the data of 41 experiments on the separation of a methanol–ethanol mixture at total reflux for a wide range of parameters [7, pp. 175–186]: $G_W = 1.09$–4.30 m/s, $Y_W = 0.170$–0.414 mol/mol, $Y_{c.pl} = 0.205$–0.520 mol/mol, $\eta = 0.578$–1.580, and $E_M = 0.22$–0.77.

The proposed mathematical model takes into account the degree of mixing between trays β, which was not measured in experiments. This parameter depends on the design of a tray; therefore, the verification of the model was performed for the β range of 0–1 with a step of 0.1.

The root-mean-square deviation of the calculated values for the mole fraction of the light key on the control tray from experimental data was taken as a criterion for the adequacy of the mathematical model as follows:

$$S = \sqrt{\frac{\sum_{k=1}^{m}\left(X_k^{\text{exp}} - X_k^{\text{calc}}\right)^2}{N_{\text{exp}} - 1}}. \qquad (17)$$

For $\beta = 0.3$, the value of the criterion was 0.017 mol/mol. At an average value of $Y_W = 0.297$ mol/mol, the relative error of calculated values was 5.7%.

CONCLUSIONS

In this study, a mathematical model for cyclic binary distillation with the continuous supply of streams to a column has been proposed. The adequacy of the proposed model has been assessed using experimental data on cyclic distillation for the case of separating a methanol–ethanol mixture. The higher efficiency of a three-section column operated in the cyclic mode compared with a conventional distillation column has been shown for the case of separating a toluene–*ortho*-xylene mixture.

The effect of the parameters of the proposed model for cyclic distillation on the separation process has been studied. The highest efficiency of separation is achieved under the conditions of the single replacement of the fluid on the tray in the liquid flow period without mixing. When hydrodynamic conditions are different from the ideal displacement (plug flow) mode, it is inexpedient to replace the entire volume of the fluid on the tray.

A method has been proposed for organizing the continuous supply of material streams to a column by dividing the column into vertical sections and distributing the liquid and vapor streams among the sections.

NOTATION

A, B, C, D, E	constants in the extended Riedel equation
E_M	Murphree tray efficiency
f	number of the feed tray
G	flow rate, mol/s
H	tray holdup, mol
K	vapor–liquid distribution ratio
m	number of experiments
N	number of trays in the distillation column

n	number of components
P	pressure, Pa
R	reflux ratio
S	root-mean-square deviation
T	temperature, K
t	time, s
U	unit step function
X	mole fraction of the light key in the liquid, mol/mol
Y	mole fraction of the light key in the vapor, mol/mol
Y^*	equilibrium mole fraction of the light key in the vapor, mol/mol
Z	mole fraction of the light key in the mixing unit, mol/mol
β	degree of mixing between trays
γ	proportion of the vapor flow period
ε	fraction of the withdrawn overhead product
η	fraction of the replaced fluid on the tray
ξ	thermodynamic criterion of column separation efficiency
τ	column section cycle time, s

SUBSCRIPTS AND SUPERSCRIPTS

c.pl	control plate
calc	calculated value
D	reflux condenser
exp	experimental value
F	feed stream
f	number of the feed tray
i	number of the separation stage
j	number of the component
k	number of the experiment
tr.d	transport delay
V	vapor stream
W	bottom of the column

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