

## A NONEQUILIBRIUM MODEL OF DISTILLATION STAGE WITH POSSIBLE OCCURRENCE OF TWO LIQUID PHASES

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A model of the rectification column stage is proposed in which the possible splitting into two liquid phases may occur. For this case, the equilibrium stage concept has hitherto been employed in the literature. In this work it is shown that even in this situation, it is possible to use, under certain simplifying assumptions, the up-to-date nonequilibrium stage concept. For solving the proposed model, a global approach is suggested with retaining the problem dimension regardless of the number of coexisting liquid phases.

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For a long time, the traditional concept of equilibrium stage in its most different variants has been used for simulating the rectification. To correct the differences in behaviour between the real and equilibrium stage, the stage or vaporization efficiencies introduced in different ways have been employed. Even though this model concept may be adequate in many cases of binary mixtures, its disadvantages for multicomponent mixtures have been evident for a long time. Therefore, the nonequilibrium stage concept (sometimes also denoted as rate-based approach), such as published by Krishnamurthy and Taylor<sup>1</sup>, is more and more often applied in the most different regions of mathematical modelling of diffusion separation processes. Even though it is possible to find in the literature differently modified approaches to the calculation of mass transfer rate through the phase boundary<sup>2,3</sup>, it is possible to say that the principle remains the same. The extension of this concept in the chemical-engineering practice is best documented by the fact that the Aspen Technology, Inc., which concerns with the development of chemical-engineering software, furnishes already from 1988 the modulus RATEFRAC for calculating the tray and packed columns, stemming from the nonequilibrium stage concept<sup>4</sup> as one of the moduli of its simulation program ASPEN PLUS. The nonequilibrium stage concept has already been utilized in simulating and modelling, besides the common two-phase separation processes such as rectification and absorption in tray and packed columns, even for special situations such as, e.g., rectification accompanied by chemical reaction catalyzed with a dispersed solid catalyst in the liquid phase<sup>5</sup> or liquid extraction respecting the distribution in the dispersed phase<sup>6</sup>. Huang, Taylor and

Kooijman<sup>7,8</sup> have recently proposed the models originating from the nonequilibrium stage concept of a so-called second generation for a series of accompanying effects, such as the longitudinal backmixing, droplet entrainment, pressure drop, etc. Seader<sup>9</sup> reports that the introduction of the nonequilibrium stage concept could be the beginning of a new era in simulating and designing the separation processes.

Recently a number of works has appeared which deal with the computation of dynamic behaviour of stage rectification in the case when the splitting of the liquid phase into two liquid phases may take place, e.g.<sup>10-12</sup>. The authors have altogether used the Michelsen method<sup>13</sup> for the identification of possible existence of the second liquid phase. Eckert and Kubicek<sup>14</sup> have published a generalized model of equilibrium stage for the case when the liquid may split up to  $p$  liquid phases and exploited it successfully even for the case of columns with a possible existence of two and three liquid phases<sup>15</sup>. For the identification of the number of coexisting phases, they used their own method<sup>16</sup>. All these works stem from the out-of-date equilibrium stage concept. Lao<sup>17</sup> as the first proposed and verified several model concepts for the three-phase distillation employing the rate based approach.

In this work, a simplified mathematical model is proposed of the rectification column stage with a possible occurrence of two liquid phases which as well employs in its formulation the rate based approach. The model is formulated for a stationary state but its modification for dynamic behaviour should not make trouble.

## THEORETICAL

Let us consider the  $j$ -th stage of rectification column illustrated schematically in Fig. 1. For the sake of simplicity, no side feed and withdrawn streams are considered because it would only complicate the model clarity but would not mean a new quality from the point of view of formulation. Owing to the simplification, let us assume further besides the stationary state:

1. The spaces filled up with the vapour and the continuous liquid phase, in which the second liquid phase might be dispersed in the form of spherical drops, behave as ideal mixers, i.e., the composition and temperature of corresponding leaving streams correspond to those in these spaces. In case of the dispersed second liquid phase, this assumption means that the drop distribution as well as their composition and temperature in leaving liquid stream from the stage, consisting of the two liquid phases, correspond to the situation on the stage as well.

2. The drop volumes in single drop-classes and the number of these classes are given in advance (see Appendix).

3. All the drops of the  $k$ -th class of dispersed liquid leaving the stage have the same composition. With regard to the coalescence and splitting of the drops of different classes it is evident that, in fact, the composition of drops even of the same size should

differ. However, the assumption considered allows us to retain an acceptable model complexity.

4. Only the vapor or continuous liquid phase can exchange heat with surroundings.

5. The axial mixing along the column is considered in none of the phases which would be caused, e.g., by droplet entrainment of the continuous and dispersed liquid phase, by weeping, etc.

6. The mass transfer takes place only between the vapour and continuous liquid phase and between the continuous and dispersed liquid phase. The mass transfer by diffusion between the vapour and liquid phase as well as between drops of different sizes does not take place with respect to the probable very short contact time and small interfacial area between the vapor bubbles (or vapour above the level) and the drops of dispersed liquid phase.

7. The leaving continuous and dispersed liquid phases have the same temperature. For the same reason as in the foregoing point, we do not consider the heat exchange between the vapour and dispersed liquid phase.

Under these assumptions we can write the component balances in the vapour phase

$$0 = V_{j+1}y_{i,j+1} - V_j y_{i,j} - N_{i,j}^* \quad i = 1, \dots, I \quad (I)$$

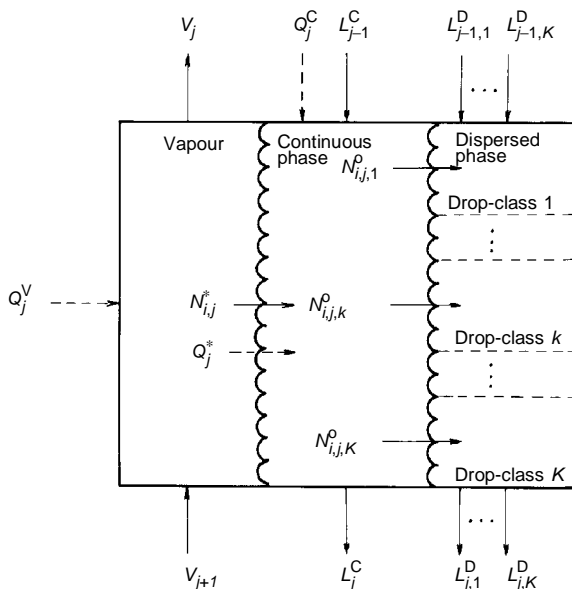


FIG. 1

Nonequilibrium  $j$ -th stage of rectification column with possible occurrence of two liquid phases

Analogously we write the component balances for the continuous liquid phase

$$0 = L_{j-1}^C x_{i,j-1}^C + N_{ij}^* - L_j^C x_{ij}^C - \sum_{k=1}^K N_{ij,k}^o \quad i = 1, \dots, I \quad (2)$$

and the component balances of the whole dispersed liquid phase

$$0 = \sum_{k=1}^K L_{j-1,k}^D x_{i,j-1,k}^D + \sum_{k=1}^K N_{ij,k}^o - \sum_{k=1}^K L_{j,k}^D x_{ij,k}^D \quad i = 1, \dots, I \quad (3)$$

completed by the component balances of  $K - 1$  classes of dispersed liquid phase

$$0 = L_{j-1,k}^D x_{i,j-1,k}^D + N_{ij,k}^o + (G_{j,k} - L_{j,k}^D) x_{ij,k}^D \quad i = 1, \dots, I, k = 1, \dots, K - 1 \quad (4)$$

Term  $G$  represents a source of drops of the  $k$ -th class by splitting and coalescing from drops of other classes. The way of its possible expressing will be shown in Appendix. The given formulation of Eqs (3) and (4) ensures the fulfilment of condition  $\sum_{k=1}^K G_{j,k} = 0$  and simultaneously the assumed re-distribution of dispersed liquid phase only to drops of permissible classes.

As next one we will write the enthalpy balances of the vapour phase

$$0 = V_{j+1} H_{j+1} - V_j H_j - \sum_{i=1}^I N_{ij}^* H_{ij}^V + Q_j^V - h_j^V a_j^* \frac{\varepsilon_j^V (T_j^V - T_j^*)}{\exp(\varepsilon_j^V) - 1} \quad (5)$$

and of both the liquid phases

$$0 = L_{j-1}^C H_{j-1}^C + \sum_{k=1}^K L_{j-1,k}^D H_{j-1,k}^D + \sum_{i=1}^I N_{ij}^* H_{ij}^C + h_j^C a_j^* (T_j^* - T_j^C) - L_j^C H_j^C - \sum_{k=1}^K L_{j,k}^D H_{j,k}^D + Q_j^C \quad (6)$$

The expression  $\varepsilon_j^V / (\exp(\varepsilon_j^V) - 1)$  in relation (5) serves for correcting the heat transfer coefficient in the vapour phase to mass transfer. The way of its calculation will be mentioned in Appendix. In case of heat transfer coefficient in the liquid phase, this correction may be neglected<sup>2</sup>.

Now let us write down the equations for mass transfer which are, between two phases, always only  $I - 1$  independent. For the mass transfer from the vapour into the continuous liquid phase, we can thus write

$$0 = N_{ij}^* - J_{ij}^{Y,*} - y_{ij} \sum_{m=1}^I N_{m,j}^* \quad i = 1, \dots, I-1 \quad (7)$$

$$0 = N_{ij}^* - J_{ij}^{C,*} - x_{ij}^C \sum_{m=1}^I N_{m,j}^* \quad i = 1, \dots, I-1 \quad (8)$$

Analogously we can write for the mass transfer from the continuous liquid phase into the dispersed liquid phase

$$0 = N_{ij,k}^o - J_{ij,k}^{C,o} - x_{ij,k}^C \sum_{m=1}^I N_{m,j,k}^o \quad i = 1, \dots, I-1, k = 1, \dots, K \quad (9)$$

$$0 = N_{ij,k}^o - J_{ij,k}^{D,o} - x_{ij,k}^D \sum_{m=1}^I N_{m,j,k}^o \quad i = 1, \dots, I-1, k = 1, \dots, K \quad (10)$$

The equation for heat transfer from the vapour into the continuous liquid phase will be

$$0 = h_j^Y a_j^Y \frac{\epsilon_j^Y (T_j^Y - T_j^*)}{\exp(\epsilon_j^Y) - 1} + \sum_{i=1}^I N_{ij}^* H_{ij}^Y - h_j^C a_j^C (T_j^* - T_j^C) - \sum_{i=1}^I N_{ij}^* H_{ij}^C \quad (11)$$

On the assumption of thermodynamic equilibrium on phase boundaries, we can write for the interface vapour–continuous liquid phase

$$0 = \psi_{ij}^* x_{ij}^* - y_{ij}^* \quad i = 1, \dots, I \quad (12)$$

Analogously for the interface continuous liquid phase–dispersed liquid phase we can write

$$0 = \gamma_{ij,k}^{C,o} x_{ij,k}^{C,o} - \gamma_{ij,k}^{D,o} x_{ij,k}^{D,o} \quad i = 1, \dots, I, k = 1, \dots, K \quad (13)$$

For single mole fractions, the constraints hold which follow from their definitions: In vapour

$$0 = \sum_{i=1}^I y_{ij} - 1 \quad (14)$$

$$0 = \sum_{i=1}^I y_{ij}^* - 1 \quad , \quad (15)$$

in the continuous liquid phase

$$0 = \sum_{i=1}^I x_{ij}^C - 1 \quad (16)$$

$$0 = \sum_{i=1}^I x_{ij}^* - 1 \quad (17)$$

$$0 = \sum_{i=1}^I x_{i,j,k}^{C,o} - 1 \quad k = 1, \dots, K \quad , \quad (18)$$

or in the dispersed liquid phase

$$0 = \sum_{i=1}^I x_{i,j,k}^{D,o} - 1 \quad \text{or} \quad 0 = L_{j,k}^D \quad k = 1, \dots, K \quad (19)$$

$$0 = \sum_{i=1}^I x_{i,j,k}^D - 1 \quad k = 1, \dots, K \quad . \quad (20)$$

## DISCUSSION

If we assume that the pressure on the stage is known, and the amount, composition and temperature of all the input streams into the stage as well as the size of heat fluxes are given, it is possible to consider relations (1)–(20) as a system of equations

$$0 = F(X) \quad , \quad (21)$$

where  $\dim F = \dim X = 5I + 4IK + K + 5$  and the vector of unknowns  $X$  consists of  $V_j, y_{ij}, N_{ij}^*, L_j^C, x_{ij}^C, L_{j,k}^D, x_{i,j,k}^D, N_{i,j,k}^o, T_j^V, T_j^*, T_j^C, y_{ij}^*, x_{ij}^*, x_{i,j,k}^{C,o}, x_{i,j,k}^{D,o}$  for  $i = 1, \dots, I$  and  $k = 1, \dots, K$  at the variables which contain these subscripts. The reader will notice that in the vector of unknowns, even the variables are considered which refer to all the classes of dispersed liquid phase which, in fact, need not exist on each stage. This is typical of the application of a so-called global approach to the solution when the total dimension of

the problem is not changed and different phase situations are realized only by interchanging the relations as it has been made in our work<sup>16</sup> and as we shall show below for this case considered. To solve system of equations (21), it is possible to use, e.g., some of modifications of the Newton method. Very important is the initial guess of the number of coexisting phases and the other initial values of variables connected with it. From experience follows that it is suitable to assume the presence of the second liquid phase<sup>16</sup> when choosing the initial values, in the given case in all the drop-classes. The alternative in relation (19) means the replacement of the first relation given there by the relation which defines that the certain drop-class of dispersed phase does not exist. Then for this class, the respective constraint of the sum of mole fractions on the respective interface need not be fulfilled. If, in the course of iteration process, the case occurs that for hitherto non-existing smallest drop-class, the sum of its mole fractions begins

to approach unity, which can be tested, e.g., as  $\sum_{i=1}^I x_{ij,1}^{D,o} > 0.99999$ , we insert the first

relation in Eq. (19) for  $k = 1$  into the considered system of equations instead of the up-to-now considered second relation. In this way we model the formation of dispersed liquid phase in the form of smallest droplets. On the contrary, if the case occurs in the course of iteration calculation, that the flow rate of hitherto existing  $k$ -th drop-class turns non-positive, i.e.,  $L_{j,k}^D \leq 0$ , we exchange the up to now considered first relation in Eq. (19) for the second (for the respective  $k$ ). In this way we model the extinction of drops of arbitrary drop-class. The smallest amount of dispersed liquid phase of  $k$ -th class corresponds just to one drop of this class, which, however, is not a condition in the given formulation. Need for this exchange is tested always after several (e.g., two) iteration steps.

Let us mention that in the given model formulation it is assumed that always the vapour and continuous liquid phase leave the tray, i.e., the case does not take place when all the incoming liquid is evaporated or all the incoming vapour is condensed owing to the supplied (taken away) heat. If the stage should function as flash, it would be necessary, such as in Eq. (19), to exchange moreover the relations in Eqs (15) and (17) for those defining the non-existence of the respective leaving phase.

Relations (1)–(13) contain a number of auxiliary functions whose enumeration will be dealt with in Appendix for the sake of clear arrangement.

## CONCLUSIONS

The development of modern methods of computing the separation equipments was enabled only by the considerable advancement of computer technique in the last thirty years. With this advancement, it is possible to observe also the effort to give more precision to the actual description of the process. Simultaneously, one passes over from the decomposed approach to the global one. This logical development can be watched

not only in the stationary but also in dynamic modelling where the standard software for solving the differential-algebraic systems<sup>18</sup> is used even for the solution of multi-phase rectification<sup>12</sup>.

The methodology presented in this work, which can be naturally simply (by inserting feeds, side withdrawals, etc.) transferred to the description of the entire rectification column including boiler and condenser, outlines, in addition to the models proposed by Lao<sup>17</sup>, another method how, even in the case of multiphase rectification, to employ the modern rate based approach. The modification of this approach to the dynamic model is of course possible as well.

Numerical verification of the proposed model forms the subject of next paper. Nevertheless it is to be expected that the agreement of calculated values with experimental data should be at least as good as that achieved by Lao<sup>17</sup> in his best model IV which could be derived from the model presented here by introducing further simplifying assumptions. But it is necessary to realize that the number of simultaneously solved equations considerably increases unlike the common equilibrium concept. In case of three components on considering five drop-classes of the dispersed second liquid phase, it is in the proposed model 85 equations for each stage in comparison with 13 equations for the equilibrium model. It is evident that the possibility of neglecting the concentration or temperature gradient between the bulk phase and interface in some phase, which corresponds to the infinitely large mass or heat transfer coefficient in this phase, would allow us to diminish the number of equations in the model and so also the simplification and acceleration of the computation. The question whether the complex approach proposed here is of any sense, is obvious especially if we consider a considerable uncertainty in the estimation of a number of model parameters calculated from regression relation obtained experimentally (see Appendix) which are themselves subject to a great error. Such a question, however, always was, is and will be in the future important not only in considerations of the advancement of this region of chemical engineering.

## APPENDIX

### *Enumeration of Auxiliary Functions*

A number of functions occur in Eqs (1)–(13) which must be enumerated in the course of computation. The enumeration of molar and partial molar enthalpies  $H$  in the liquid and vapour phase as well as the enumeration of distribution coefficients between the continuous liquid and vapour phase  $\psi$ , activity coefficients in the liquid phase  $\gamma$  and molar densities of all phases  $\rho$  are undoubtedly well-known to the reader and therefore we will not treat them.

The important quantity in the presented model is the source term  $G$  in Eq. (4) which represents the effect of splitting and coalescence of drops. One of the possibilities how it can be formulated is, e.g., use of HD (homogeneous distribution of the sources)



model by Jiricny et al.<sup>19</sup>. The discrete distribution of drops into drop-classes must be postulated in advance:

Set of eligible drop sizes of dispersed phase (as far as it exists) contains drops of minimum given volume  $v_1^D$  and drops of maximum volume  $v_K^D$ .

All drops of the set have volumes equal to multiples of volume of the smallest drop

$$v_k^D = kv_1^D \quad k = 1, \dots, K. \quad (A1)$$

The source term for drops of  $k$ -th class can then be formulated as follows

$$G_{j,k} = \left[ \sum_{l=1}^M (k_{l,k}^c n_{j,l} n_{j,k-l} - k_{k,l}^d n_{j,k}) + \sum_{l=k+1}^K (1 + \delta_{l-k,k}) (k_{l,k}^d n_{j,l} - k_{k,l}^c n_{j,k} n_{j,l-k}) \right] v_{S,j} v_k^D \rho_{j,k}^D. \quad (A2)$$

Here we have omitted, for the sake of simplicity, the stage subscript at rate constants for splitting  $k^d$  and coalescence  $k^c$  whose estimation will have a great significance for the model reliability. For  $M$  in Eq. (A2) holds

$$M = \frac{k - [1 - (-1)^k / 2]}{2}. \quad (A3)$$

The first summation term in Eq. (A2) represents the coalescence of drops of  $k$ -th class from smaller drops and simultaneously their splitting, the second term represents the formation of drops of  $k$ -th class by splitting larger drops and reverse synthesis of larger drops with drops of  $k$ -th class as one of participants of this synthesis. Similarity of these relations with those from reaction kinetics is evident.

Number concentration of drops of  $k$ -th class on the stage can be, under the above-mentioned assumption that the mixture of liquids leaves the stage in the same "mixed" state as it occurs on it, expressed by the relation

$$n_{j,k} = \frac{L_{j,k}^D}{K \rho_{j,k}^D v_k^D \left( \sum_{m=1}^K L_{j,m}^D / \rho_{j,m}^D + L_j^C / \rho_j^C \right)}. \quad (A4)$$

Now let us pay attention to Eqs (7)–(10). In literature one can find several ways of their formulation and enumeration. In front of the summation terms, the arithmetic or logarithmic average value of concentration between the bulk of the respective phase on the stage and the concentration in the inlet stream of this phase coming on the plate<sup>3</sup> is sometimes used instead of the concentration in the bulk of the respective phase. Similar approach is applied even for temperatures  $T_j^V$  and  $T_j^C$  in Eqs (5) and (6). However, the

way of solution of Eqs (7)–(10) and the enumeration of transport terms following from it appears to be more substantial. As it has been said, there exist several approaches<sup>3</sup> from which the method by Krishna and Standard<sup>20</sup> based on the exact solution of Maxwell–Stefan equations and the method based on the linearization theory by Toor<sup>21</sup> and Stewart and Prober<sup>22</sup> are recommended most often. However, in none of these approaches we dispense with the enumeration of binary mass transfer coefficients in the respective phase whose values are decisive for the model reliability. For the case of occurrence of the second liquid phase on the stage, there exist no verified methods of their estimation in the literature. Lao<sup>17</sup> used in his work the procedures originating from theoretical concepts on the flow of phases on stage. On the assumption that the amount of dispersed liquid phase is not large, it would be possible to employ a suitably modified AIChE method<sup>23</sup> in the case of tray column and the mass transfer between the vapour and continuous liquid phase. It provides the value of mass transfer coefficient already multiplied by the interfacial area both for the continuous liquid phase and for the vapour phase. A survey of recent methods which could be used for the given purpose even for packed columns is reported by Taylor et al.<sup>8</sup> For the mass transfer coefficient between the continuous and dispersed phase, it would similarly be possible to use some of regression relations published for the extraction usually in the form of dependence of the Sherwood number on the Reynolds and Schmidt numbers, see, e.g.<sup>24</sup>.

Inserting the drop population model makes it possible, on the assumption of spherical shape of drops of dispersed liquid phase, to estimate the interfacial area between the  $k$ -th drop-class of dispersed liquid phase and continuous liquid phase directly from the relation

$$a_{j,k}^0 = 4.836 v_{S,j} n_{j,k} (v_k^D)^{2/3} . \quad (\text{A5})$$

Parameter  $\varepsilon$  in Eq. (5) is defined<sup>20</sup> as

$$\varepsilon_j^V = \left[ \sum_{i=1}^I N_{i,j}^* C_{pi}^V / h_j^V a_j^* \right] . \quad (\text{A6})$$

The estimation of heat transfer coefficient in the vapour and continuous liquid phase is usually done on the basis of the Chilton–Colburn analogy<sup>25</sup> from the average value of mass transfer coefficient, Lewis number and the value of molar heat capacity in the respective phase.

## SYMBOLS

$a$	interfacial area, $\text{m}^2$
$C_p$	molar heat capacity at constant pressure, $\text{J mol}^{-1} \text{K}^{-1}$

$F$	vector of equations
$G$	source of drops, $\text{mol s}^{-1}$
$H$	molar or partial molar enthalpy, $\text{J mol}^{-1}$
$h$	heat transfer coefficient, $\text{W m}^{-2} \text{K}^{-1}$
$I$	number of components
$J$	transport term in Eqs (7)–(10), $\text{mol s}^{-1}$
$K$	number of drop-classes
$k^c$	rate constant of coalescence, $\text{m}^3 \text{s}^{-1}$
$k^d$	rate constant of dispersion (drop splitting), $\text{s}^{-1}$
$L$	liquid phase flow rate, $\text{mol s}^{-1}$
$M$	summation limit
$N$	mass transfer rate through phase boundary, $\text{mol s}^{-1}$
$n$	number concentration of drops, $\text{m}^{-3}$
$Q$	heat flux, $\text{W}$
$T$	temperature, $\text{K}$
$V$	vapour phase flow rate, $\text{mol s}^{-1}$
$v$	volume, $\text{m}^3$
$v_s$	total hold-up on stage, $\text{m}^3$
$X$	vector of unknowns
$x$	mole fraction in liquid phase
$y$	mole fraction in vapour phase
$\gamma$	activity coefficient
$\delta$	Kronecker delta
$\varepsilon$	parameter defined by Eq. (A6)
$\Psi$	continuous liquid phase–vapour distribution coefficient
$\rho$	molar density, $\text{mol m}^{-3}$

## Subscripts

$i$	component $i$
$j$	stage $j$
$K$	class of largest drops
$k$	drop-class $k$
$l, m$	auxiliary index

## Superscripts

C	continuous liquid phase
D	dispersed liquid phase
V	vapour phase
*	value at vapour–continuous liquid phase interface
o	value at continuous liquid phase–dispersed liquid phase interface

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