

# Distillation Models for Two Partially Immiscible Liquids

GUIDO BUZZI FERRARIS

and

MASSIMO MORBIDELLI

Istituto di Chimica Industriale  
Politecnico di Milano  
Piazza Leonardo Da Vinci, 32  
20133 Milano, Italy

We hereby submit three different numerical methods for the solution of the multicomponent distillation model for two partially immiscible liquids. A procedure to define the real number of phases present on each stage has been developed. The proposed Newton-Raphson method allows efficient solution of problems with a large selection of the unknowns and the specification equations.

## SCOPE

A large amount of literature has been devoted to the modeling of multicomponent separators in which two phases are present. However, the case with three phases, two liquids and one vapor, has received much less attention (Block and Hegner 1976). These distillation columns are very frequent in the chemical engineering processes. For example, the liquid-phase separation on the top of the column, with one phase refluxed to the column and the other one withdrawn as a distillate, or the introduction of a side stream phase separator can improve the separation efficiency of the distillation column. Besides the considerable progress made in the last few years in describing the multicomponent three phases equilibria: liquid-liquid-vapor, allows modeling of such units.

In developing a computer program for the solution of the large system of nonlinear equations which constitutes the model, two problems have to be considered. The first problem is the difficulty to define the number of phases present on each stage that is usually unknown until the computations are performed. The second problem is the possibility to give as an input different kind of specification equations. In particular, it can be useful to assign the values of some particular variables (or their functions) such as the molar fraction of one component on a particular stage, the flow rate of one component from a stage, or its ratio with respect to another flow rate on another stage, or to the feed itself.

## CONCLUSIONS AND SIGNIFICANCE

Three different numerical methods for the solution of the mathematical model of a multicomponent multistaged separator in which the liquid phase separation can occur, are offered. Each of these methods shows different characteristics and efficiency in the way they reach the solution of the problem. Therefore, they have been combined into a computer program which switches from one method to the other in order to optimize the required amount of computation time. However, the core of the developed computer program is the Newton-Raphson method which appeared to be the most powerful and efficient in reaching the solution.

Because a three-phase system always admits also a two-phase pseudo-solution, the number of phases present on each

stage has to be assigned "a priori" and can not remain unspecified, when we require a unique solution problem. Therefore, the obtained solution is verified through the multflash method to make sure the liquid phase separation does not in actual fact take place, in all those stages where two phases only were assumed. This method then also provides a powerful tool to pin down the real phase type of the distillation column under examination.

The offered computer program can solve problems in which a large variety of particular specification equations are given. This provides a useful tool to the process engineer who can directly impose to the problem the more useful requirements.

## BASIC EQUATIONS

Figure 1 shows the generic  $j$ -th stage of the three-phase multicomponent, multistaged separator. On each stage we may consider the presence of a feed stream, side streams of both the two liquid phases, a side stream of the vapor phase and a withdrawal or an introduction of heat. The mathematical model of the  $j$ -th stage, where equilibrium conditions are assumed,

Correspondence concerning this paper should be addressed to G. Buzzi Ferraris. M. Morbidelli is presently with Istituto di Chimica Fisica, Elettrochimica e Metallurgia del Politecnico, Milano, Italy.

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consists of the material balances and the equilibrium relationships of each component  $i$ , and of the enthalpy balance, the global material balance and the two stoichiometric conditions.

With reference to Figure 1, in which the second liquid phase is indicated with an apex, these equations can be written as follows:

• Material balance of the  $i$ -th component on the  $j$ -th stage ( $V_{N+1} = L_0 = L'_0 = 0$ )

$$L_{j-1}x_{i,j-1} + L'_{j-1}x'_{i,j-1} - (L_j + U_j)x_{i,j} \\ - (L'_j + U'_j)x'_{i,j} - (V_j + W_j)y_{i,j} + V_{j+1}y_{i,j+1}$$

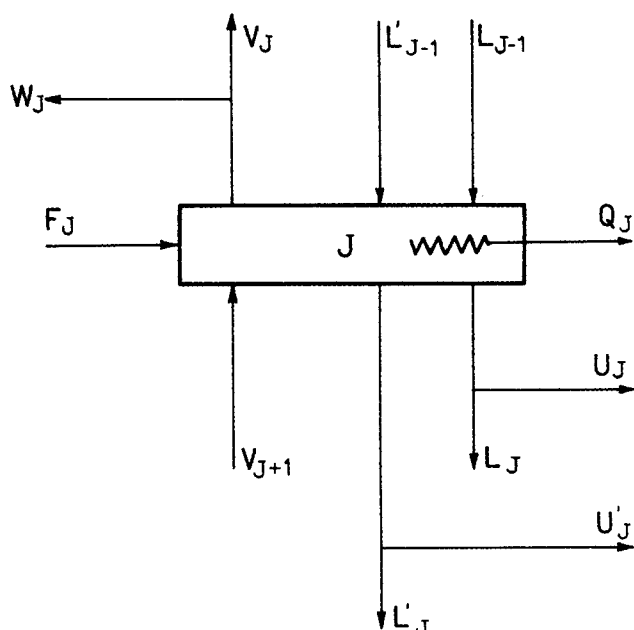


Figure 1. Schematic diagram of the separator stage.

$$+ F_j z_{i,j} = 0 \quad (i = 1, M; j = 1, N) \quad (1)$$

• Equilibrium equations of the  $i$ -th component on the  $j$ -th stage

$$y_{i,j} - K_{i,j} x_{i,j} = 0 \quad (2)$$

$$y_{i,j} - K'_{i,j} x'_{i,j} = 0 \quad (i = 1, M; j = 1, N) \quad (3)$$

or

$$x'_{i,j} - K^+_{i,j} x_{i,j} = 0 \quad (3')$$

• Stoichiometric equations on the  $j$ -th stage

$$\sum_n (y_{n,j} - x_{n,j}) = 0 \quad (4)$$

$$\sum_n (y_{n,j} - x'_{n,j}) = 0 \quad (j = 1, N) \quad (5)$$

or

$$\sum_n (x_{n,j} - x'_{n,j}) = 0 \quad (5')$$

• Global material balance on the  $j$ -th stage

$$L_{j-1} + L'_{j-1} - (L_j + U_j) - (L'_j + U'_j) - (V_j + W_j) + V_{j+1} + F_j = 0 \quad (j = 1, N) \quad (6)$$

• Enthalpy balance on the  $j$ -th stage

$$L_{j-1} h_{j-1} + L'_{j-1} h'_{j-1} - (L_j + U_j) h_j - (L'_j + U'_j) h'_j - (V_j + W_j) H_j + V_{j+1} H_{j+1} + F_j H F_j - Q_j = 0 \quad (j = 1, N) \quad (7)$$

It is important to point out that this system always admits the trivial solution in which  $x_{i,j} = x'_{i,j}$ , that is the two liquid phases are coincident. This corresponds to the two-phase solution which always exists, whether or not the liquid phase separation occurs (Henley and Rose, 1969).

## SPECIFIED AND UNSPECIFIED VARIABLES

Equations 1 to 7 constitute a non linear system of  $(3M + 4)N$  equations. Different choices of the unspecified and specified variables are available to solve the system. We will refer to the following set of variables:  $y_{i,j}$ ,  $x_{i,j}$ ,  $x'_{i,j}$ ,  $T_j$ ,  $V_j$ ,  $L_j$  and  $L'_j$  as the "standard set of unspecified variables," and consequently  $P_j$ ,  $U_j$ ,  $U'_j$ ,  $W_j$ ,  $Q_j$ ,  $F_j$ ,  $z_{i,j}$  and  $H F_j$  will constitute the "standard set of specified variables."

However, it is interesting, for practical purposes, to also consider problems with a different choice of the unspecified

variables. In this case one or more of the specified variables of the "standard set" can be considered as unknown, and likewise one or more equations have to be added to the system. These equations can either be very simple (for example,  $L_5 = 100$ .) or complex functions of all present unspecified variables (for example,  $L_{5,3.5}/F_{5,3.8} = 0.9$ ). The possibility to solve this kind of problems depends upon the used mathematical algorithm as we will show shortly.

The physicochemical functions describing the vapor-liquid ( $K_{i,j}$ ,  $K'_{i,j}$ ) and the liquid-liquid equilibria ( $K^+_{i,j} = K_{i,j}/K'_{i,j}$ ), as well as the enthalpy functions ( $h_j$ ,  $h'_j$ ,  $H_j$ ), are assumed to be known functions of the temperature, pressure and phase composition.

The system of Eqs. 1 to 7 describes a multicomponent, multistaged separator with three phases in equilibrium on each stage. In most cases of import the liquid phase separation occurs only on few stages. Therefore the solving system has to be modified to describe those stages where only two phases are present. Also this possibility depends upon the used solving method and therefore it will be described in detail in the following.

## BOILING POINT TYPE METHOD

This method splits the solving system 1 to 7 in three smaller subsystems which are solved separately. The sequence of solution of the subsystems is similar to the one applied by the well known "BP method" for two phases distillation columns (Wang and Henke, 1966; Ball and Beitrag, 1961; Boston and Sullivan, 1972). For this reason we will refer to this method as a boiling point type method, although the subsystems are different.

A method of this type has been proposed for the solution of three-phase distillation columns by Block and Hegner (1976).

The following variables can be defined:

$$\bar{L}_j = L_j + U_j + L'_j + U'_j \quad (8)$$

$$\eta_j = (L_j + U_j)/\bar{L}_j \quad (9)$$

$$\bar{x}_{i,j} = \eta_j x_{i,j} + (1 - \eta_j) x'_{i,j} \quad (10)$$

where  $\bar{L}_j$  and  $\bar{x}_{i,j}$  can be regarded as the overall flowrate and the overall composition of the liquid streams leaving the  $j$ -th stage, respectively;  $\eta_j$  is the phase split parameter describing the phase separation of the liquid at the  $j$ -th stage. If  $\eta_j = 1$ ,  $L'_j = 0$  and  $U'_j = 0$ ; if  $\eta_j = 0$ ,  $L_j = 0$  and  $U_j = 0$ ; if  $0 < \eta_j < 1$  both liquid phases are present. The system of Eqs. 1 to 10 can be rearranged and splitted in the following three groups of equations:

Systems 1 ( $j = 1, N$ )

$$\bar{x}_{i,j} - x_{i,j} \eta_j - (1 - \eta_j) x'_{i,j} = 0 \quad (i = 1, M) \quad (11)$$

$$y_{i,j} - K_{i,j} x_{i,j} = 0 \quad (i = 1, M) \quad (12)$$

$$y_{i,j} - K'_{i,j} x'_{i,j} = 0 \text{ or } x'_{i,j} - K^+_{i,j} x_{i,j} = 0 \quad (i = 1, M) \quad (13, 13')$$

$$\sum_n (y_{n,j} - x_{n,j}) = 0 \quad (14)$$

$$\sum_n (y_{n,j} - x'_{n,j}) = 0 \text{ or } \sum_n (x_{n,j} - x'_{n,j}) = 0 \quad (15, 15')$$

System 2

$$L_j + U_j - \eta_j \bar{L}_j = 0 \quad (j = 1, N) \quad (16)$$

$$L'_j + U'_j - (1 - \eta_j) \bar{L}_j = 0 \quad (j = 1, N) \quad (17)$$

$$L_{j-1} + L'_{j-1} - \bar{L}_j - (V_j + W_j) + V_{j+1} + F_j = 0 \quad (j = 1, N) \quad (18)$$

$$L_{j-1} h_{j-1} + L'_{j-1} h'_{j-1} - (L_j + U_j) h_j - (L'_j + U'_j) h'_j - (V_j + W_j) H_j + V_{j+1} H_{j+1} + F_j H F_j - Q_j = 0 \quad (j = 1, N) \quad (19)$$

Systems 3 ( $i = 1, M$ )

$$\frac{L_{j-1} + L'_{j-1} K^+_{i,j-1}}{\eta_{j-1} + (1 - \eta_{j-1}) K^+_{i,j-1}} \bar{x}_{i,j-1} - \left( \bar{L}_j + \frac{(V_j + W_j) K_{i,j}}{\eta_j + (1 - \eta_j) K^+_{i,j}} + \lambda \right) \bar{x}_{i,j} +$$

$$\frac{V_{j+1}K_{i,j+1}}{\eta_{j+1} + (1 - \eta_{j+1})K_{i,j+1}^+} \bar{x}_{i,j+1} = -F_j z_{i,j} - \lambda \bar{x}_{i,j} \quad (j = 1, N) \quad (20)$$

where  $\lambda$  is a positive parameter which describes the dynamic behavior of the column (Ball and Beirag, 1961). When  $\lambda = 0$ , Eq. 20 is the steady state material balance of the  $i$ -th component on the  $j$ -th stage. This parameter can be very useful in the below described iterative procedure of solution, because with increasing values of  $\lambda$  the approach to the solution is more stable, although the convergence is slower.

The solution procedure may be summarized as follows:

- 1) A first trial set of values of  $\bar{x}_{i,j}$  is assumed.
- 2) Solution of all the  $N$  Systems 1 to evaluate the unknowns  $y_{i,j}$ ,  $x_{i,j}$ ,  $x'_{i,j}$ ,  $T_j$  and  $\eta_j$ , on each  $j$ -th stage.
- 3) Solution of the linear System 2, with the unknowns  $V_j$ ,  $L_j$ ,  $L'_j$  and  $\bar{L}_j$ .
- 4) Solution of all the  $M$  tridiagonal Systems 3 to evaluate the overall molar fraction  $\bar{x}_{i,j}$  of each  $i$ -th component on all the  $N$  stages.
- 5) The procedure at this point is iterated.

The solution of each of the three systems requires some remarks. Each of Systems 1 contains some non linear equations due to the highly non linear dependence of the equilibrium ratios to the compositions. Therefore, it requires an efficient method of solution of non linear systems. It is important to note that the trivial solution ( $\eta_j = 1$ ;  $x_{i,j} = x'_{i,j} = \bar{x}_{i,j}$ ;  $y_{i,j} = K_{i,j}x_{i,j}$ ;  $T_j =$  boiling point temperature of the overall liquid mixture) must be avoided when the two liquid phases are present. This aim has been pursued in the present program by solving the system with a particular subroutine (Buzzi Ferraris, 1972) which allows restriction of values of the unknowns in a specified range. It is thus possible to avoid that  $x_{i,j} = x'_{i,j}$  by giving two distinct allowable ranges to the compositions of the two liquid phases. It is clear that this approach requires that the phases number on each stage is *a priori* specified. If only two phases are present it can be or  $\eta_j = 0$  and Eqs. 12 and 14 are replaced respectively by:

$$x_{i,j} = 1/M \quad (i = 1, M) \quad (12')$$

$$\eta_j = 0 \quad (14')$$

or  $\eta_j = 1$  and then Eqs. 13 and 15 are replaced respectively by:

$$x'_{i,j} = 1/M \quad (i = 1, M) \quad (13')$$

$$\eta_j = 1 \quad (15')$$

and the procedure reduces to a two-phase boiling-point method.

The linear System 2 has a block tridiagonal form and therefore it can be solved straight away.

Each of the  $M$  Systems 3 consists of  $N$  equations with a tridiagonal structure. These systems have been solved using some formulas which has been derived applying the procedure proposed by Boston and Sullivan (1972) for the two-phase problems.

The value of  $\bar{x}_{i,j}$  on each stage  $j$  is then obtainable by using the following equations, starting from  $j = 1$ :

$$\tau_j = \alpha_{j+1} \quad (21)$$

$$\epsilon_j = \alpha_j \frac{\epsilon_{j-1}}{\delta_{j-1}} + \beta_j \quad (22)$$

$$\delta_j = \epsilon_j + \gamma_j \quad (23)$$

$$\rho_j = \omega_j + \gamma_j \mu_{j-1} \quad (24)$$

$$\nu_j = -\tau_j / \delta_j \quad (25)$$

$$\mu_j = \rho_j / \delta_j \quad (26)$$

$$\bar{x}_{i,j} = \mu_j - \nu_j \bar{x}_{i,j} \quad (\bar{x}_{i,N} = \mu_N) \quad (27)$$

where, in the case under examination is:

$$\alpha_j = \frac{V_j K_{i,j}}{\eta_j + (1 - \eta_j) K_{i,j}^+}$$

$$\beta_j = U_j + U'_j K_{i,j}^+ + W_j K_{i,j} + \lambda$$

$$\gamma_j = L_j + L'_j K_{i,j}^+$$

$$\omega_j = F_j z_{i,j} + \lambda \bar{x}_{i,j}$$

and the initial conditions are  $\epsilon_0 = \delta_0 = 1$  and  $\mu_0 = 0$ .

These solution equations are more stable than the classical Thomas algorithm (Holland, 1975), because the denominators of Eqs. 25 and 26 are given by the sum (Eq. 23) of all positive terms. Therefore, numerical instabilities due to the difference between two numbers of the same order of magnitude are avoided.

The iterative solving procedure outlined in the points 1 to 5 refers to the standard set of unspecified variables. The solution of problems in which a different choice of specified and unspecified variables is done, is not however in general always obtainable with this procedure. In particular, it is impossible to give some specification equations involving unknowns which are evaluated from different systems. For example it is impossible to specify the flowrate of component  $i$  on the  $j$ -th stage,  $L_{x,i,j}$ , because  $L_j$  is evaluated from System 2 while  $x_{i,j}$  is evaluated from System 1. In our program this particular method has been developed in order to solve problems where  $s$  of the following variables of the "standard set of the specified variables"  $U_j$ ,  $U'_j$ ,  $W_j$  and  $F_j$  can be unknown. Furthermore, the corresponding  $s$  equations that have to be added, can be any linear combination of the variables  $L_j$ ,  $L'_j$ ,  $V_j$ ,  $U_j$ ,  $U'_j$ ,  $W_j$  and  $F_j$  on the same or different stages. This requires a modification of the solving procedure of the only System 2. In particular the new unknowns are added on the right hand side of the last column, and the new equations after the last row, of the original coefficient matrix of the linear system. In this way, the coefficient matrix retains its block-tridiagonal structure except for the new columns and rows, and the problem is basically reduced to the solution of a linear system of  $s$  equations as reported in more detail by Buzzi Ferraris (1980).

In conclusion, this solution procedure on the whole presents the same advantages and limits that are well known for the two phases boiling point method (Friday and Smith, 1964). In particular this method shows a limited possibility to solve problems with "non standard" choices of specified and unspecified variables, and an uncertain convergence in cases where the relative volatility of the components in the mixture are largely different. However this method requires a small amount of computation time and may approach the solution also when the starting solution is poor.

## GLOBAL NEWTON-RAPHSON METHOD

This method regards the whole system of Eqs. 1 to 7 as a general non linear system, and does not split it in subsystems of smaller dimension, but solves it altogether by applying the Taylor's approximation truncated after the first term to all the involved equations. The resulting linear system with the unknowns  $\delta y_{i,j}$ ,  $\delta x_{i,j}$ ,  $\delta'_{i,j}$ ,  $\Delta T_j$ ,  $\delta V_j$ ,  $\delta L_j$  and  $\delta L'_j$ , is given by the following equations with  $j = 1, N$ :

$$\begin{aligned} L_{j-1} \delta x_{i,j-1} + L'_{j-1} \delta x'_{i,j-1} - (L_j + U_j) \delta x_{i,j} - (L'_j + U'_j) \delta x'_{i,j} \\ - (V_j + W_j) \delta y_{i,j} + V_{j+1} \delta y_{i,j+1} + x_{i,j-1} \delta L_{j-1} + x'_{i,j-1} \delta L'_{j-1} \\ - x_{i,j} \delta L_j - x'_{i,j} \delta L'_j - y_{i,j} \delta V_j + y_{i,j+1} \delta V_{j+1} = -f_{i,j} \quad (i = 1, M) \end{aligned} \quad (28)$$

$$\begin{aligned} \delta y_{i,j} - K_{i,j} \delta x_{i,j} - x_{i,j} \frac{\partial K_{i,j}}{\partial T_j} \delta T_j - \sum x_{i,j} \frac{\partial K_{i,j}}{\partial x_{n,j}} \delta x_{n,j} \\ - \sum x_{i,j} \frac{\partial K_{i,j}}{\partial y_{n,j}} \delta y_{n,j} = -\phi_{i,j} \quad (i = 1, M) \end{aligned} \quad (29)$$

$$\begin{aligned} \delta y_{i,j} - K'_{i,j} \delta x'_{i,j} - x'_{i,j} \frac{\partial K'_{i,j}}{\partial T_j} \delta T_j - \sum x'_{i,j} \frac{\partial K'_{i,j}}{\partial x'_{n,j}} \delta x'_{n,j} \\ - \sum x'_{i,j} \frac{\partial K'_{i,j}}{\partial y'_{n,j}} \delta y'_{n,j} = -\phi'_{i,j} \quad (i = 1, M) \end{aligned} \quad (30)$$

$$\Sigma(\delta y_{n,j} - \delta x_{n,j}) = -\sigma_j \quad (31)$$

$$\Sigma(\delta y_{n,j} - \delta x'_{n,j}) = -\sigma'_j \quad (32)$$

$$\delta L_{j-1} + \delta L'_{j-1} - \delta L_j - \delta L'_j - \delta V_j + \delta V_{j+1} = -r_j \quad (33)$$

$$\begin{aligned} L_{j-1} \Sigma \frac{\partial h_{j-1}}{\partial x_{n,j-1}} \delta x_{n,j-1} + L'_{j-1} \Sigma \frac{\partial h'_{j-1}}{\partial x'_{n,j-1}} \delta x'_{n,j-1} \\ - (L_j + U_j) \Sigma \frac{\partial h_j}{\partial x_{n,j}} \delta x_{n,j} - (L'_j + U'_j) \Sigma \frac{\partial h'_j}{\partial x'_{n,j}} \delta x'_{n,j} \\ - (V_j + W_j) \Sigma \frac{\partial H_j}{\partial y_{n,j}} \delta y_{n,j} + V_{j+1} \Sigma \frac{\partial H_{j+1}}{\partial y_{n,j+1}} \delta y_{n,j+1} \\ + h_{j-1} \delta L_{j-1} + h'_{j-1} \delta L'_{j-1} - h_j \delta L_j \\ - h'_j \delta L'_j - H_j \delta V_j + H_{j+1} \delta V_{j+1} \\ + \left( L_{j-1} \frac{\partial h_{j-1}}{\partial T_{j-1}} + L'_{j-1} \frac{\partial h'_{j-1}}{\partial T_{j-1}} \right) \delta T_{j-1} + V_{j+1} \frac{\partial H_{j+1}}{\partial T_{j+1}} \delta T_{j+1} \\ - \left( (L_j + U_j) \frac{\partial h_j}{\partial T_j} + (L'_j + U'_j) \frac{\partial h'_j}{\partial T_j} \right. \\ \left. + (V_j + W_j) \frac{\partial H_j}{\partial T_j} \right) \delta T_j = -g_j \end{aligned} \quad (34)$$

These equations describe the  $j$ -th stage where there are two immiscible liquid phases. If, on this stage, there is only one liquid phase  $L_j$  and is  $L'_j = 0$ , then Eqs. 30 and 32 are replaced by:

$$\delta x'_{i,j} = 0 \quad (i = 1, M) \quad (30')$$

$$\delta L'_j = 0 \quad (32')$$

On the other hand, if the other phase  $L'_j$  is present while  $L_j = 0$ , Eqs. 29 and 31 are replaced by:

$$\delta x_{i,j} = 0 \quad (i = 1, M) \quad (29')$$

$$\delta L_j = 0 \quad (31')$$

It clearly appears that also this method requires the *a priori* knowledge of the stages in which the liquid phase separation occurs.

The resulting system of Eqs. 28 to 34 arranged stage by stage, has a block-tridiagonal structure, similar to that obtained for the two-phases multicomponent, multistaged separators, with the method proposed by Naphtali and Sandholm (1971). In the present case each submatrix has the dimension of  $3M + 4$  rows and columns.

As the vapor phase may often prove close to ideal, when practically applied we may neglect the terms  $\partial K_{i,j}/\partial y_{n,j}$  and  $\partial K'_{i,j}/\partial y_{n,j}$  in Eqs. 28 to 34. In this case, Eq. 29 can be analytically solved with respect to  $\delta y_{i,j}$  and then removed from the linearized system. The dimension of the submatrices is so reduced to  $2M + 4$ .

To improve the efficiency of the solution method, it is important to evaluate the derivatives of enthalpy and equilibrium ratios functions in the most efficient way. For the enthalpy functions this may be obtained by expressing the enthalpy of the liquid and vapor phase using the molar enthalpy of each component

$$h = \Sigma \tilde{h}_n x_n + h_{\text{mix}} \quad (35)$$

$$H = \Sigma \tilde{H}_n y_n \quad (36)$$

so that the partial derivatives of  $h$  and  $H$  comparing in Eq. 34 with respect to the composition can be estimated as:

$$\frac{\partial h}{\partial x_n} = \frac{\partial h'}{\partial x'_n} = \tilde{h}_n \quad (37)$$

$$\frac{\partial H}{\partial y_n} = \tilde{H}_n \quad (38)$$

neglecting, only for the estimation of these derivatives, the dependence of the mixing enthalpy on the composition.

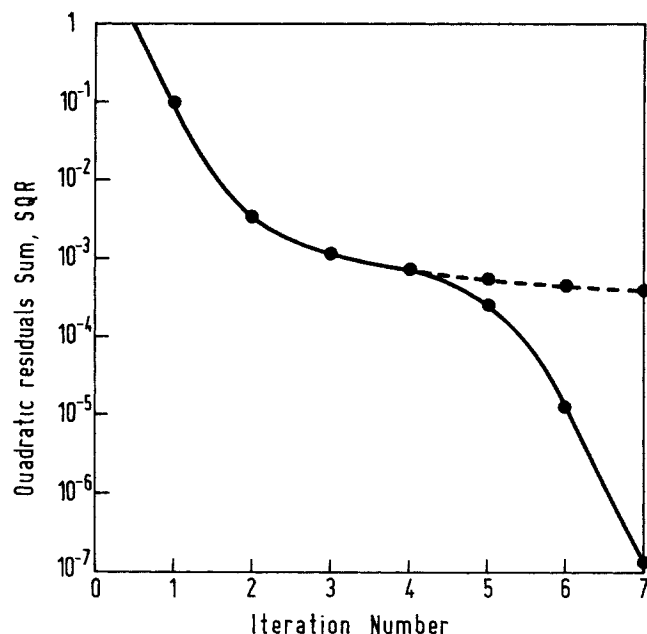


Figure 2. Influence of the derivatives of the equilibrium ratios with respect to the liquid compositions on the convergence speed. Dotted line: the derivatives are always neglected. Solid line: the derivatives are evaluated on all the stages from iteration 4 on.

Due to the high non ideality of liquid mixtures where the phase separation can occur, the derivatives of equilibrium ratios with respect to the compositions and to the temperature can not be neglected. Evaluation of these derivatives can be performed either analytically or, more often, numerically; in both cases they may require a large amount of computation time. Therefore, it is convenient to evaluate it only on few stages, using a linear interpolation to estimate it on the intermediate stages, where the number of the present equilibrium phases is unchanged. The effect of the presence of these derivatives in Eqs. 29 and 30 on the convergence speed of the algorithm is reported in Figure 2.

It can be observed that, in the reported case, increasing the number of stages in which such derivatives are evaluated, the number of iterations required to reach the solution decreases. In some other cases if the number of such stages is too low, the convergence to the solution may also not be achieved. However, computation time required from each iteration increases by increasing the number of stages in which such derivatives are evaluated. Therefore, it is convenient to start with few of these stages, and then increase it if the solution method slows its convergence speed or if it diverges.

As it has been already pointed out the Newton-Raphson method faces the system of Eqs. 1 to 7 as a whole. At each iteration, the corrections of the previous set of solution variables is obtained by solving a linear system with a block-tridiagonal structure. This allows solution of problems where the set of specified and unspecified variables is different from the "standard set." In particular, we can assume as unknown any one of the variables  $U_j$ ,  $U'_j$ ,  $W_j$ ,  $F_j$ ,  $z_{i,j}$ ,  $Q_j$  and at the same time introduce any equation relating all the variables of the problem. These specification equations are linearized, as all other equations of the system, and their coefficients are added after the last row of the original coefficient matrix. The new corresponding unknowns are instead added after the last column. The resulting coefficient matrix has then still a block-tridiagonal structure except for the new columns and rows. The solution of such a system, in the same way as the boiling point type method, can be obtained as reported by Buzzi Ferraris (1980).

In conclusion, this method allows to solve a wide variety of problems, and it requires the *a priori* knowledge of the stages in which the liquid phase separation occurs.

It is worth, however, remembering that the Newton-Raphson approach is a very efficient procedure if the initial guess is "close enough" to the final answer. Therefore the method must be integrated with other algorithms if we look for a general program for multicomponent, multistaged separators.

## MULTIFLASH METHOD

Let us consider the  $j$ -th stage of a distillation column rearranged as shown in Figure 3. The liquid streams coming from the stage  $j - 1$ :  $L_{j-1}$  and  $L'_{j-1}$ , the vapor stream coming from the stage  $j + 1$ :  $V_{j+1}$ , as well as the feed stream  $F_j$  are mixed all together, in order to build up a single feedstream to the  $j$ -th stage. Composition, flowrate and enthalpy of this stream can be easily evaluated when the variables  $L_{j-1}$ ,  $L'_{j-1}$ ,  $h_{j-1}$ ,  $h'_{j-1}$ ,  $x_{i,j-1}$ ,  $x'_{i,j-1}$ ,  $y_{i,j+1}$ ,  $V_{j+1}$ ,  $H_{j+1}$ ,  $F_j$ ,  $z_{i,j}$  and  $HF_j$  are assigned.

The system of Eqs. 1 to 7 can then be solved with respect to the unknowns  $T_j$ ,  $L_j$ ,  $L'_j$ ,  $V_j$ ,  $x_{i,j}$ ,  $x'_{i,j}$ ,  $y_{i,j}$ . It is important to point out that this system can be regarded as a three-phase flash model of the previously obtained stream, in which the values of  $Q_j$ ,  $P_j$ ,  $U_j$  and  $W_j$  are fixed.

By applying this procedure to all the stages of the column, it is possible to establish an input-output sequence in order to build up an iterative solving procedure. Namely the solution of the three-phase flash problem for each stage, allows us to obtain the new trial values of the variables  $T_j$ ,  $L_j$ ,  $L'_j$ ,  $V_j$ ,  $x_{i,j}$ ,  $x'_{i,j}$ ,  $y_{i,j}$  ( $i = 1, M$ ) starting from trial values of the corresponding variables in the adjacent stages  $j - 1$  and  $j + 1$ . By means of this procedure the multistage problem reduces to a set of three-phase flash problems.

In Figures 4 a, b, c, some possible sequences for the simple case of a five stage column are shown. Figure 4a represents a sequence in which each stage is considered as separated from the others; at each iteration, the values of all the variables are simultaneously changed. In Figure 4b, a top-bottom sequence in which each stage transmits informations to the next one and finally to the bottom stage is shown. In Figure 4c, a chessboard type sequence with transmission among even and odd stages is represented.

Each stage of a distillation column operating in a steady state condition is then simulated with a discontinuous and orderly series of elementary acts. The approach to the solution obtained by means of this procedure is then similar, even if there is no explicit time variable, to the situation of the start up and stabilization of a distillation column.

This method, although extremely stable in approaching the solution, usually requires many iterations, and therefore a large amount of computation time. Furthermore, we may come across the problem of avoiding the trivial two-phase solution on those stages where a liquid phase separation occurs. This difficulty can be overcome, as in the previous methods, by constraining the unspecified variables  $x_{i,j}$  and  $x'_{i,j}$  so that the trivial solution  $x_{i,j} = x'_{i,j}$  is avoided.

It is worthwhile to note that this method proves effective to verify the solution obtained by the above reported methods. In particular, by using the sequence reported in Figure 4a it is possible to consider each stage separately in order to verify that, in those stages where only two phases are considered, the liquid phase separation does not actually occur.

## COMPUTER PROGRAM

Each of the above reported methods shows some peculiar characteristics and different efficiency in approaching the solution. It may prove convenient to combine all of them to build up a computer program able to reach the solution in the most efficient way. In order to control the convergence of the program, it is necessary to define an objective function SQR as the square root of the sum of weighted squared residuals of all equations in the system. The weights for each equation are defined so that a SQR value of, say,  $10^{-6}$  means that the average error in the residuals of all the equations involves the sixth significant digit. The SQR function is evaluated at each iteration

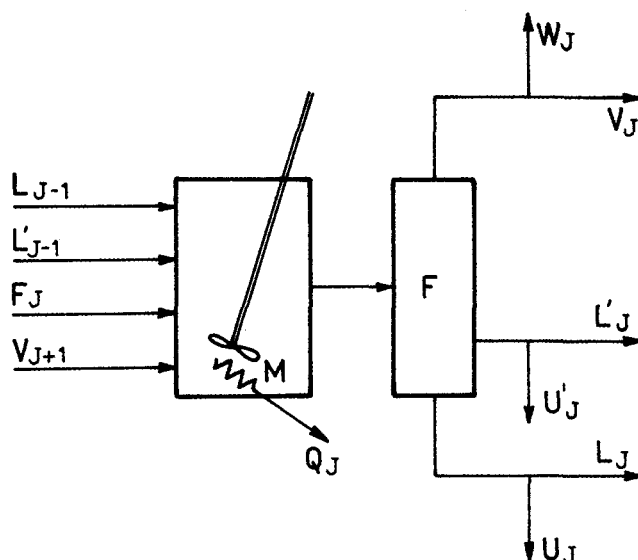


Figure 3. The model of the column stage in the multiflash method.

and the new trial solution is accepted only when the SQR value decreases.

The global Newton-Raphson method is assumed to be the core of the computer program here developed, because of its effectiveness to reach the solution. When the new trial solution proposed by this method increases the objective function SQR, a one dimensional research is performed. This procedure consists of a SQR minimum research along the direction predicted by the Newton global method. If the newly obtained solution is considerably different from the previous one (1% for temperature and flow rates and 0.01 for mole fractions), a new iteration of the Newton global method is performed, otherwise the control switches to the boiling point type method or, if possible, it simply increases the number of stages in which the derivatives  $\partial K_{i,j}/\partial x_{n,j}$ ,  $\partial K'_{i,j}/\partial x'_{n,j}$ ,  $\partial K_{i,j}/\partial T_j$ , and  $\partial K'_{i,j}/\partial T_j$  are estimated, allowing the same method to retain control.

The boiling point type method proves very useful when the Newton-Raphson global method fails. In fact it has a completely different way to approach the solution and it requires a small amount of computation time.

Due to the high nonideality of the liquid mixture under examination, it is not possible to evaluate automatically a first trial solution using the ideal equilibrium ratios. Therefore, it is necessary to present it as an input. Naturally to obtain a quick convergence of the program it is necessary to give a reasonable first trial solution. When the available starting solution is poor

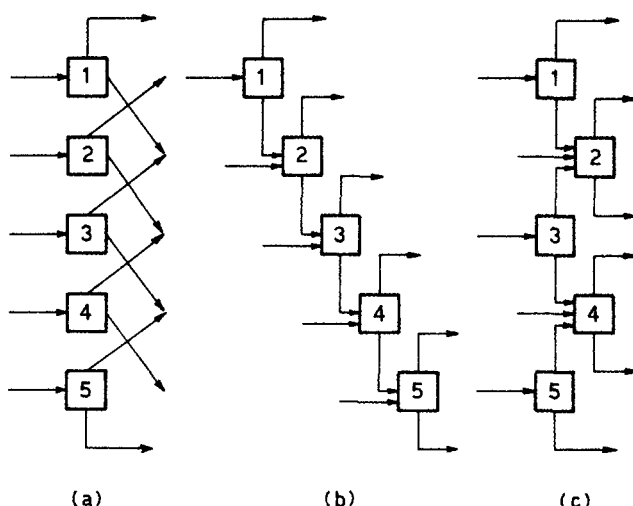


Figure 4. Possible sequences of flash to represent a five stages distillation column. a—each stage is separated from the others; b—top-bottom sequence; c—chessboard-type sequence.

the Newton-Raphson method often fails, while the BP type method may be useful to improve the trial solution.

A boiling point type method has been proposed by Block and Hegner (1976) using a Newton-Raphson procedure to solve the above mentioned system 3 in the unknowns  $\bar{x}_{i,j}$ . This procedure involves the numerical evaluation of the derivatives  $\partial y_{i,j}/\partial \bar{x}_{n,k}$ ,  $\partial x_{i,j}/\partial \bar{x}_{n,k}$  and  $\partial x'_{i,j}/\partial \bar{x}_{n,k}$  for all the components and on each stage. Each of such evaluation requires the solution of a non linear system of equations. Therefore, this procedure is too much time consuming to be applied in the computer program here developed, where the boiling point type method is used only as an alternative to the more powerful global Newton method.

Finally the multiflash method is used to verify the obtained solution and to define the trial solution for the new solving procedure when the assumed phase type of the column is not correct, as it will be shown shortly.

As already pointed out, the number of equilibrium phases present on each stage along the column has to be assigned as an input. Actually this is *necessary*, in order to obtain a problem with a *unique* solution. In fact, as pointed out by Henley and Rosen (1969), a system in which three phases are present always admits a two-phase mathematical solution at the same time. Therefore, the correct solution is such as to show three phases in some or all stages; yet along with this you find solutions where only two phases are considered in some of these stages.

Therefore, it is necessary to assess a first trial number of phases on each stage, solve the problem, and then verify the solution with the multiflash method. If the multiflash method predicts a liquid phase separation in some of the stages, where only two phases were assumed, then the whole solving procedure has to be repeated. The starting solution assumed for the new solving procedure is the one obtained by the multiflash method, which appeared to be very close to the correct one. This procedure is iterated until the multiflash method predicts the same number of phases on each stage assumed to obtain the solution.

In conclusion, if the given number of stages in which three phases are present is lower than the correct one, the program reaches a mathematical solution of the system but the multiflash method will predict a liquid phase separation also in some other stage; whereas if greater, the program will not find any solution, and the problem has to be repeated with fewer such stages.

## APPLICATIONS OF COMPUTER PROGRAM

The aim of this section is to show some peculiarities of the developed program mentioned above. The examined example is a 12-stage distillation column in which the separation of the mixture propanol-butanol-water is performed. This problem is similar to that reported by Block and Hegner (1976) and the physicochemical data of the system are resumed in Table 1.

For all the examined problems the following starting solution has been used:

TABLE 1. PHYSICO-CHEMICAL DATA FOR THE SYSTEM: PROPANOL (1), BUTANOL (2), AND WATER (3)

Component	1	2	3
$C_p^L$	12.28	50.92	18.00
$\Delta H_{tr}(25^\circ\text{C})$	11,240	12,280	10,681
A	8.37895	7.83800	8.07131
B	1,788.02	1,558.19	1,730.63
C	-45.722	-76.279	-39.734
$a_0$	0.59	0.78	8.22
$a_1$	0.07942	0.0998	0.0015
$a_2$	-4.431-05	-5.35-05	0.134-05

$$\log p^\circ = A - B/(T + C)$$

$$C_p^V = a_0 + a_1T + a_2T^2$$

NRTL Parameters (Block and Hegner, 1976)

	$\tau_{i,j}$	$\tau_{j,i}$	$\alpha_{i,j}$
1-2	-0.61259	0.71640	0.30
2-3	0.90047	3.51307	0.48
3-1	2.7425	-0.07149	0.30

$$T_j = 360.0; V_1 = 0.0; V_j = 100.0; L_j = 100.0; (j = 1, N)$$

$$x_{1,1} = 0.3; x_{2,1} = 0.1; x_{3,1} = 0.6; y_{1,1} = 0.3; y_{2,1} = 0.1; y_{3,1} = 0.6; x_{1,12} = 0.1; x_{2,12} = 0.3; x_{3,12} = 0.6; y_{1,12} = 0.1; y_{2,12} = 0.3; y_{3,12} = 0.6$$

the mole fractions  $x_{i,j}$  and  $y_{i,j}$  in the intermediate stages have been evaluated by linear interpolation between their values in the first and last stage above reported. Furthermore, in those stages in which liquid separation was assumed, the following relationships were also imposed

$$x'_{1,j} = x'_{2,j} = 0.0; x'_{3,j} = 1.0; L'_j = 10.$$

The first peculiarity taken into account is the possibility to use a set of specified and unspecified variables different from the "standard set." For example it can be useful to assign the mole fraction of a particular component, or its percentage with respect to the feed, in the distillate, as an input.

In the examined example, the following two specification equations are considered:

$$1) \text{ Specified: } x_{1,1} = 0.35; L_1 = 87.; V_1 = 0.$$

$$\text{Unspecified: } Q_1; Q_{12}; U_1$$

$$2) \text{ Specified: } \frac{L_{12}x_{2,12}}{F_{52.5}} = 0.85; V_1 = 0.$$

$$\text{Unspecified: } Q_1; U_1$$

Further, in the second problem, the reflux stream,  $L_1$ , is cooled of  $30^\circ\text{C}$  under the equilibrium temperature,  $T_1$ , of the condenser and the reboiler duty is set,  $Q_{12} = 1.25 \cdot 10^6$ .

It is worthwhile to point out that these problems, due to the particular specification equations introduced, are solvable only

TABLE 2. NUMERICAL SOLUTION OF PROBLEM 1

j	$x_{1,j}$	$x_{2,j}$	$x_{3,j}$	$x'_{1,j}$	$x'_{2,j}$	$x'_{3,j}$	$y_{1,j}$	$y_{2,j}$	$y_{3,j}$	$T_j$	$V_j$	$L_j$	$L'_j$
1	0.3500	0.0279	0.6221	0.0000	0.0000	0.0000	0.3807	0.0133	0.6061	361.8	0.0000	87.00	0.0000
2	0.2997	0.0528	0.6475	0.0000	0.0000	0.0000	0.3500	0.0279	0.6221	362.2	115.6	87.56	0.0000
3	0.2493	0.0786	0.6721	0.0000	0.0000	0.0000	0.3121	0.0466	0.6413	362.8	116.1	88.15	0.0000
4	0.2086	0.1014	0.6900	0.0000	0.0000	0.0000	0.2740	0.0662	0.6598	363.3	116.7	88.64	0.0000
5	0.1871	0.1251	0.6878	0.0000	0.0000	0.0000	0.2431	0.0834	0.6735	363.8	117.2	139.5	0.0000
6	0.1597	0.1399	0.7004	0.0000	0.0000	0.0000	0.2126	0.0995	0.6879	364.1	118.1	140.1	0.0000
7	0.1334	0.1550	0.7116	0.0000	0.0000	0.0000	0.1802	0.1170	0.7028	364.5	118.7	140.6	0.0000
8	0.1130	0.1766	0.7104	0.0306	0.0257	0.9438	0.1490	0.1351	0.7160	364.8	119.2	135.0	6.187
9	0.0966	0.2057	0.6977	0.0227	0.0242	0.9531	0.1206	0.1528	0.7266	365.2	119.7	124.6	17.02
10	0.0800	0.2337	0.6863	0.0167	0.0231	0.9602	0.0950	0.1693	0.7357	365.4	120.2	116.8	25.21
11	0.0641	0.2597	0.6761	0.0121	0.0223	0.9657	0.0727	0.1839	0.7434	365.6	120.6	110.9	31.52
12	0.0493	0.2835	0.6672	0.0085	0.0216	0.9699	0.0537	0.1967	0.7496	365.8	121.0	20.01	1.414

$$U_1 = 28.575 \quad Q_1 = 1.2414 + 06 \quad Q_{12} = -1.2455 + 06 \quad P = 1.$$

$$F_5 = 50.0 \quad z_{1,5} = 0.22 \quad z_{2,5} = 0.13 \quad z_{3,5} = 0.65 \quad HF_5 = 65,000.0$$

TABLE 3. NUMERICAL SOLUTION OF PROBLEM 2

$j$	$x_{1,j}$	$x_{2,j}$	$x_{3,j}$	$x'_{1,j}$	$x'_{2,j}$	$x'_{3,j}$	$y_{1,j}$	$y_{2,j}$	$y_{3,j}$	$T_j$	$V_j$	$L_j$	$L'_j$
1	0.3416	0.0320	0.6264	0.0000	0.0000	0.0000	0.3760	0.0155	0.6086	361.8	0.0000	82.55	0.0000
2	0.2877	0.0589	0.6535	0.0000	0.0000	0.0000	0.3416	0.0320	0.6264	362.4	112.2	87.10	0.0000
3	0.2368	0.0851	0.6781	0.0000	0.0000	0.0000	0.3014	0.0521	0.6466	362.9	116.7	87.69	0.0000
4	0.1981	0.1070	0.6949	0.0000	0.0000	0.0000	0.2633	0.0717	0.6650	363.5	117.3	88.17	0.0000
5	0.1789	0.1296	0.6915	0.0000	0.0000	0.0000	0.2342	0.0881	0.6777	363.9	117.8	139.1	0.0000
6	0.1510	0.1448	0.7042	0.0000	0.0000	0.0000	0.2022	0.1051	0.6927	364.3	118.7	139.7	0.0000
7	0.1252	0.1602	0.7146	0.0000	0.0000	0.0000	0.1694	0.1230	0.7076	364.6	119.3	140.2	0.0000
8	0.1075	0.1865	0.7060	0.0276	0.0250	0.9474	0.1392	0.1411	0.7197	365.0	119.8	130.7	10.03
9	0.0911	0.2152	0.6938	0.0205	0.0238	0.9556	0.1118	0.1584	0.7298	365.3	120.3	121.3	19.81
10	0.0749	0.2422	0.6829	0.0151	0.0228	0.9621	0.0876	0.1741	0.7383	365.5	120.8	114.3	27.19
11	0.0596	0.2670	0.6734	0.0109	0.0220	0.9670	0.0668	0.1879	0.7454	365.7	121.1	109.0	32.89
12	0.0456	0.2893	0.6651	0.0077	0.0214	0.9708	0.0492	0.1998	0.7511	365.8	121.5	19.10	1.281

$$U_1 = 29.618 \quad Q_1 = 1.2611 + 06 \quad Q_{12} = -1.2500 + 06 \quad P = 1.$$

$$F_5 = 50.0 \quad z_{1,5} = 0.22 \quad z_{2,5} = 0.13 \quad z_{3,5} = 0.65 \quad HF_5 = 65,000.0$$

TABLE 4. PSEUDOSOLUTION OF PROBLEM 1 (THREE PHASES ARE PRESENT ONLY ON THE LAST TWO STAGES)

$j$	$x_{1,j}$	$x_{2,j}$	$x_{3,j}$	$x'_{1,j}$	$x'_{2,j}$	$x'_{3,j}$	$y_{1,j}$	$y_{2,j}$	$y_{3,j}$	$T_j$	$V_j$	$L_j$	$L'_j$
1	0.3500	0.0279	0.6221	0.0000	0.0000	0.0000	0.3807	0.0133	0.6061	361.8	0.0000	87.00	0.0000
2	0.2997	0.0528	0.6475	0.0000	0.0000	0.0000	0.3500	0.0279	0.6221	362.2	115.5	87.56	0.0000
3	0.2493	0.0786	0.6721	0.0000	0.0000	0.0000	0.3121	0.0467	0.6413	362.8	116.0	88.15	0.0000
4	0.2085	0.1014	0.6901	0.0000	0.0000	0.0000	0.2739	0.0662	0.6599	363.3	116.6	88.64	0.0000
5	0.1870	0.1252	0.6878	0.0000	0.0000	0.0000	0.2429	0.0835	0.6735	363.8	117.1	139.5	0.0000
6	0.1594	0.1400	0.7006	0.0000	0.0000	0.0000	0.2123	0.0997	0.6880	364.1	118.0	140.1	0.0000
7	0.1326	0.1546	0.7128	0.0000	0.0000	0.0000	0.1796	0.1173	0.7031	364.5	118.6	140.7	0.0000
8	0.1084	0.1682	0.7235	0.0000	0.0000	0.0000	0.1479	0.1347	0.7174	364.9	119.1	141.2	0.0000
9	0.0875	0.1803	0.7322	0.0000	0.0000	0.0000	0.1192	0.1508	0.7301	365.2	119.7	141.7	0.0000
10	0.0710	0.1949	0.7341	0.0000	0.0000	0.0000	0.0946	0.1651	0.7403	365.4	120.1	142.0	0.0000
11	0.0659	0.2569	0.6772	0.0125	0.0223	0.9651	0.0751	0.1823	0.7426	365.6	120.4	111.5	30.86
12	0.0507	0.2812	0.6681	0.0088	0.0217	0.9695	0.0554	0.1955	0.7491	365.8	120.8	20.19	1.344

$$U_1 = 28.469 \quad Q_1 = 1.2403 + 06 \quad Q_{12} = -1.2444 + 06 \quad P = 1.0$$

$$F_5 = 50.0 \quad z_{1,5} = 0.22 \quad z_{2,5} = 0.13 \quad z_{3,5} = 0.65 \quad HF_5 = 65,000.0$$

TABLE 5. NUMERICAL SOLUTION OF THE MULTIFLASH METHOD APPLIED TO THE RESULTS REPORTED IN TABLE 4

$j$	$x_{1,j}$	$x_{2,j}$	$x_{3,j}$	$x'_{1,j}$	$x'_{2,j}$	$x'_{3,j}$	$y_{1,j}$	$y_{2,j}$	$y_{3,j}$	$T_j$	$V_j$	$L_j$	$L'_j$
1													
2													
3													
4													
5													
6													
7													
8	0.1128	0.1770	0.7103	0.0304	0.0256	0.9440	0.1487	0.1353	0.7161	364.9	119.1	132.5	8.667
9	0.0969	0.2054	0.6977	0.0228	0.0243	0.9529	0.1210	0.1525	0.7264	365.2	119.6	120.9	20.86
10	0.0812	0.2316	0.6872	0.0171	0.0232	0.9597	0.0968	0.1681	0.7351	365.4	120.0	115.3	26.79
11	0.0659	0.2569	0.6772	0.0125	0.0223	0.9651	0.0751	0.1823	0.7426	365.6	120.4	111.5	30.86
12	0.0507	0.2812	0.6681	0.0088	0.0217	0.9695	0.0554	0.1955	0.7491	365.8	120.8	20.19	1.344

NO THREE-PHASE SOLUTION

through the global Newton-Raphson method. The obtained full solutions are reported in Tables 2 and 3. The second point that we want to emphasize is the research of the stages in which the liquid-phase separation occurs.

Table 4 reports the solution of problem 1), where the three phases presence is considered only on the last two stages. You may notice that such solution exists, together with the solution with liquid phase separation on the last five stages (Table 2). Therefore, the number of phases present on each stage can not remain unspecified during the solution of the problem, since multiple solutions of such a problem are workable. It is, therefore, necessary to solve the problem with a first trial phase type of column, and then verify that on each stage the assigned number of phases is correct. This procedure is followed, as seen above, by the multiflash method.

In Table 5, it can be observed that the multiflash method, applied to the results reported in Table 4 (with presence of the second liquid only on the last two stages), predicts that the liquid

phase separation occurs also on the stages 8, 9 and 10. Therefore the problem has been solved assuming three coexisting phases on the last five stages and the obtained results are reported in Table 2. It appears from the comparison of Tables 2 and 5 that the predicted compositions and flow rates on stages 8, 9 and 10 were very close to the correct values.

#### NOTATION

$F$	= feed stream molar flow rate
$h$	= liquid stream molar enthalpy
$\bar{h}$	= molar enthalpy of a liquid component
$\bar{H}$	= vapor stream molar enthalpy
$\bar{H}$	= molar enthalpy of a vapor component
$HF$	= molar feed enthalpy
$h_{\text{mix}}$	= molar mixing enthalpy
$K$	= vapor-liquid equilibrium ratio
$K^+$	= liquid-liquid equilibrium ratio
$L$	= liquid stream molar flow rate

$\bar{L}$  = overall liquid stream molar flow rate defined by Eq. 8  
 $M$  = number of components of the mixture  
 $N$  = number of stages, including reboiler and condenser  
 $P$  = pressure  
 $Q$  = rate of heat leaving the stage  
 $s$  = number of specification equations  
 $T$  = temperature  
 $U$  = liquid side stream molar flow rate  
 $V$  = vapor stream molar flow rate  
 $W$  = vapor side stream molar flow rate  
 $x$  = liquid mole fraction  
 $\bar{x}$  = overall liquid mole fraction defined by Eq. 10  
 $y$  = vapor mole fraction  
 $z$  = feed mole fraction

#### Greek Letters

$\eta$  = phase splitting parameter defined by Eq. 9  
 $\lambda$  = parameter in Eq. 20

#### Superscript

' = second liquid phase

#### Subscripts

$i$  = component index  
 $j$  = stage index

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# Collection Efficiency of Cyclone Separators

Based on terLinden's (A. J. terLinden, "Investigations into Cyclone Dust Collectors," IME Proc., 160, 233, 1949) experimental observations, a three-region model is proposed for the fluid flow in a cyclone. Within each region, turbulence is assumed to promote mixing of the suspended particles. Incorporation of this mixing concept into the three-region model allows an analytic expression for the collection efficiency of the cyclone to be developed.

The theoretical result is compared with data obtained in the high temperature, high pressure exhaust from a pressurized fluidized bed combustor.

P. W. DIETZ

General Electric Company  
 Corporate Research and Development  
 Schenectady, NY 12301

## SCOPE

In the present paper, an analytic expression is developed for the efficiency of reverse flow cyclone separators. To develop this equation, a simple model for the gas flow within the cyclone is proposed. Based on this model, the cyclone is separated into three regions: an inlet region, a downflow region, and an upflow region. Within each of the regions, radial particle-transport is assumed to be dominated by turbulent mixing.

Consequently, a simple equation for conservation of particles can be stated and the expression for cyclone efficiency results from the solution of the equation subject to appropriate boundary conditions.

Comparisons of the resultant expression with data from the second stage cyclone at Exxon's PFB miniplant yield satisfactory agreement.

## CONCLUSIONS AND SIGNIFICANCE

With the growing concern for the environmental effects of particulate pollution, it becomes increasingly important to be able to design optimized pollution control devices. For instance, although measurements of the fractional efficiency of

cyclone separators can be used to predict the performance of an existing design in new applications, the efficiency of new designs cannot be predicted based on current theories. In the present paper, a new, analytic model is developed for the performance of cyclone separators which may provide the needed design tool.

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