

# New Simultaneous Modular Method for Calculating Multistage Multicomponent Separation Processes

A new flexible method has been developed for modelling multistage multicomponent separation processes. The method is based on the simultaneous modular approach and can be referred to as a quasilinear two-level modelling. The network is solved within each iteration as a linear one, and after each network solution the split streams are updated modularly using nonlinear submodels. The method is suited for modelling the most important separation processes with arbitrary structure and permits to use different models for the subunits or processing units. Illustrative examples and computational results are presented showing the efficiency, flexibility and capability of the method.

**Z. FONYO**

Department of Chemical Engineering  
Technical University of Budapest  
1521 Budapest, Hungary

**H. NISHIMURA and  
Y. YAMASHITA**

Department of Chemical Engineering  
University of Tokyo  
Tokyo 113, Japan

## SCOPE

The computation of multistage multicomponent separation processes is a basic chemical engineering problem of considerable practical importance both in preliminary design and in the study and improvement of existing plants. Four types of model equations are to be solved simultaneously: equilibrium relationships, material balance for each component, enthalpy balances, and the restrictions for summation of mole fractions. Since the equations describing material and heat balances are strongly nonlinear, the calculation of compositions is customarily carried out by some suitably chosen iterative procedure. The model equations for the solution method can be grouped by type or by stages. The former is called equation-solving approach, and the latter modular approach. Both solution procedures can be performed either in sequential or in simultaneous

manner. Although the equation-solving methods and the sequential modular approaches are commonly used in the steady-state material and heat balancing, no attempt on the simultaneous modular approach has been made for modelling the separation processes.

The main objective of this study was to demonstrate that the simultaneous modular approach can be successfully applied for calculating any kind of multistage, multicomponent separation processes with arbitrary structures. The method we present incorporates the assignment procedure for general network solution developed by Nishimura and Yamashita (1978), which precedes the rigorous material and heat balancing, thereby leading to considerable savings in computational effort.

## CONCLUSIONS AND SIGNIFICANCE

The simulation of the multistage multicomponent separation processes is a subject which has, in the last two decades, reached various literature. Despite the dramatic progress in our understanding which this has produced, a comprehensive simultaneous modular method capable of modelling the various multistage, multicomponent separation processes has remained out of reach. In this work, a new, flexible method has been developed and proposed to find a rigorous solution of any separation problem. The proposed simultaneous modular method is referred to as quasilinear two-level modelling because the network is solved within each iteration as a linear one, and two types of models are used for each unit. The network solution using linear submodels creates the upper level, and the nonlinear submodels create the lower level of modelling. The illustrative examples

proved the stable application of the method to distillation with narrow and wide boiling mixtures as well as with nonideal mixtures, especially to absorption, extraction and interlinked distillation columns.

The principal significance of the proposed modular method is that it is competitive and quite flexible; besides, it can be used with different types of models for subunits; e.g., the concept of equilibrium stage can be combined with short-cut methods. Another potentially useful feature of the method is that its technique is highly system-engineering-oriented, since the adaptation of the simultaneous modular strategy renders to use the same network solution package possible at different hierarchical levels for the computation of processing units, chemical plants, chemical complexes, etc.

## PROBLEM STATEMENT

The most important separation processes (distillation, absorption, stripping, extraction and washing) can all be modelled by use of the concept of equilibrium stage (also called a theoretical plate or

perfect plate). The concept of the equilibrium stage is based on the presumption that the phases leaving the stage are in thermodynamic equilibrium. The equilibrium stage processes are taken to be at steady-state operation, and the term model as used herein means the complete set of assumptions and corresponding equa-

tions required to describe a process. The solution of the model for separation processes is obtained by finding a set of unknown variables which satisfies all the equations of the model after fixing the parameters according to the degrees of freedom.

The models equations for the stage  $n$  can be expressed as follows:

- a. The equilibrium relationships ( $N \times C$  equations)

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

- b. The component material balances ( $N \times C$  equations)

$$(L_n + P_{Ln})x_{in} + (V_n + P_{Vn})y_{in} - L_{n-1}y_{in-1} - V_{n+1}y_{in+1} - F_n x_{iFn} = 0 \quad (2)$$

- c. The enthalpy balance ( $N$  equations)

$$(L_n + P_{Ln})h_n + (V_n + P_{Vn})H_n - L_{n-1}h_{n-1} - V_{n+1}H_{n+1} - q_n = 0 \quad (3)$$

- d. The restrictions of mole fractions ( $2N$  equations)

$$\sum_i x_{in} = 1$$

$$\sum_i y_{in} = 1 \quad (4)$$

The  $K_{in}$ ,  $h_n$  and  $H_n$  are functions of temperature and the composition; thus, the model of separation problems is nonlinear and contains  $NC + NC + N + 2N = N(2C + 3)$  equations with

$y_{i,n}$	...	$NC$
$x_{i,n}$	...	$NC$
$L_n$	...	$N$
$V_n$	...	$N$
$T_n$	implicite	$N$

altogether  $N(2C + 3)$  unknown variables.

The formulation of the solution method involves the decision of grouping the model equations. Either each individual type of restriction can be satisfied for all stages (one type of restriction for all stages at a time) or all the four types of restriction can be satisfied for each stage (all restrictions for one stage at a time). The former when the equations are grouped by type is called *equation-solving approach*, the latter when the equations are grouped by stages is called *modular approach*.

At the equation-solving approach (Friday and Smith, 1964; King, 1971), the rates of products and the number of stages are specified usually, and the composition of the products are computed, i.e., simulation of a defined separation process is executed. At the common modular approach (Lewis and Matheson, 1932; Greenstadt et al., 1958), the mole fractions of key components in the products are specified, and the rates of products, the number of stages as well as the distribution of nonkey components are calculated, i.e., design of separation column is performed. The latter problem specification is very disadvantageous from numerical point of view. The structure of the system is not determined beforehand; thus, simultaneous approach cannot be used. Consequently, the Lewis-Matheson type of stage to stage calculations are actually *sequential modular methods* which work from both ends of the column toward the middle. The top-down and bottom-up calculations for each component must mesh at a feed stage of the column. All sequential stage to stage solutions suffer from several disadvantages:

- The solution method is essentially restricted to single-feed distillation problem; if more than one-feed stage exist, a choice of mesh point must be chosen for each component.
- The method requires the initial assumption of end compositions hence the procedure is unstable and very susceptible to buildup of truncation errors since stage compositions must be generated from assumed end compositions.
- Arbitrary procedures should be set up to handle nondistributed components since these concentrations are very sensitive to the initialization.

- Rate of convergence of the stage approach is usually slow compared to the equation-solving approaches.

Because of these disadvantages of the sequential modular methods and since the equation-solving type of methods can be applied not only for simulation but also for design purpose indirectly, almost all existing commercial flowsheeting system have adapted or preferred the equation-solving approach for calculating multistage, multicomponent separation processes. These procedures don't require mesh points, work equally well for any number of feeds and side streams and no arbitrary methods require for handling nondistributed components.

Under certain conditions, however, the modular approach is more desirable than equation-solving in the flowsheeting. The fulfilment of this desire is especially important in case of the necessity of using different type of models for the subunits like in the petroleum industry. The case of modelling distillation in petroleum industry, e.g., the concept of equilibrium stage, sometimes should be combined with short-cut methods like Fenske, Underwood, Gilliland type of distillation model.

Although the equation-solving methods and the sequential modular approaches with accelerated convergence methods for solution are commonly used in the steady-state process simulation, no one attempt on the simultaneous modular approach has been made for modelling the separation processes.

The purpose of this paper is to show that the *simultaneous modular approach* can be used for rigorous calculation of multistage, multicomponent separation processes. The approach developed in this work makes use of simultaneous solution of the whole system as a quasilinear network (assuming constant multipliers between inputs and outputs), and after each network solution all multipliers are updated separately using nonlinear submodels of the concept of equilibrium stage. The calculation method to be proposed in this paper offers a means of solution to a wide variety of separation problems such as complex separation towers as well as interconnected set of separation columns. This paper demonstrates a successful application of the simultaneous modular method to different separation problems.

## COMPUTATION BY SIMULTANEOUS MODULAR TECHNIQUE

Originally linear problems were treated by simultaneous modular technique having constant relationships between the inputs and outputs of the subunits. In this case between input vector ( $X$ ) and output vector ( $Y$ ) of the subunit linear relations exist with constant  $P$ -matrices (Mason, 1953, 1956)

$$Y = PX \quad (5)$$

The linear networks could be solved very straightforwardly by using graph representation.

In the case of nonconstant relationships between inputs and outputs of subunits, the  $P$ -matrix is a function of inputs. The form of the  $P$ -matrix in a very general case is (Nishimura et al., 1968)

$$P = \text{diag}(\xi) * (\zeta * S + I) \quad (6)$$

$$\xi = f(X, U) \quad (7)$$

$$\zeta = g(X, U) \quad (8)$$

where

- $\xi$  = vector of separation ratios
- $\zeta$  = matrix of dimensionless extent of reaction in case of reaction
- $S$  = matrix of stoichiometric coefficients (in case of reaction)
- $I$  = identity matrix
- $U$  = vector of process parameters

Retaining the simultaneous network solution using Eq. 5, the actual modelling is partitioned, because after each network solution (iteration) the values of  $P$ -matrices (split-streams) should be updated separately using nonlinear submodels, i.e., Eqs. 6, 7 and 8.

The method of computation outlined above is referred to as *quasilinear two-level modelling*. Quasilinear because the network is solved within each iteration as a linear one; two-level modelling because the procedure uses two types of models. The processing network solution using the linear submodels (Eq. 5) creates the upper level, the nonlinear submodels (Eqs. 6, 7 and 8) create the lower level of the modelling. For the first part of the computation it is necessary to construct network solution procedure before execution of material and heat balancing.

The simultaneous modular method of computation consists of two separate parts. The first is the so-called topological part or the construction of network solution procedure; the second is the rigorous material and heat balancing itself. A very important feature of the computation procedure is to assign the network calculation formulas by general network solution technique before the first iteration of the rigorous calculation. Accordingly, the first part of the method is performed merely once before the material and heat balancing and never repeated during the course of the calculation.

## NETWORK SOLUTION

The network solution technique is based on the *signal flow graph approach* which makes network-decomposition possible for components, thus unnecessary paths can be omitted. This part of the method is essentially the same as was used in the Proceval general process simulator of the simultaneous modular type (Nishimura and Yamashita, 1978).

After reading the problem specification the construction and solution of the signal flow graph are performed automatically. After the identification and classification of nodes the reduction of the graph is made by using equivalent transformations. The sequence of eliminating the middle nodes is assured by special preference rules. At the end of the reduction the graph contains only the input and the original looped nodes without self-loops. The values of looped nodes can be readily obtained by using the path values. The remaining nodes can be calculated easily by inserting these values back into the original graph, i.e., redefining the original structure. The construction of the algebraic solution of network is entirely general and can be used for any kind of simulation and any arbitrary structure.

## MATERIAL AND HEAT BALANCING

A separation column consists of a series of stages. According to our simultaneous modular strategy, all the four types of model equations (Eqs. 1, 2, 3 and 4) should be satisfied for each stage (all restrictions for one stage at a time). The column, in addition to vapor-liquid or liquid-liquid equilibrium stages, consists of reboiler, total or partial condenser, mixers and distributors.

The vapor-liquid and liquid-liquid stages, the reboiler and the partial condenser can be calculated by use of the concept of equilibrium stage. Considering an equilibrium stage ( $n$ ) the total feed of component  $i$ ,  $F_{in}$ , is separated into two phases,  $F_{in}\xi_{in} = y_{in}V_n$  and  $F_{in}(1 - \xi_{in}) = x_{in}L_n$ , respectively. The component balance equation around this stage is

$$\frac{y_{in}V_n}{\xi_{in}} = \frac{x_{in}L_n}{1 - \xi_{in}} \quad (9)$$

The expression of separation ratio by using the definition of equilibrium coefficient,  $K_{in} = y_{in}/x_{in}$ , is

$$\xi_{in} = \frac{K_{in}}{(L_n/V_n) + K_{in}} \quad (10)$$

which represents a general formula to compute separation ratio for any equilibrium stage. The equilibrium coefficient is dependent on pressure, temperature and composition.

The modelling of equilibrium stage requires the selection of the appropriate type of equation to provide a given variable. As in general, in our algorithm too, the component material balances

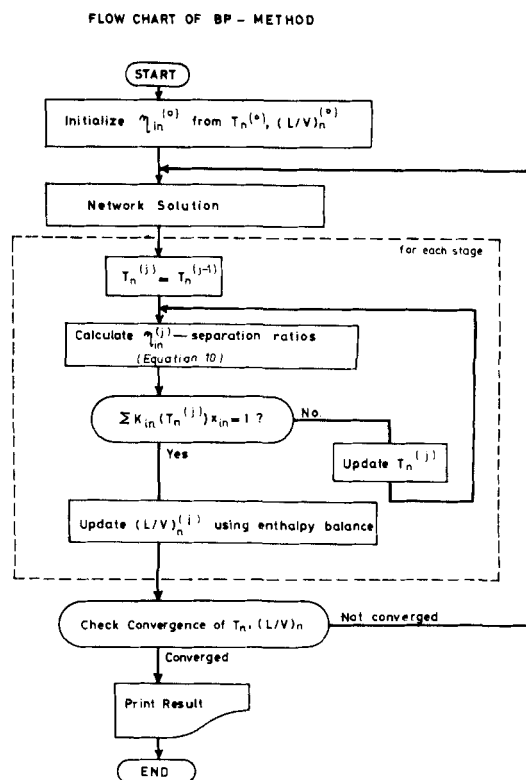


Fig. 1. Flow chart of BP-method.

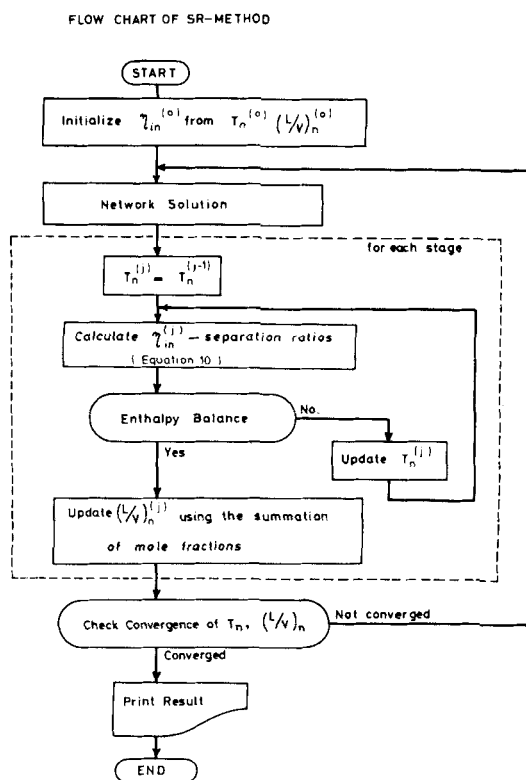


Fig. 2. Flow chart of SR-method.

combined with equilibrium relationships (Eqs. 1 and 2) are used to generate  $x_{in}$  and  $y_{in}$ —the molar composition of phases. After solving the component material balances two alternative exist

- Obtain the new  $T_n$  values from the restriction of summation of mole fractions (Eq. 4) and update  $L_n/V_n$  using enthalpy balances (Eq. 3), which is called bubble point (BP) type of method, or

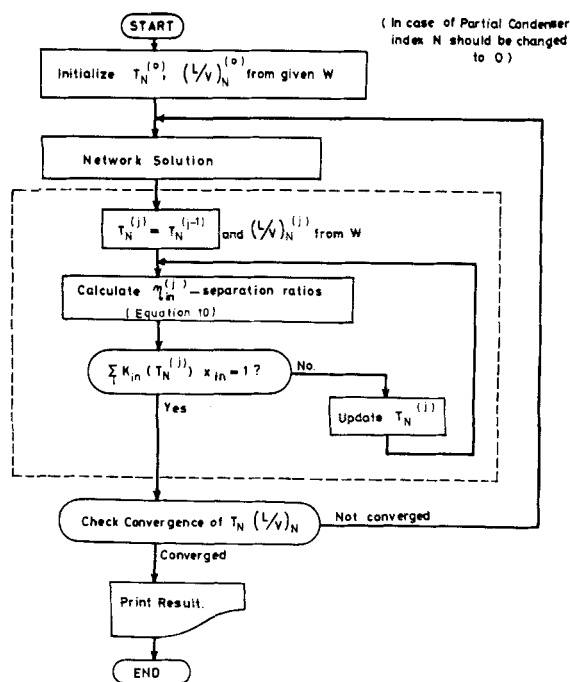


Fig. 3. Convergence loops of reboiler and partial condenser.

• Obtain the new  $T_n$  values from enthalpy balances (Eq. 3) and update  $L_n/V_n$  using the restriction of summation of mole fractions (Eq. 4), which is called sum of rates (SR) type of method. The logic sequence of our BP type of computation is shown in Figure 1. When latent heat effects predominate over sensible heat effects in the enthalpy balance calculation; i.e., the stage temperatures are physically determined more by compositions (restriction of summation of mole fractions) than by enthalpy balances, and total flows are determined more by enthalpy balances than by compositions, the BP method should be preferred. The criteria are satisfied by separations such as close-boiling distillation.

The logic sequence of our SR type of computation is shown in Figure 2. When sensible heat effects predominate over latent heat effects in the enthalpy balance calculation—the stage temperatures are physically determined more by enthalpy balances than by compositions and total flows are determined more by compositions than by enthalpy balances—the SR method should be preferred. Gas absorbers can be characterized by these criteria and distillation of wide boiling mixture also tends to favor this method (King, 1971). In the case of extraction process, the SR method is simplified by the absence of the energy balance equations from the model set.

The interpretation and calculation of mixer, total condenser and distributor are obvious. At mixers the flow rates of each component should be summarized, at distributors and total condensers no concentration-change occurs. The strategies of computation of partial condenser and reboiler are similar (Figure 3). Inside the dotted line of these flow charts, the modular computation is depicted and after calculating each stage separately, outside the dotted line, the simultaneous network solution is performed.

#### PROBLEM SPECIFICATION, INITIALIZATION AND PROMOTION

The problem specification of the suggested simultaneous modular method corresponds to that of the common equation solving methods; i.e., the number of equilibrium stages and the total flow rate of the distillate are specified. (At the sequential modular methods of the Lewis-Matheson type, instead of these specifications, the recoveries of the two key components in the products are specified.) The necessary input data include such specifications as structure of the process, connections between the units, com-

position and thermal condition of the feeds, reflux ratios, type of condensers, and criteria for convergence. The program is able to handle arbitrary process structure; i.e., the specification of any number of columns is allowable with arbitrary connections and allowance has been made for any number of feeds and side products.

The revaporization ratio of the reboiler ( $R' = V'/W$ ) can also be specified instead of the rate of distillate ( $R'$ -specification). This problem specification is more suitable for computation of absorbers and extractors. Since in case of  $R'$ -specification the recovery ratios of both ends of the column are fixed, the convergence is naturally more stable and quicker. In case of  $D$ -specification the value of  $R'$  should be updated continuously by normalization in each iteration.

In this method, the computation is commenced with assumed temperature profile, and it is continued until the temperature and the  $(L/V)$ -ratio at each stage do not change simultaneously any longer with further iterations. The temperature initialization is read by data input. The temperatures of the terminal stages have to be initialized, but the assignment of the initial values for the other stages is optional. A special linear interpolation routine assures the calculation between the given values. The initialization of  $\xi$ -values is performed automatically by applying Eq. 10 and using the concept of equimolar overflow for calculating  $(L/V)_{\text{mean}}$  from  $D, R, T$  and  $q$ :

$$\left(\frac{L}{V}\right)_{\text{Rect}} = \frac{RD}{(R+1)D} \quad (11)$$

$$\left(\frac{L'}{V'}\right)_{\text{Strip}} = \frac{qF + RD}{(R+1)D - (1-q)F} \quad (12)$$

$$\left(\frac{L}{V}\right)_{\text{mean}} = \frac{(L/V)_{\text{Rect}} + (L/V)_{\text{Strip}}}{2} \quad (13)$$

In case of polar mixtures, when equilibrium coefficients depend on composition, all recovery ratios are initialized as 0.5.

Inside the modular calculations the regula falsi method was used for determining  $(L/V)$  or  $T$  values. For the  $(L/V)$  and  $T$ -promotion, before the simultaneous network solution, the following five methods were tried and applied alternatively on our examples: direct substitution, group type of modified direct substitution, modified direct substitution, continual acceleration, and dominant eigenvalue method. In addition, allowance has been made for the promotion of the recovery ratios or node values, but from our experience the promotion of  $(L/V)$  and  $T$  values proved to be more efficient.

#### SEQUENCE OF CALCULATIONS

The calculational steps of the proposed method in the sequence are the following:

1. Read all data.
2. Initialize a set of temperatures ( $T_n^0$ ).
3. Interpret the structural data and make signal flow graph.
4. Initialize path values ( $\xi_{in}$ ).
5. Classify the nodes into input, looped, middle and tail nodes.
6. Reduce the graph until it contains only input and looped nodes.
7. Calculate the values of looped nodes from input nodes, then calculate the values of the remaining nodes (simultaneous linear network solution).
8. Call the necessary number and type of subroutines like: a. partial or total condenser; b. vapor-liquid or liquid-liquid equilibrium stage; c. reboiler; and d. mixer and distributor. Subroutines are to update  $(L/V)_n$ ,  $T_n$  and  $\xi_{in}$  by using nonlinear submodels (modular nonlinear calculation).
9. Check convergence of  $(L/V)_n$ ,  $T_n$  and, if necessary, promote these values.

The above procedure is repeated from Step 7 with the new values of  $(L/V)_n$  and  $T_n$  until the convergent criteria—the condition

when both the  $(L/V)_n$  and  $T_n$  at each plate show no trend to change with successive iterations—is satisfied.

## ILLUSTRATIVE EXAMPLES

This method has been programed for a HITAC 8800/8700 computer system at the Computer Center of University of Tokyo, and many test problems have been successfully solved. To illustrate the algorithm suggested, 16 examples are presented. The illustrative examples proved the stable application of the method to distillation with narrow and wide boiling mixtures as well as with nonideal mixtures, especially to absorption, extraction and interlinked distillation columns. The convergence criteria used for these problems were

$$\left| \frac{\Delta T}{T} \right| \leq 0.001 \text{ and } \left| \frac{\Delta(L/V)}{L/V} \right| \leq 0.001$$

between successive trials. The schematic diagrams of the calculated structures are shown in Figure 4. The most important features, the necessary number of iterations, and the total CPU times of the illustrative example are given in Table 1.

**Examples 1 . . . 4.** The first four examples are the variations of the 11-stage, five-component separation problem with a relative narrow-boiling mixture described by Holland (1963). Propane, *i*-C<sub>4</sub>, *n*-C<sub>4</sub>, *i*-C<sub>5</sub> and *n*-C<sub>5</sub>, is to be separated by distillation and the feed is fed to Plate No. 4 (Stage No. 5). The amounts of each component in the feed in kmol/h are: 5, 15, 25, 20 and 35. Here the rate of distillate is fixed to 48.9 kmol/h, and the reflux ratio,  $L_0/D$  is 2.58.

The polynomial functional forms of the equilibrium and enthalpy data used in these examples are given in the Holland book. Using the same initial assumptions as Holland in Example 2, the solution set of temperatures, vapor rates, and product distributions are found to be in close agreement with the reported results. Figure 5 shows the steady mode of convergence of the method for the examples. Here for simplicity only the variation of vapor rate at the top plate with each iteration is plotted. It is important to notice, that the required CPU time was less than 5 seconds for each example.

**Examples 5 . . . 8.** These examples are the variations of the 13-stage, 11-component distillation problem with a wide-boiling mixture described by Holland (1963). The mixture of CH<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub>, *i*-C<sub>4</sub>, *n*-C<sub>4</sub>, *n*-C<sub>5</sub>, *n*-C<sub>6</sub>, *n*-C<sub>7</sub>, *n*-C<sub>8</sub> and the 360<sup>+</sup>-fraction is separated and the feed is fed to Plate 4. The component flow rates are 2, 10, 6, 12.5, 3.5, 15, 15.2, 11.3, 9, 8.5 and 7 kmol/h, the rate

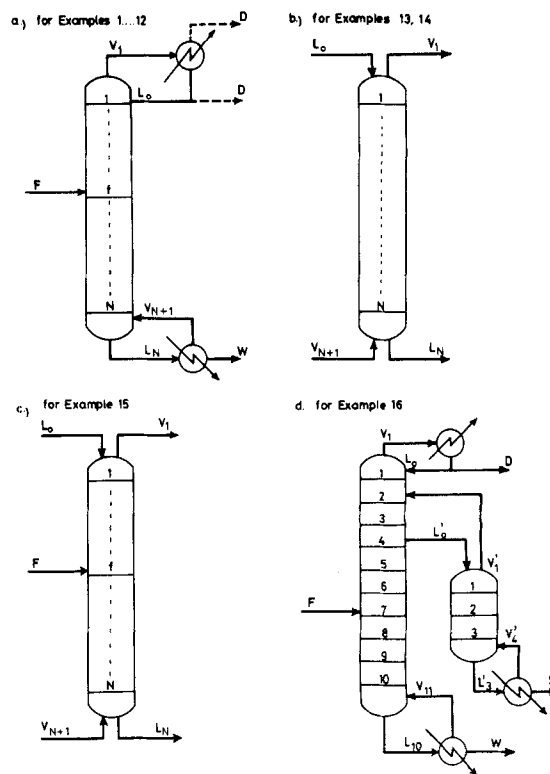


Fig. 4. Schematic diagrams of the calculated structures.

of distillate is fixed to 31.6 kmol/h and the reflux ratio is 2.0. The functional forms used for calculating the equilibrium and enthalpy data in these examples are presented in the original literature.

Using the same initial assumptions as Holland in Example 8, we obtained for product distributions the results shown in Table 2 with comparison to the reported values. These compare favorably with each other. We can notice that the number of iterations for the wide-boiling problem is higher than that for narrow-boiling problem. The required CPU computer time was reasonable, between 22 and 40 seconds for each example.

**Examples 9, 10.** The system of four components *i*-C<sub>5</sub>, *n*-C<sub>5</sub>, *n*-C<sub>6</sub> and *n*-C<sub>7</sub><sup>+</sup> is to be separated by distillation. The column corresponds to the 87 theoretical-stage industrial *i*-C<sub>5</sub>, *n*-C<sub>5</sub> separation tower investigated by Földes and Fonyó (1978). The component flow rates of the feed are 78.58, 83.36, 95.44, 35.83 kmol/h, and

TABLE 1. SPECIFICATIONS AND MOST IMPORTANT FEATURES OF THE ILLUSTRATIVE EXAMPLES

Items	Example	No. of Stages (N + 2)	No. of Components (C)	(N + 2)/C	Type of Condenser†	Convergence Method Used	Specification	No. of Iteration Necessary to Reach Convergence	Total CPU-Time, s
Distillation with Narrow-Boiling Mixture	1	11	5	55	T	BP	R'	15	3
	2	11	5	55	T	BP	D	22	5
	3	11	5	55	P	BP	R'	14	3
	4	11	5	55	P	BP	D	13	3
Distillation with Wide-Boiling Mixture	5	13	11	132	T	SR	R'	28	22
	6	13	11	132	T	SR	D	33	29
	7	13	11	132	P	SR	R'	31	26
	8	13	11	132	P	SR	D	35	30
Distillation for Industrial Pentan Separation	9	87	4	348	T	BP	R'	36	80*
	10	87	4	348	T	BP	D	90	140**
Distillation with Nonideal Mixture	11	12	3	36	T	BP	R'	26	32
	12	12	3	36	T	BP	D	24	3
Absorption	13	8	14	112	—	SR	R'	31	10
	14	20	14	280	—	SR	R'	71	44
Extraction	15	15	4	60	—	Special SR	R'	14	3
Interlinked Distillation Columns	16	16	6	96	T	BP	R'	22	10

† P = partial; T = total.

\* 57 s for network solution procedure and 31 for calculation.

\*\* 57 s for network solution procedure and 83 s for calculation.

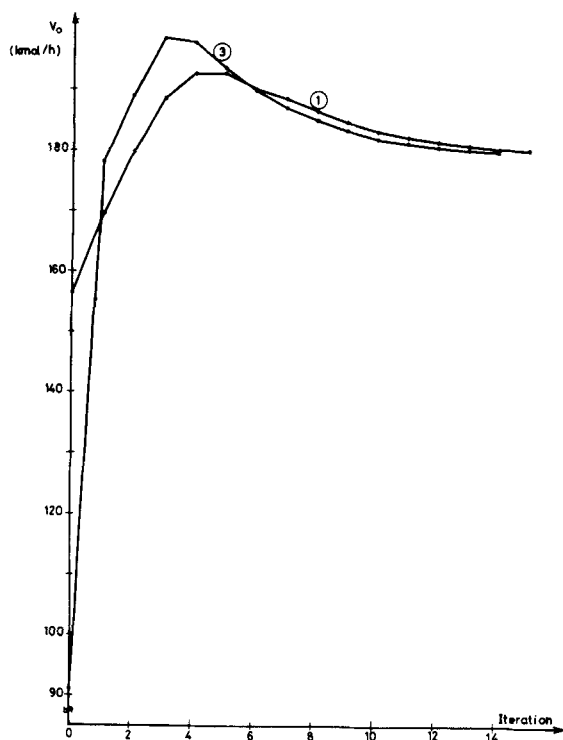


Fig. 5. Mood of Convergence.

TABLE 2. PRODUCT DISTRIBUTIONS FOR EXAMPLE 8

Components	Simultaneous Modular Method		Holland's Method	
	Distillate (kmol/h)	Bottoms (kmol/h)	Distillate (kmol/h)	Bottoms (kmol/h)
CH <sub>4</sub>	2.000	0.0	2.000	0.0
C <sub>2</sub> H <sub>6</sub>	9.99989	0.00007	9.99994	0.00005
C <sub>3</sub> H <sub>6</sub>	5.9541	0.045864	5.96602	0.03397
C <sub>3</sub> H <sub>8</sub>	12.20349	0.2964	12.27752	0.22247
i-C <sub>4</sub>	0.67313	2.826816	0.6694	2.83059
n-C <sub>4</sub>	0.813577	14.18626	0.6820	14.31799
n-C <sub>5</sub>	0.01034	15.1895	0.00505	15.19495
n-C <sub>6</sub>	0.000016	11.2998	0.0	11.29994
n-C <sub>7</sub>	0.0	8.99999	0.0	8.99999
n-C <sub>8</sub>	0.0	8.49999	0.0	8.49999
360+	0.0	6.99999	0.0	6.99999
Total	31.6546	68.3446	31.601	68.399

the feed is sent to plate 42. The rate of distillate is fixed to 81.02 kmol/h, and the reflux ratio is 12.0. The calculated results as well as the favorable comparison with industrial data is presented in Table 3. Unlike the preceding examples, the topological part requires considerable CPU time here (57 s), while the computation time of material and heat balancing is reasonable (31–83 s). To save computation time in this case a sort of system engineering approach was followed; i.e., the network solution procedure was assigned merely once, and these intermediate results are stored and retrieved for every calculation.

**Examples 11, 12.** These examples are the variations of the 12-stage distillation problem with a three-component nonideal system described by Hanson et al. (1962). The feed is liquid and fed to Plate 6. The composition effect on the equilibrium relation is expressed by means of activity coefficients as  $K_{mod} = \gamma (K_{ideal})$ , where the values of gamma (activity coefficients) are determined by a three suffix Margules equation described in the reference.

Using the same initial assumptions as Hanson et al., in Example 12, we obtained for product distributions the results shown in Table 4, with comparison to the reported values.

**Examples 13, 14.** These examples correspond to the 14-component hydrocarbon absorption problems described by Holland

TABLE 3. PRODUCT DISTRIBUTIONS FOR EXAMPLE 10

Components	Simultaneous Modular Method		Industrial Data	
	Distillate (kmol/h)	Bottoms (kmol/h)	Distillate (kmol/h)	Bottoms (kmol/h)
i-C <sub>5</sub>	78.04	0.5289	78.29	0.293
n-C <sub>5</sub>	3.06	80.282	2.74	80.623
n-C <sub>6</sub>	0	95.437	0	95.441
n-C <sub>7</sub> <sup>+</sup>	0	35.829	0	35.831
Total	81.1	212.076	81.03	212.188

TABLE 4. PRODUCT DISTRIBUTIONS FOR EXAMPLE 11

Components	Simultaneous Modular Method		Hanson's Method	
	Distillate (mol)	Bottoms (mol)	Distillate (mol)	Bottoms (mol)
A	0.1830	0.0560	0.1817	0.0573
B	0.0224	0.0226	0.0330	0.0119
C	0.0946	0.6214	0.0853	0.6307
Total	0.2999	0.7001	0.3000	0.7000

TABLE 5. TEMPERATURES AND VAPOR RATE PROFILES FOR EXAMPLE 13

Plate No.	Simultaneous Modular Method		Holland's Method	
	Temperature Profile (°F)	Vapor Rate Profile (kmol/h)	Temperature Profile (°F)	Vapor Rate Profile (kmol/h)
1	29.52	2,774	27.93	2,659
2	32.25	2,889	31.04	2,813
3	30.36	2,899	30.90	2,825
4	27.80	2,907	29.59	2,834
5	25.91	2,915	27.66	2,843
6	23.58	2,924	25.10	2,855
7	19.37	2,936	21.47	2,873
8	14.21	2,955	15.32	2,905

TABLE 6. NUMBER OF ITERATIONS FOR EXAMPLE 15

Method	Relaxation Factor	No of Iterations
Hanson's Method	—	19
Newton-Raphson	—	7
Relaxation	5	21
Relaxation	10	17
Relaxation	100	20
Simultaneous Modular	—	14

(1975) (Examples 4-3 and 4-4). The towers are provided with eight and 20 theoretical stages, respectively. Using the same initial assumptions and the same equilibrium, enthalpy and correction data, we obtained the results shown in Table 5 for product distribution. Here also a comparison with Holland's results are presented for Example 13. From these two examples we can notice that the number of iterations and the CPU times are proportional to the number of theoretical stages.

**Example 15.** The example is the 15-stage, four-component extraction problem described by Hanson et al. (1962). Two solvents are used to separate an equimolar mixture of acetone and ethanol by extraction. The solvents are chloroform and water. The feed is sent to Stage 5, chloroform to Stage 1, and water to Stage 15. The flow rates are: feed, 0.2 kmol/h; chloroform, 0.8 kmol/h; water, 1.0 kmol/h.

The equilibrium data are described by the Margules equation and reported in the reference cited above. To demonstrate the effectiveness of the simultaneous modular method for lack of

product specification, a comparison is presented in Table 6 with the number of iterations by Jelinek and Hlavacek (1976).

**Example 16.** The two columns shown in the sketch (Figure 4d) are interlinked. The example is a slight modification of the two-column problem described by Hanson et al. (1962). The only feed to the system is a six-component feed of 1 kmol/h to the seventh plate. The main column has a total condenser, reboiler, and ten plates. The reflux ratio and revaporization ratio are set at 3.105 and 2.141, respectively. The liquid side stream is set at 0.5 times the actual liquid flow and drawn from the fourth plate and sent to the top plate of the side stripper. Vapor from the top plate of the stripper is returned to Plate 2 of the main column. The side stripper has a reboiler and three plates, and the revaporization ratio is set at 1.422.

It is important to note here that the algebraic solution of the network is performed for the whole system all at once and the two columns are calculated simultaneously, moreover the number of necessary iteration and CPU-time are fairly favorable.

## DISCUSSION

A new flexible method has been developed for calculating the multistage, multicomponent separation processes. The numerical examples with widely different characters showed the usefulness and competitiveness of the simultaneous modular approach, although it cannot be said that the convergence by the new method in all cases were better than that by the equation-solving approach.

In scarcity of product specification (or in abundance of reflux ratio and revaporization ratio specification) and in case of more complicated system structure, the simultaneous modular method proved to be more effective. The present method is highly system engineering oriented, since the adaptation of the simultaneous modular strategy renders to use the same network solution package possible at different hierarchical levels for the computation of processing units, chemical plants, chemical complexes, etc.

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## NOTATION

$C$  = total number of components  
 $D$  = total flow rate of distillate  
 $F_n$  = feed flow rate to stage  $n$   
 $H$  = enthalpy  
 $I$  = identity matrix  
 $K_{in}$  = equilibrium vaporization constant or distribution coefficient of component  $i$  in stage  $n$   
 $L_n$  = total flow rate of liquid or extract phase leaving the  $n$ th stage  
 $N$  = total number of theoretical plates  
 $P$  = transformation matrix  
 $P_{LN}$  = liquid side stream flow rate from stage  $n$   
 $P_{Vn}$  = vapor side stream flow rate from stage  $n$   
 $q$  = factor related to the thermal condition of feed ( $q = (L_{strip} - L_{rect})/F$ )  
 $q_n$  = heat entering stage  $n$   
 $R$  = reflux ratio ( $L_o/D$ )

$R'$  = revaporization ratio ( $V'/W$ )  
 $S$  = total flow of side product  
 $S$  = matrix of stoichiometric coefficients in case of reaction  
 $T$  = temperature  
 $T_n$  = temperature on stage  $n$   
 $U$  = vector of process parameters  
 $V_n$  = total flow rate of vapor or raffinate phase leaving the  $n$ th stage  
 $W$  = total flow rate of bottoms  
 $x$  = input vector  
 $x_{in}$  = mole fraction of component  $i$  in liquid or extract phase in stage  $n$   
 $Y$  = output vector  
 $y_{in}$  = mole fraction of component  $i$  in vapor or raffinate phase in stage  $n$   
 $\xi$  = vector of separation ratios  
 $\xi_{in}$  = separation ratio of component  $i$  on stage  $n$

## Subscripts

$F$  = variables associated with feed  
 $f$  = variables associated with feed stage  
 $i$  = component number  
 $in$  = input  
 $n$  = stage number  
 $out$  = output  
 $rect$  = rectifying section  
 $strip$  = stripping section

## Superscript

$(j)$  = iteration number

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