OPTIMUM DESIGN OF DISTILLATION SYSTEMS

by

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ABSTRACT

The optimum design of a distillation system is posed as a constrained nonlinear problem, which enables the simultaneous determination of the optimal configuration and the design variables of the given configuration. Initially the problem is formulated as a mixed-integer nonlinear problem because the set of optimization variables includes integer number for the ideal stages of each section of a given column. Since no explicit algorithm is yet available for solving this class of problem, the integer optimization variables are treated as real ones and a constrained nonlinear problem is solved.

The optimization of a single column with two or more products is carried out minimizing an objective function which is the nonlinear total cost of the column and the specifications for the products are expressed as nonlinear constraints. The use of short cut methods to model the distillation column, widely used in the design of distillation systems, has been replaced by a simple, but more rigorous, plate to plate model of the column that can be made as sophisticated as desired as the design evolves, thus avoiding the restrictions imposed by short cut methods and obtaining more realistic information on the system.

The optimal configuration of a series of distillation columns is found by optimizing a general configuration that contains most of the possible arrangements between columns. The objective function is the total cost of the general configuration, the nonlinear constraints are the specifications of the products and bounds on the internal flow rates of the columns and the optimization variables are the ideal stages in all the sections of the columns, the flow rates of the intermediate streams interconnecting columns and the heat exchanged in the condensers,

reboilers and intermediate heat exchangers of the system. Some of the configurations frequently used in industry can be found as particular simplifications of the general configuration proposed, but when energy costs dominate unconventional systems with strong thermal coupling between columns are usually obtained.

CONTENTS

ERRATA

p. 8, line 22: replace 'though an outer loop' by 'where an outer loop was used to search for the total number of stages on each column and an inner loop'.

p. 22, line 11: replace 'exchanger rate much' by 'transfer rate must'.

p. 30, line 18: replace 'compnent' by 'component'.

p. 43, line 5: replace 'Shubert' by 'Schubert'.

p. 60: insert at the beginning 'Broyden Method'.

p. 69, line 16: replace 'trys' by 'trays'.

p. 70, line 3: replace 'volatiles' by 'volatilities'.

p. 75, line 3: replace 'percentage' by 'percentage rate'.

p. 159, line 2: insert 'AIChE J'.

p. 162, line 25: replace 'Happel and Jordon' by 'Happel, J. and D.G. Jordan'.

p. 163, line 1: replace 'Shaham' by 'Shacham'.

p. 165, line 11: replace','by', AIChE J. 26, 916 (1980).

p. 176, line 1: replace 'increase' by 'inverse'.

p. 176, line 23: replace 'unity' by 'the value of unity'.

p. 176, line 16: replace 'Shubert' by 'Schubert'.

CHAPTER I

INTRODUCTION

Distillation is one of the most widely used separation processes in industry, accounting for 20 to 50 percent of the total energy consumption in petroleum refineries; it is a highly energy-intensive process. When a complex mixture is to be separated into several fractions, the designer is faced with the problem of choosing the configuration of columns and heat exchangers to be used, as well as the size and the operation conditions of each unit.

The distillation system to be designed must satisfy given specifications, which are represented by bounds on the system variables or algebraic expressions between the system variables. A good model of the distillation process is necessary to achieve the design requirements and is expressed by a set of algebraic equations. The system of equations, including the model and the specifications, is likely to be underdetermined mainly because most of the design requirements are inequality expressions. Therefore an infinite number of alternatives exists where a number of design variables equivalent to the degrees of freedom of the system has to be specified. A good, or the best, alternative can be selected following some specific criteria.

It is often possible to formulate a quantitative measure of desirability, called an objective function, so that the optimal choice corresponds to maximizing this function. Frequently the basis of this measure is economic, and the objective function is simply the cost; in this case of course the optimum choice corresponds to minimizing the cost.

Techniques developed in the area of nonlinear programming can be used to solve the design problem, where the extremun of the objective function is sought in the feasible space defined by the constraint equations, which are the design specifications, operating restrictions and sometimes the equations modelling the system.

In the last decade, a great deal of effort has been directed towards the development of computer based methods for the selection of good configurations for chemical processes in general and for distillation columns in particular. The selection of the design variables has been carried out on two levels: an outer loop where the structure of the system, mainly the connections between different units of equipment, is determined and an inner loop where the size and operating conditions of each unit is found. The use of decomposition techniques is mainly due to the big number of variables involved in the solution of the problem and the fact that different alternatives frequently involve discrete decisions, such as which columns should be used and in which order they should be connected. Furthermore, in the case of distillation columns, there is a set of integer variables representing the plate numbers in each section of a given column. Because of these integer variables, the resulting optimum design is a mixed-integer nonlinear problem.

The optimization of the total number of plates in each column was first attempted by Gaminibandara and Sargent (5), though an outer loop was used to search for the values of the continuous variables that would minimize the same objective function.

In Chapter IV, the optimum design of a single distillation column is carried out. The integer variables were chosen simultaneously using a standard nonlinear programming algorithm to minimize the cost of running the distillation column. The model and the algorithm used for the simulation of a single distillation column are described in Chapter II.

Most of the work on synthesis of distillation columns has been concerned with the configurations of columns carrying out sharp separations, thus reducing the synthesis problem to search for the optimum configuration among a predefined set of possibilities, where each column has only two products and the use of mass recycles is severely limited. Coupled columns, as the ones shown by Petlyuk et al (56, 57), often give substantial savings over more conventional separation sequences. It is unlikely that the non-conventional coupled arrangement between columns will be synthesized using heuristic or evolutionary rules. Gaminibandara and Sargent (5) developed a general system of interlinked columns from which most configurations of interest can be obtained by deletion of parts of the general structure. The appropriate deletions were obtained by direct optimal design of the general configuration. The same approach has been used in the present work. Therefore, the optimum arrangement between columns will be found by analysis of the general structure, the synthesis step being the one of finding the general structure. A slightly more general configuration than the one presented by Gaminibandara and Sargent (5) will be used in this thesis. They used two optimization levels while in the algorithm presented in Chapter V all the design variables are optimized simultaneously, thus requiring only one optimization level. The simulation of the general configuration is presented in Chapter III.

CHAPTER II

SIMULATION OF A DISTILLATION COLUMN

2.1 Introduction

In an ordinary distillation column, the feed is introduced into a vertical cascade of stages. Vapour rising in the section above the feed is washed with liquid to remove the less volatile components. The liquid is provided by condensing some of the vapour leaving the top of the column and returned as reflux liquid. In the section below the feed, the liquid is stripped of volatile components by vapour produced at the bottom by vaporization of the liquid in the reboiler. Some of the more volatile components are vaporized from the liquid, increasing the light component concentrations on the vapour and some of the less volatile components are condensed from the vapour increasing the heavy component concentrations in the liquid. Most of the columns are fitted with plates which are arranged to bring about the best possible contact between liquid and vapour. The liquid flows by gravity from one stage to the other.

For mass transfer to take place the streams entering each stage must not be in equilibrium with each other, for it is the departure from equilibrium that provides the driving force for transfer. The leaving streams are usually not in equilibrium either but are much closer to being so than the entering streams. If the mixing in a given stage is so effective that the leaving streams are in fact in equilibrium, the stage is, by definition, an ideal stage.

The purities obtained for the withdrawn products will depend upon the liquid-gas ratios used and the number of stages provided.

In Figure 2.1 a typical column and plate are shown.

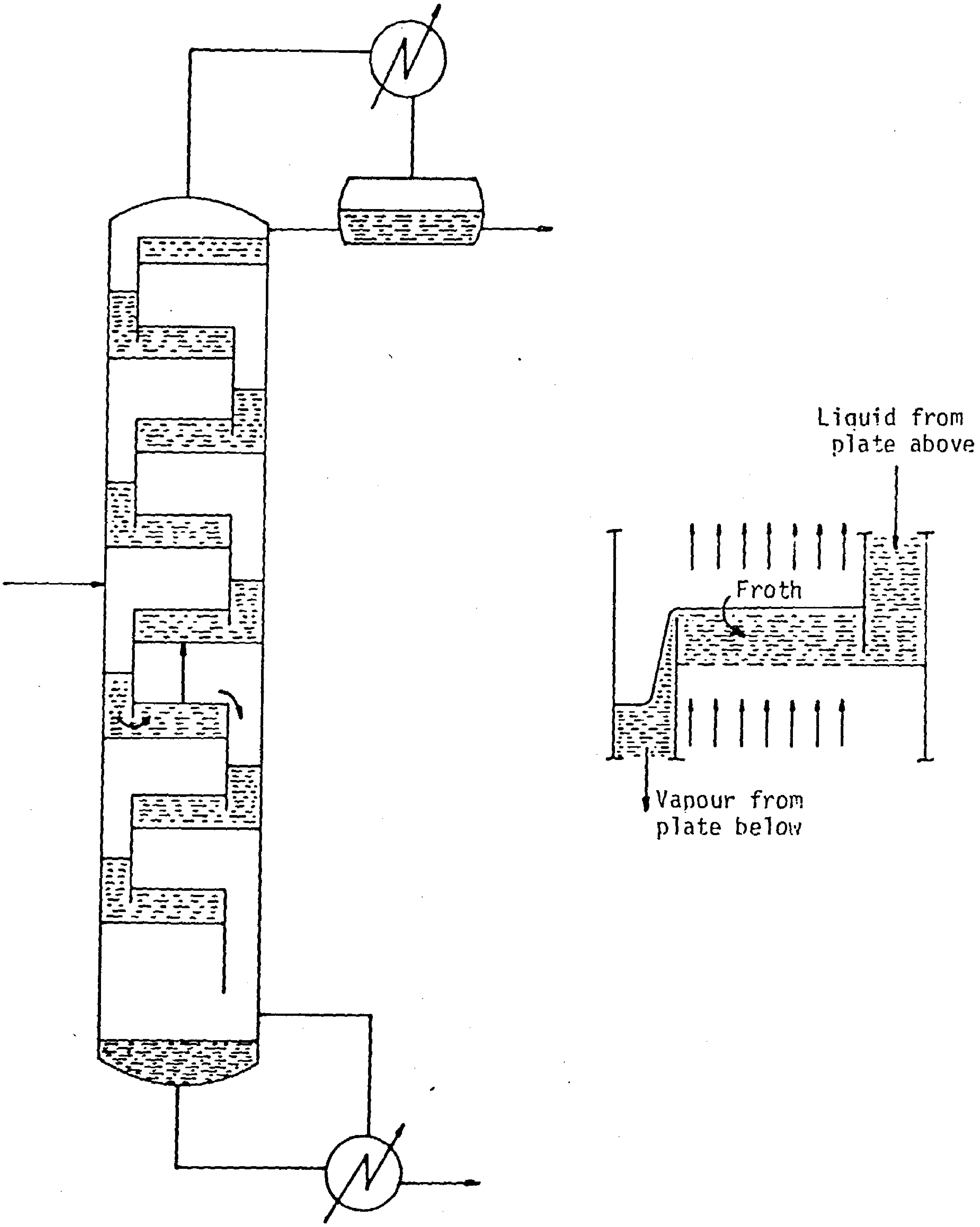

Figure 2.1

2.2 Statement of the Problem

The solution of a multistage distillation problem requires mass balances, heat balances and equilibrium conditions to be satisfied over each stage of the process. The basic equations are often simple but the countercurrent method of operation interconnects all stages. More specifically, the solution of the equilibrium stage model for a separation process is obtained by finding a set of temperatures, phase flow rates and compositions which satisfy the following equations:

1. The equilibrium relationship

$$y_{i,p} = K_{i,p} \, x_{i,p} \qquad (2.1)$$

2. The component mass balance around stage p

$$L_p \, x_{i,p} + V_p \, y_{i,p} - L_{p-1} \, x_{i,p-1} - V_{p+1} \, y_{i,p+1} - f_{i,p} = 0 \qquad (2.2)$$

3. The energy balance around stage p

$$L_p h_p + V_p H_p - L_{p-1} \, h_{p-1} - V_{p+1} \, H_{p+1} - F_p \, h_{Fp} - Q_p = 0 \qquad (2.3)$$

4. The summation restrictions

$$\sum_i x_{i,p} = 1.0 \quad \text{and} \quad \sum_i y_{i,p} = 1.0 \qquad (2.4)$$

Some methods of solution are based on tearing and partitioning the system, so that subsets of variables are paired with subsets of equations through which they show their greatest effects. For example, Equation (2.1) can be used to eliminate the vapour compositions throughout the other three equations. The compositions are obtained from the component mass balance Equations (2.2). Two arrangements are then possible, the "boiling point" (BP) arrangement matches the temperatures with the summation Equations (2.4) and the flow rates with enthalpy balance Equations (2.2). The "sum-rates" (SR) arrangement matches the enthalpy balances with stage temperatures and the summation equations with flow rates. Friday and Smith (1) analyzed the circumstances under which different pairings are appropriate. To consider the convergence characteristics of the BP and SR approaches, they studied these methods as a system of two sets of variables, the flow rates and temperatures and two sets of equations representing the successive substitution procedures for generating mean values of the variables, the enthalpy and summation equations. The rate of convergence of the Direction Substitution (DS) method near the solution can be predicted knowing the dominant eigenvalue of the Jacobian of the iteration matrix at the solution, as will be seen in more detail in Appendix B. If the absolute value of the dominant eigenvalue is less than unity the DS method will converge, otherwise it will diverge. Friday and Smith (1) calculated the values of the dominant eigenvalues of the Jacobian matrices for different examples and found that BP methods have an absolute value of the dominant eigenvalue less than unity for relatively narrow boiling feeds and that the dominant eigenvalue increases as the range of temperatures of the components of the mixture increases, reaching values bigger than unity for wide boiling mixtures. On the other hand, the SR methods have the absolute value of the dominant eigenvalue less than unity for wide boiling mixtures and the magnitude of the dominant eigenvalue increases as the difference in temperatures of the different elements in the mixture decreases.

From this work it follows that the BP method should be used for narrow-boiling mixtures and the SR method for wide-boiling mixtures. However, Friday and Smith (1) found that the range of applicability of the BP and SR methods increases considerably by damping the changes on the variables from iteration to iteration.

Amundson and Pontinen (2) noticed that iterating on the temperatures and flow rates, the component mass balance equations form a set of linear equations which are easily solved by standard matrix procedures. They reduced the nonlinear part of the problem to one involving plate temperatures only, which they treated as non-interacting and corrected using the Newton-Raphson method after normalizing liquid plate compositions.

Wang and Henke (3) eliminated the inversion of the matrix proposed by Amundson and Pontinen (2) and solved the linearized mass balance equations using Thomas' method for tridiagonal matrices; Muller's method was used to accelerate convergence of the temperatures. Sargent and Murtagh (4) used the same linear subsets and introduced an inner loop to obtain temperatures from boiling point or dew point calculations, thus being able to deal with non-ideal mixtures. Sargent and Gaminibandara (5) performed a flash calculation for each plate to find the temperatures, avoiding the instability due to the presence of very light and very heavy components in the bubble and dew points calculations.

One disadvantage of the BP approach arises when the equilibrium ratios are composition dependent. The liquid and vapour compositions are not known at the point when it is necessary to calculate composition dependent equilibrium ratios, that is, just before the component mass balance equations are solved. In such problems, either the compositions from a previous trial must be used or an extra iteration loop must be introduced. If the composition lag approach is adopted, the convergence properties of the iteration algorithm may be adversely affected, especially for highly non-ideal systems. The second approach could be costly in terms of computational effort.

The pairing problem is avoided if all the equations are solved simultaneously. Stainthorp and Whitehouse (6) solved all the equations describing the system, including mass and energy balances, physical data correlations and performance specifications, as one large set of nonlinear algebraic equations, using the Newton method. The method involves the computation of the Jacobian matrix and makes large demands on computer storage. In a second paper, Stainthorp, Whitehouse and Kandela (7) solved the mass balance for each component separately as a linear system, thus reducing the nonlinear set of equations to be solved with the Newton method. As equilibrium and enthalpy data were independent of compositions, the storage requirement was independent of the number of components, thus reducing the working space. They placed constraints on the relative change of variables to stabilize the method. Using the Newton method, quadratic convergence can be obtained near the solution but the calculation of the Jacobian matrix each iteration could be time consuming.

Tierney and Yanosik (8) noticed that the Jacobian matrix changes little near solution. Orbach, Crowe and Johnson (9) kept the Jacobian matrix unchanged for several iterations and only when the rate of convergence was slow was a new Jacobian matrix calculated. Tomich (10) adjusted temperatures and flow rates simultaneously at each iteration, solving the enthalpy and summation equations with Broyden's method (11). The tridiagonal matrices for the component mass balance were solved separately.

In Broyden's method, which is described in Appendix D, an approximation of the Jacobian or its inverse is used and the approximation is revised after each step on the basis of information generated during the step. No additional function evaluations are required.

The Jacobian matrix has a stagewise sparse structure for multistage procedures and advantage can be taken of the fact that the sparsity pattern is known a priori to develop more efficient solutions. Naphtali and Sandholm (12) exploited the block tridiagonal structure of the Jacobian when the equations were grouped by stage. Ishii and Otto (13) solved all the equations simultaneously using the Newton-Raphson method to obtain temperatures, flow rates and compositions, but the linearized equations only included the partial derivatives that have a dominant influence on the solution and this greatly reduces the computational effort required.
Shewchuk and Hutchinson (14) solved the set of nonlinear equations by linearizing the coefficients describing the equilibrium relationships and enthalpies of the internal streams. These coefficients are re-estimated between iterations and since all components are treated simultaneously it is possible to let the equilibrium ratio depend on all compositions on a plate. Shewchuk (15) extended the method for non-ideal distillation calculations. In this so-called "Quasi-Linear" method, to obtain second order convergence, it is necessary and sufficient that the coefficients of the linear equations be chosen such that they are the partial derivatives of the nonlinear functions with respect to the appropriate variables, and in this case the method reduces to Newton's method.

A large fraction of the computer time is consumed in thermodynamic calculations. Barrett and Walsh (16) introduced a Thermodynamic Interface Package designed to provide local approximation models of fugacities and enthalpies to be used in the process simulation. These models are generated from a full thermodynamic computer package. The availability of simple analytical expressions for their derivatives allow the use of better algorithms in the solution of equations which contain these properties, thus accelerating convergence.

Boston and Sullivan (17) introduced the use of newly defined energy and volatility parameters as the primary iteration variables together with a third parameter defined for each stage as a combination of the liquid and vapour phase rates and temperatures. Reasonably good initial estimates of the energy parameters can be obtained by using quite inaccurate initial estimates of the temperatures and interstage phase rates. Boston (18) used direct substitution or the bounded Wegstein method for the outer loop variables, in this case the energy and volatility parameters, but the Broyden method with the identity matrix as the initial guess for the Jacobian matrix was used to improve convergence on the third parameter defined for each stage. The main advantage of this so-called "inside out" approach is that the new variables are relatively free of interaction with each other, and are relatively independent of the primitive variables (temperatures, flow rates), hence precise initialization is not critical to good algorithm performance.

Hlavacek and Van Rompay (19) classify the separation problems in five differential degrees of difficulty and propose different methods of solution for each case. The use of the Leverberg-Marquardt method with the Broyden formula to update the Jacobian is recommended for difficult problems of small or medium size. For simple distillation problems in which there is a weak interaction in the liquid phase and the volatilities in the column are of the same order of magnitude, they reckon that decomposition methods (e.g. Wang and Henke (3)) will easily solve the system.

The main advantage of the Newton or quasi-Newton methods is that any process specification that can be expressed as an algebraic equation can be solved simultaneously with the rest of the equations describing the distillation system. The storage required for the method describing the distillation is very important, considering the dimension of the problem to be solved in the optimization of the general configuration presented in Chapter V. Pairing methods require much less storage than simultaneous solution methods, and so a traditional approach of decomposing the general system and coupling certain equations with given variables was chosen. The BP method was used given the fact that it solves most distillation problems.

2.3 Equations Describing the Distillation Column

The notation for a typical plate is illustrated in Figure 2.2 where the stages have been numbered from the top to the bottom of the column and the nomenclature used was

$x_{i,p}$	liquid mole compositions
$y_{i,p}$	vapour mole compositions
V_p	vapour molar flow rate
L_p	liquid molar flow rate
F_p	feed molar flow rate
P_p^V	vapour molar product flow rate
P_p^L	liquid molar product flow rate

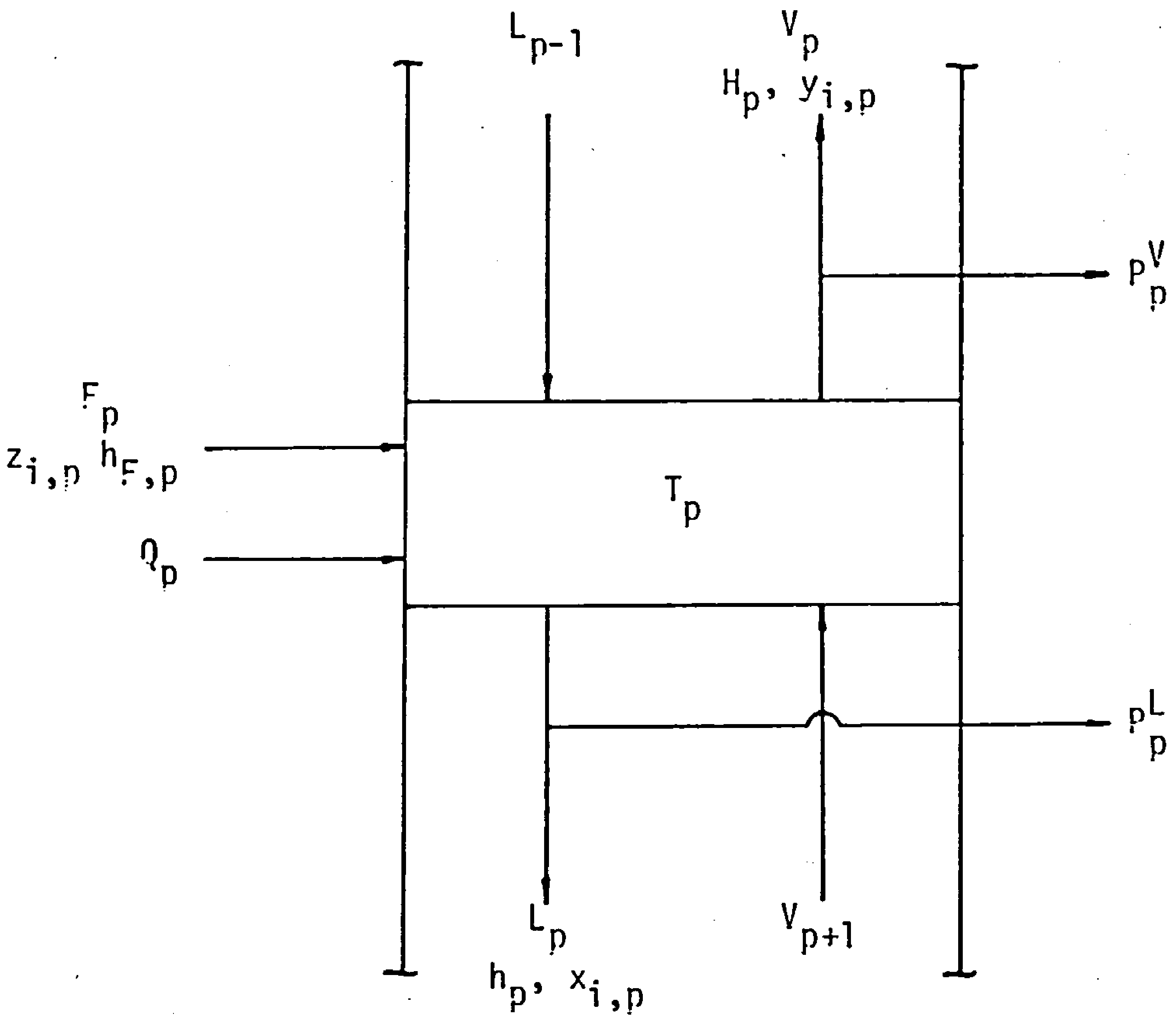

Figure 2.2 - Notation of a typical plate

Q_p	heat exchanged
T_p	temperature
H_p	vapour enthalpy
h_p	liquid enthalpy
p	stage number

The basic relations are derived from heat and material balances and the use of equilibrium conditions for each stage.

Ideal stages are assumed throughout, so that vapour and liquid leaving a given plate are in thermal and phase equilibrium given by the relations

$$y_{i,p} = K_{i,p}\, x_{i,p} \quad , \quad i=1,2, \ldots \quad c \tag{2.5}$$

where $K_{i,p}$ is the equilibrium ratio defined in terms of the fugacity coefficients $\phi^L_{i,p}$, $\phi^V_{i,p}$ for the liquid and vapour phases respectively:

$$K_{i,p} = \frac{\phi^L_{i,p}}{\phi^V_{i,p}} \quad , \quad i=1,2, \ldots \quad c \tag{2.6}$$

The fugacity coefficients are functions of pressure π_p, temperature T_p and composition of the phase in question:

$$\phi^L_{i,p} = \phi^L_i\,(\pi_p, T_p, x_{i,p}) \tag{2.7}$$

$$\phi^V_{i,p} = \phi^V_i\ (\pi_p,\ T_p,\ y_{i,p}) \tag{2.7}$$

A material balance on plate p yields

$$F_p + V_{p+1} + L_{p-1} = V_p + P^V_p + L_p + P^L_p \tag{2.8}$$

For the top plate of a column (p=1) there are no upper plates so we have

$$V_1 = L_o = 0 \tag{2.9}$$

while if m is the bottom plate of a column we have

$$L_m = V_{m+1} = 0 \tag{2.10}$$

A similar balance for a typical component i yields

$$z_{i,p}\ F_p + y_{i,p+1}\ V_{p+1} + x_{i,p-1}L_{p-1} = y_{i,p}(V_p+P^V_p) + x_{i,p}(L_p+P^L_p) \tag{2.11}$$

while the energy balance yields

$$Q_p + h_{FP}F_p + H_{p+1}V_{p+1} + h_{p-1}L_{p-1} = H_p(V_p+P^V_p) + h_p(L_p+P^L_p) \tag{2.12}$$

where the enthalpies are functions of π_p, temperature and compositions of the relevant phase:

$$\begin{aligned} h_p &= h(\pi_p,\ T_p,\ x_{i,p}) \\ H_p &= H(\pi_p,\ T_p,\ y_{i,p}) \end{aligned} \tag{2.13}$$

We also have by definition of the mole fractions

$$\sum_{i=1}^{c} z_{i,p} = \sum_{i=1}^{c} x_{i,p} = \sum_{i=1}^{c} y_{i,p} = 1 \qquad (2.14)$$

where $z_{i,p}$ molal feed composition

Summing (2.8) and (2.11) over all plates in the column we obtain the overall balances:

$$\begin{aligned} &\sum_{p\varepsilon P} (F_p - P_p^P - P_p^V) = 0 \\ &\sum_{p\varepsilon P} (Q_p + h_{FP}F_p - h_pP_p^P - H_pP_p^V) = 0 \end{aligned} \qquad (2.15)$$

where P is the set of plates with non zero streams of material or energy to the exterior.

To specify the operating conditions, the state of each product stream, and all but one product flow rate and one heat exchanger rante much be specified, these two quantities being computed from (2.15) to assume a steady state.

From Equation (2.8)

$$L_p = F_p + V_{p+1} + L_{p-1} - (V_p + P_p^V) - P_P^L \qquad (2.16)$$

replacing Equation (2.16) in Equation (2.12) we obtain

$$\begin{aligned} &Q_p + h_{FP}F_p + H_{p+1}V_{p+1} + h_{p-1}L_{p-1} = \\ &\qquad = H_p(V_p+P_p^V) + h_p(P_p^L+F_p+V_{p+1}+L_{p-1}-P_p^L-V_p-P_p^V) \end{aligned}$$

$$Q_p + F_p(h_{Fp}-h_p) + L_{p-1}(h_{p-1}-h_p) =$$

$$= -H_{p+1}V_{p+1} + V_p(H_p-h_p) + P_p^V(H_p-h_p) + V_{p+1}h_p$$

$$V_{p+1} = \frac{1}{(H_{p+1}-h_p)} \left[V_p(H_p-h_p)+P_p^V(H_p-h_p)-Q_p-F_p(h_{FP}-h_p)-L_{p-1}(h_{p-1}-h_p)\right] \tag{2.17}$$

For given values of enthalpies, obtained using Equation (2.13), Equations (2. 8) and (2.12) can be used alternately to generate all internal flow rates, L_p and V_p.

Using Equations (2.5) and (2.11) we obtain

$$z_{i,p}F_p+x_{i,p+1}K_{i,p+1}V_{p+1}+x_{i,p-1}L_{p-1}=x_{i,p}K_{i,p}(V_p+P_p^V)+x_{i,p}(L_p+P_p^L)$$

$$-x_{i,p-1}L_{p-1}+x_{i,p}(L_p+P_p^L+K_{i,p}(V_p+P_p^V))-x_{i,p+1}K_{i,p+1}V_{p+1}=F_pz_{i,p} \tag{2.18}$$

Each set of equations for each component can be solved for the values of liquid compositions if the internal flow rates and equilibrium ratios are known. In general the $K_{i,p}$ are dependent upon compositions as well as temperature and pressure as shown in Equations (2.6) and (2.7), making Equation (2.17) nonlinear.

If the equilibrium ratios depend weakly upon compositions, it is possible to remove the nonlinearity by evaluating the $K_{i,p}$ for the compositions obtained in the previous iteration.

The component mass balance equations may be written as

$$G^i x^i = f^i \qquad (2.19)$$

where G^i is a tridiagonal matrix whose non-zero elements are:

$$g_{i,p-1} = -L_{p-1}$$

$$g_{i,p} = L_p + P_p^L + K_{i,p} (V_p + P_p^V) \qquad (2.20)$$

$$g_{i,p+1} = -K_{i,p+1} V_{p+1}$$

Usually the equilibrium ratios and internal flow rates are unknown, and we shall have to iterate on the values of these variables. This will result in Equation (2.19) being a set of linear equations to be solved within each iteration for each component. The Thomas method with partial pivoting was used to solve the tridiagonal matrix and it is described in Appendix A.

Equation (2.19) has to be solved for each component in turn, obtaining $x_{i,p}$ for all i and p, and then from Equation (2.5) $y_{i,p}$ for all i and p can be calculated. However, unless the $K_{i,p}$ were consistent with these compositions and temperatures, the computed compositions do not sum to unity on each plate.

Amundson and Pontinen (2) normalized the liquid composition on each plate by dividing each composition by the composition sum on that plate. The $K_{i,p}$, then, had to be chosen to satisfy the following equation:

$$\sum_{i=1}^{c} K_{i,p} x_{i,p} = 1 \text{ , } p = 1,2, \ldots \qquad (2.21)$$

If the $K_{i,p}$ are functions only of the temperature and pressure, Equation (2.21) can be used to correct temperatures. Amundson and Pontinen treated the temperatures as noninteracting variables and corrected them using Newton's method applied to Equation (2.21). This process diverges in general if the liquid compositions are not scaled.

The inherent separation factor $\alpha_{i,j}$ in a vapour-liquid system is commonly called the relative volatility. The reasons for this name are apparent from Equation (2.25), where for an ideal system $\alpha_{i,j}$ is simply the ratio of the vapour pressures of i and j.

$$\alpha_{i,j} = \frac{y_i/x_i}{y_j/x_j} \tag{2.22}$$

If the components of the mixture obey Raoult's and Dalton's law,

$$p_i = Py_i = P_i^V x_i \tag{2.23}$$

where P total pressure

P_i^V vapour pressure of pure liquid i at the temperature of the mixture.

In such a case

$$K_i = \frac{y_i}{x_i} = \frac{P_i^V}{P} \tag{2.24}$$

and

$$\alpha_{i,j} = \frac{P_i^V}{P_j^V} = \frac{K_i}{K_j} \tag{2.25}$$

The vapour pressures of i and j depend upon temperature. Since

$\alpha_{i,j}$ is proportional to the ratio of vapour pressures, and since both vapour pressures increase with increasing temperature, $\alpha_{i,j}$ will be less sensitive to temperature than k_i and k_j. Over short ranges of temperature, $\alpha_{i,j}$ can often be taken to be a constant.

The selection of a base component can be done by choosing one of the components of the mixture or defining a hypothetical component. If the mixture contains a wide range of boiling temperatures, then a middle component should be taken as the reference component.

For the base component $K_b = y_b/x_b$ and $\alpha_b = 1.0$. Expressions needed for the calculation of the bubble point and dew point temperatures are developed in the following manner. For any component i,

$$\frac{y_i}{y_b} = \frac{K_i\ x_i}{K_b\ x_b} \qquad (2.26)$$

which can be rearranged to

$$\frac{y_i}{K_b} = \alpha_{i,b}\ x_i \qquad (2.27)$$

Summation of both sides of the expressions over all components yields

$$\frac{1}{K_b} = \sum_{1=j}^{c} \alpha_{i,b}\ x_i \quad \text{or} \quad K_b = \frac{1}{\sum_{i=1}^{c} \alpha_{i,b} x_i} \qquad (2.28)$$

where c is the number of components present in the mixture.

Elimination of K_b from Equations (25) and (26) gives

$$y_i = \frac{\alpha_{i,b}\, x_i}{\sum_{i=1}^{c} \alpha_{i,b} x_i} \qquad (2.29)$$

In an analogous manner it is readily shown that

$$K_b = \sum_{i=1}^{c} y_i/\alpha_{i,b} \qquad (2.30)$$

and that

$$x_i = \frac{y_i/\alpha_{i,b}}{\sum_{i=1}^{c} y_i/\alpha_{i,b}} \qquad (2.31)$$

In developing a simple K-value model, the dependence of K values on temperatures could be represented by a model for the form:

$$\ell_n\, K_{bp} = Ab_p + \frac{Bb_p}{T} \qquad (2.32)$$

where p is the ideal stage number and Ab_p and Bb_p are the Antoine constant for the reference component.

In the inside-out approach, Ap and Bp are the iteration variables in the outside loop.

Supposing that the liquid compositions are obtained from Equation (2.19) and then normalized, the equilibrium ratio for the reference component from Equation (2.28), the vapour compositions from Equation (2.29), the temperatures from Equation (2.32), and the enthalpies from Equations (2.13), then the internal flow rates can be calculated from Equations (2.16), (2.17), (2.9) and (2.10).
The equilibrium ratios can be obtained from a property prediction package or simple equations like the Antoine equation and the relative volatilities from Equation (2.25). The liquid compositions can be calculated

again from Equation (2.19) with the updated values for the equilibrium constant and internal flow rates.

In this algorithm, the vapour composition will not satisfy Equation (2.14) until a solution of the system is found, because unless the estimated equilibrium ratios are all correct, the calculated vapour mole fraction of the different components on a given stage will not add to unity. The convergence rate using direct substitution on the K_b could be very slow in some cases;an acceleration technique like the Wegstein method (35) will improve the efficiency in these cases, as it will be seen in some of the results.

2.4 Method of Solution

Interstage flows are linked together by the overall mass balance in such a way that if vapour flow increases in a certain direction through the section, the liquid flow will also increase in the same direction. Since the fractionation is mainly dependent on the ration L/V, considerable changes can occur in flows without greatly disturbing L/V, and hence the separation.

For the purposes of finding an optional configuration of distillation columns in the early stages of design, constant molar flows were therefore assumed. The changes at feed and products that correspond to intermediate plates in a given column are easily dealt with in terms of "q-factors". This simplification eliminates the need for heat balances and enthalpy determination. There will then be no need to determine temperatures in each ideal stage, temperatures will only be needed in the places in which heat exchange takes place.

The feed q^F factor is defined by equation

$$q_F = \frac{h_{Fp}-h_p}{H_p-h_p} \qquad (2.33)$$

The liquid product q^L factor is defined by

$$q_p^L = \Delta V_p / P_p^L \tag{2.34}$$

where $Q_p = \Delta V_p \; (h_p - H_p)$ (2.35)

and the vapour product q^V is defined as

$$q_p^V = \Delta L_p / P_p^V \tag{2.36}$$

and $$Q_p = \Delta L_p \; (H_p - h_p)$$

$$q_p^L = q_p^V = 0 \quad \text{correspond to } Q_p = 0$$

Equations (12) and (13), then, reduce to

$$L_p = L_{p-1} + F_p(1-q_p^F) - (1-q_p^L)\, P_p^L - q_p^V\, P_p^V \tag{2.37}$$

$$V_{p+1} = V_p - F_p q_p^F + (1-q_p^V)\, P_p^V + q_p^L\, P_p^L \tag{2.38}$$

and the overall balance equations (11) reduce to

$$\begin{aligned} &\sum_{p \varepsilon P} (F_p - P_p^L - P_p^V) = 0 \\ &\sum_{p \varepsilon P} (F q_p^F - P_p^V - P_p^L q_p^L) = 0 \end{aligned} \tag{2.39}$$

The summation equations can be written as

$$\sum_{i=1}^{c} x_{i,p} - 1 = S_p \qquad (2.40)$$

The algorithm used to solve the simplified model follows.

2.4a Simplified Algorithm

Given all the F_p, q_p^F and $Z_{i,p}$

all the q_p^L and q_p^V except one

all the P_p^V and P_p^L except one of them

the relative volatilities and the number of plates in each section of the column, then

Step 1 - Compute the remaining product rate and heat exchanged from Equation (2.39)

Step 2 - Starting at the bottom of the column and using alternately Equations (2.37) and (2.38), compute V_p and L_p upwards through the column and use Equations (2.9) and (2.10) to calculate V_1, L_0, L_m, V_{m+1}

Step 3 - Compute the equilibrium ratio for the reference component from Equation (2.28) using the feed composition as the liquid compositions and assume that the equilibrium ratios for this compnent in all the plates are the same

Step 4 - Solve Equations (2.19) for each component to give all the $x_{i,p}$, then compute S_p from Equation (2.40)

Step 5 - If $|S_p| < \varepsilon$ for all p, STOP, otherwise normalize the $x_{i,p}$ and go to step 6

Step 6 - Adjust K_{bp} using Equation (2.28) for each plate. A secant method to accelerate the convergence of the Direct Substituion method can be used in this step. Go to step 4.

The accelaration of the Direct Substitution method will be described in Appendix C.

2.5 Numerical Examples

The acceleration of the direct substitution method is analyzed in Appendix C. To avoid instabilities in the application of the Wegstein method some bounds on the extrapolation can be used or otherwise the first acceleration step is used with some delay, for example after a few iterations using direct substitution steps only, and from then on, using the acceleration method every n direct substitution steps.

Some examples will be run to see whether the Wegstein method improves the convergence of the direct substitution method and under which conditions. Two binary mixtures and three ternary mixtures will each be separated into their components in a single column, at atmospheric pressure. The binary cases, of a column with two liquid products will be run with different composition of the feed. For the ternary mixtures, the three products were liquid. The feed in all the cases was 100 lb mole/hr as a liquid at its bubble point.

The nomenclature used was:

NT	total number of ideal stages in the column
NF	plate number where the feed is introduced

NI	plate number where the intermediate product is obtained from
R	reflux ratio defined as the amount of liquid being recycled over the liquid product at the top of the column
D	top product molal flow rate (lb mole/hr)
PH	intermediate product molal flow rate (lb mole/hr)
X_F	molal composition of the feed
α	relative volatilities
it	number of iterations
minws	first iteration number in which the acceleration method was used
(nac-1)	number of direct substitution steps used between the acceleration steps
error	tolerance error
time	seconds (CDC 6500)

Example Ia

NT	NF	R	D	error
21	9	1.4733	30.0	0.0001

	X_F	α	product purity
benzene	0.3	2.8	0.899
toluene	0.7	1.0	0.957

minws	nac	it	time
no acceleration		15	0.1695
14	6	18	0.2055
10	6	20	0.2355

Example Ib

NT	NF	R	D	error
17	8	0.71	50.0	0.0001

	X_F	α	product purity
benzene	0.5	2.8	0.900
toluene	0.5	1.0	0.900

minws	nac	it	time
no acceleration		14	0.1305
10	4	14	0.1320

Example Ic

NT	NF	R	D	error
18	9	0.56	70.0	0.001

	X_F	α	product purity
benzene	0.7	2.8	0.957
toluene	0.3	1.0	0.901

minws	nac	it	time
no acceleration		13	0.126
10	4	16	0.170

Example IIa

NT	NF	R	D	error
31	14	3.55	30.0	0.0001

	X_F	α	product purity
chloroform	0.3	1.8	0.899
carbon tetrachloride	0.7	1.0	0.957

minws	nac	it	time
no acceleration		11	0.1800
8	4	12	0.2020
10	6	12	0.1980
6	6	13	0.2175

Example IIb

NT	NF	R	D	error
26	12	1.8676	50.0	0.0001

	X_f	α	product purity
chloroform	0.5	1.8	0.899
carbon tetrachloride	0.5	1.0	0.899

minws	nac	it	time
no acceleration		11	0.1530
10	6	11	0.1590
6	6	11	0.1610
6	4	11	0.1695
4	4	16	0.2250

Example IIc

NT	NF	R	D	error
29	15	1.455	70.0	0.0001

	X_F	α	product purity
chloroform	0.7	1.8	0.958
carbon tetrachloride	0.3	1.0	0.901

minws	nac	it	time
no acceleration		13	0.2010
10	6	13	0.2100
6	4	10	0.1575
4	4	14	0.2220

Example IId

NT	NF	R	D	error
19	9	2.092	50.0	0.0001

	X_F	α	product purity
chloroform	0.5	1.8	0.899
carbon tetrachloride	0.5	1.0	0.899

minws	nac	it	time
no acceleration		10	0.1020
6	4	10	0.1035
4	4	8	0.0855

Example III

NT	NF	NI	R	D	PH
30	21	13	6.807	30.0	40.0

	X_F	α	product purity
dichloromethane	0.3	4.9	0.901
chloroform	0.4	1.8	0.904
carbon tetrachloride	0.3	1.0	0.972

minws	nac	it	time	error
no acceleration		46	1.098	0.001
18	6	34	0.855	0.001
no acceleration		69	1.6170	0.0001
18	6	38	0.8940	0.0001
14	6	37	0.8595	0.0001
10	6	39	0.9345	0.0001

Example IV

NT	NF	NI	R	D	PH
27	19	12	10.2	20.0	40.0

	X_F	α	product purity
dichloromethane	0.2	4.9	0.900
chloroform	0.4	1.8	0.900
carbon tetrachloride	0.4	1.0	0.950

minws	nac	it	time	error
no acceleration		34	0.8685	0.001
18	6	26	0.5820	0.001
14	6	22	0.4815	0.001
10	6	25	0.5535	0.001
no acceleration		44	0.9810	0.0001
18	6	31	0.7560	0.0001
14	6	27	0.5805	0.0001

Example V

NT	NF	NI	R	D	PH
24	19	15	6.23	30.0	40.0

	X_F	α	product purity
benzene	0.3	6.7	0.898
toluene	0.4	2.4	0.888
o-xylene	0.3	1.0	0.953

minws	nac	it	time	error
no acceleration		41	0.7635	0.001
18	6	41	0.7680	0.001
10	6	38	0.7095	0.001
10	4	29	0.5595	0.001
8	6	30	0.5655	0.001
8	4	38	0.7095	0.001
no acceleration		56	1.014	0.0001
18	6	47	0.885	
10	6	42	0.789	

2.6 Comments

In the first six examples, dealing with a distillation column with two products to separate a binary mixture into its components, the rate of convergence of the direct substitution method was good. In the mixture composed of benzene and toluene the relative volatilities were (2.8, 1.0) while in the second case the mixture of chloroform and carbon tetrachloride has smaller relative volatilities (1.8, 1.0). More ideal stages are required in the second mixture; thus the system of equations to be solved is bigger and more time is required to obtain the solution but the iterations required to converge using direct substitution do not increase with the size of the system, as can be seen comparing the iterations required for both mixtures with the same feed composition, as in cases Ic and IIc. An increase of 38 percent in the number of plates corresponds to an increase of 37 percent in the computing time used, for the same number (13) of iterations. Comparing examples IIb and IId, with the same feed but different ideal stages and reflux ratio in the column, in both cases to obtain 90 percent purity in both products, it can be observed that an increase of 33.3 percent in the number of ideal stages corresponds to an increase of 34.5 percent in the computing time. The number of iterations are nearly the same, eleven and ten respectively. The use of the Wegstein method did not improve the convergence in most of the cases, as can be seen in examples Ib, IIb and IId and is likely to make things worse as is the case in examples Ia, Ic and IIa.

For the last three examples, in which a ternary mixture is fed in the column, the convergence of the direct substitution method is quite slow, requiring from 44 iterations in example IV to 70 iterations in example III, for the same error tolerance as in the previous examples. In these cases the improvement achieved using the Wegstein method to accelerate convergence is not negligible, provided that some delay is introduced before the use of the acceleration procedure and that the unbounded secant step is used every "nac" direct substitution steps.

In the third example, the computing time was reduced by 47 percent of the original computing time required using DS only. DS was applied for the first fourteen iterations, followed by Wegstein acceleration every sixth iteration thereafter.

CHAPTER III

SIMULATION OF A SERIES OF DISTILLATION COLUMNS

3.1 Introduction

The simulation of a set of distillation columns can be done either by solving the equations for all the columns simultaneously or, alternatively, solving each column separately as a unit and iterating on the values of the recycle streams being torn. If the first approach is followed, the tridiagonal matrices for the component mass balances or the block band matrices in the simultaneous solution of all the linearized equations, will be filled in with elements corresponding to the interconnecting streams. Bending and Hutchinson (20) developed a Gaussian elimination method for sparse sets of linear equations which consisted of two stages, triangularization and back substitution and two programs were written. The primary equation routine solves the equations using only the operator list and the updated list of element values. This operator list method can save a significant amount of time if the matrix is sparse as the secondary linear equation routine does not need to search for elements which probably do not exist. This method was used by Shewchuk and Hutchinson (14) for multiple distillation towers.

Some efforts have recently been made to use modified Gaussian elimination to solve band matrices having some off-ban submatrices by Kuriček, Hlavácek and Procháska (21), Browne, Ishii and Otto (22), Holefing and Seader (23) and Hidalgo, Correa and Seader (24). The minimization of the number of non-zero off-band elements in the triangularization as proposed by Hidalgo <u>et al</u> (24) is time consuming, though storage is saved if all non-zero elements of the matrix are stored in a linear vector.

The key for an efficient simultaneous solution of all the equations of the system is an efficient handling of a set of sparse algebraic equations. In the context of quasi-Newton methods and a set of nonlinear equations with a sparse Jacobian matrix, Shubert (25) found that the updating procedure can be made more efficiently for the approximate Jacobian than for the inverse of the Jacobian where the sparsity is destroyed. If the constant elements in the Jacobian are known, they will remain unchanged in the updating procedure. Curtis, Powell and Reid (26) showed how to use known constant elements in the Jacobian matrix to reduce the work required to estimate the remaining elements by finite differences.

The advantages and disadvantages of solving simultaneously all the equations have been discussed in the previous chapter. For the general configurations described in Chapter V, the storage requirement for the Jacobian in the simultaneous solution would be excessively large. The possibility of solving the general system simultaneously will depend on the development of powerful sparse matrix procedures and on the robustness of the algorithm used to solve the nonlinear equations. At present, research is being done in both areas. It is likely that in the near future with an increased capacity of the computers, the simultaneous approach will compete favourably with the modular approach.

The modular approach was used in this work for the simulation of a system of distillation columns, the main advantages of this approach being the simplicity and robustness of the direct substitution method in dealing with arbitrary initial guesses. However, sometimes the convergence is very slow, as expected in the DS method and some acceleration procedure is needed to improve the rate of convergence, as has been seen in the previous chapter.

In the next section the general configuration for a ternary mixture being separated into its components will be analyzed. The use of the direct substitution approach to update the torn variables will be compared with the Broyden method.

3.2 General Configuration for a Ternary Mixture

The general configuration for a ternary mixture in which all the final products are liquids is shown in Figure 3.1. All the flow rates are expressed in moles, where

L(J) and V(J)	are the liquid and vapour internal flow rates in the first column
PL(J) and PV(J)	are the liquid and vapour product flow rates in the first column
L2(J) and V2(J)	are the liquid and vapour internal flow rates in the second column
PL2(J) and PV2(J)	are the liquid and vapour product flow rates in the second column
N1	represents the ideal stages above the feed in the first column
N2	represents the ideal stages below the feed in the first column

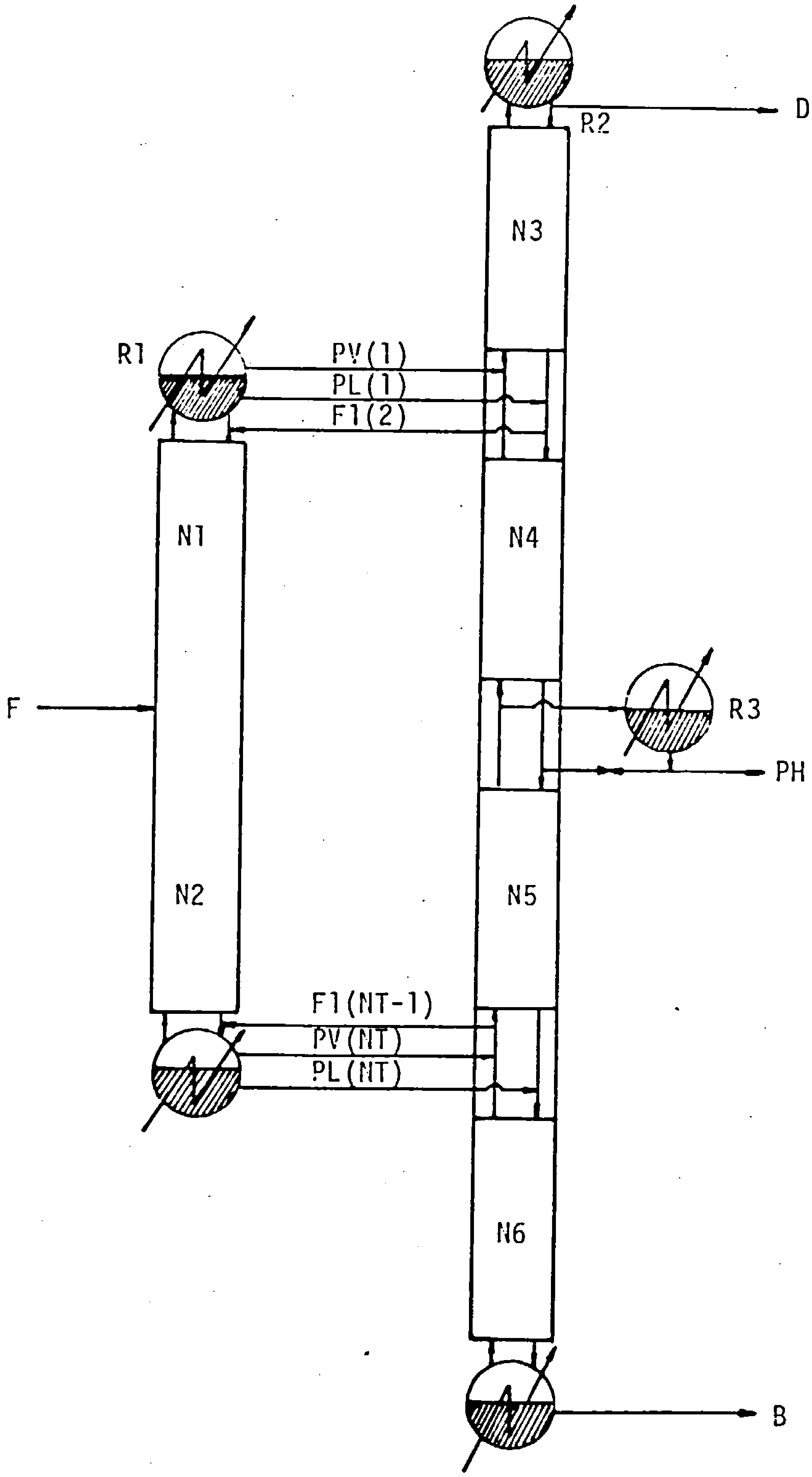

Figure 3.1

N3	represents the ideal stages in the top section of the second column
N4	represents the ideal stages in the intermediate section of the second column, above the extraction of the intermediate final product
N5	represents the ideal stages in the intermediate section of the second column, below the intermediate heat exchanger
N6	represents the ideal stages in the bottom section of the second column
F	feed flow rate to the system
PV(1)	vapour top product of the first column
PL(1)	liquid top product of the first column
F1(2)	liquid feed from the second column into the first column
PV(NT)	vapour bottom product of the first column
PL(NT)	liquid bottom product of the first column
F1(NT-1)	vapour feed from the second column into the first column

D	liquid flow rate of the lightest product
PH	liquid flow rate of the intermediate product
B	liquid flow rate of the heaviest product
R1	defined as the reflux ratio of the liquid returned to the top of the first column over the total product flow rate at the top of the column $R1 = L(1)/(PV(1)+PL(1)) \qquad (3.1)$
R2	defined as the reflux ratio of the liquid returned to the top of the second column over the top product $R2 = L2(1)/D \qquad (3.2)$
R3	is defined as follows $R3 = (PV2(N3+N4+1) - PL2(N3+N4))/PH \qquad (3.3)$

Dropping the sub index in PV and PL, if

I - $PV > PH$ and $PL = 0$ then $R3 > 1$ and a condenser is needed

II - $PV = PH$ and $PL = 0$ then $R3 = 1$ and a condenser is needed

III - $PV < PH$, $PL > 0$ and $PV > PL$ then $0 < R3 < 1$ and a condenser is required

IV - $PV = PL$, $PL > 0$ and $PV > 0$ then $R3 = 0$ and a condenser is needed

V - $PV < PL$, $PL > 0$ and $PV > 0$ then $-1 < R3 < 0$ and a condenser is needed

VI - $PV = 0$ and $PL = PH$ then $R3 = -1$ and no heat exchanger is needed

VII - $PV = 0$ and $PL > PH$ then $R3 < -1$ and a reboiler is needed

In the first case, more vapour than the amount needed to provide for the intermediate liquid product PH is condensed and recycled as reflux into the column. In the second case, an amount of vapour equivalent to PH is condensed. In the third, fourth and fifth cases the liquid product, PH, is made up of condensed vapour and liquid in different proportions. The liquid product is obtained directly from the internal liquid flow in the sixth case. More liquid is extracted from the column than needed for PH in the seventh case, thus some liquid is vaporized and returned to the column.

The degrees of freedom of this system will be obtained in the next section.

3.3 Degrees of Freedom

To solve a set of simultaneous equations, a sufficient number of variables must be specified for the number of remaining unknown variables to be exactly equal to the number of independent equations. The unknown variables to which we assign values are the design variables. If there are m variables and n independent equations, the degrees of freedom of the system are (m-n) and (m-n) values have to be specified for the design variables.

In order to describe a separation process uniquely, the number of variables which must be specified is equal to the number which can be set by construction or controlled during the operation by external means.

Analyzing each column of the general configuration in turn, we can find the degrees of freedom of the system.

In Figure (3.2) the first column can be seen.

If we assumed that all the feeds to the first column are known, the number of plates above and below the feed F are set during construction. The column can be operated at an arbitrary chosen pressure between certain limits. An arbitrary amount of heat can be introduced in the reboiler and an arbitrary amount of heat, between limits, can be removed from the condenser. The liquid leaving the condenser can be split in any desired ratio by adjusting the valve in the reflux line and the same is true for the vapour leaving the reboiler.

Thus, after all the feeds have been set, seven more variables must be defined. The seven variables mentioned were:

$$N1,\ N2,\ p_1,\ Q_c,\ Q_R,\ r_c,\ r_R$$

where p_1 is the pressure in the column, Q_c and Q_R are the heat exchanged in the condenser and reboiler and r_c and r_R are the split ratio of the liquid and vapour, condensed and boiled in both exchangers. Instead of Q_c, Q_R, r_c and r_R, the design variables specified were:

PV(1), PL(1), PV(NT) and R1

The second column is shown in Figure (3.3). Assuming that all the feeds to the column are known, four more variables than in the

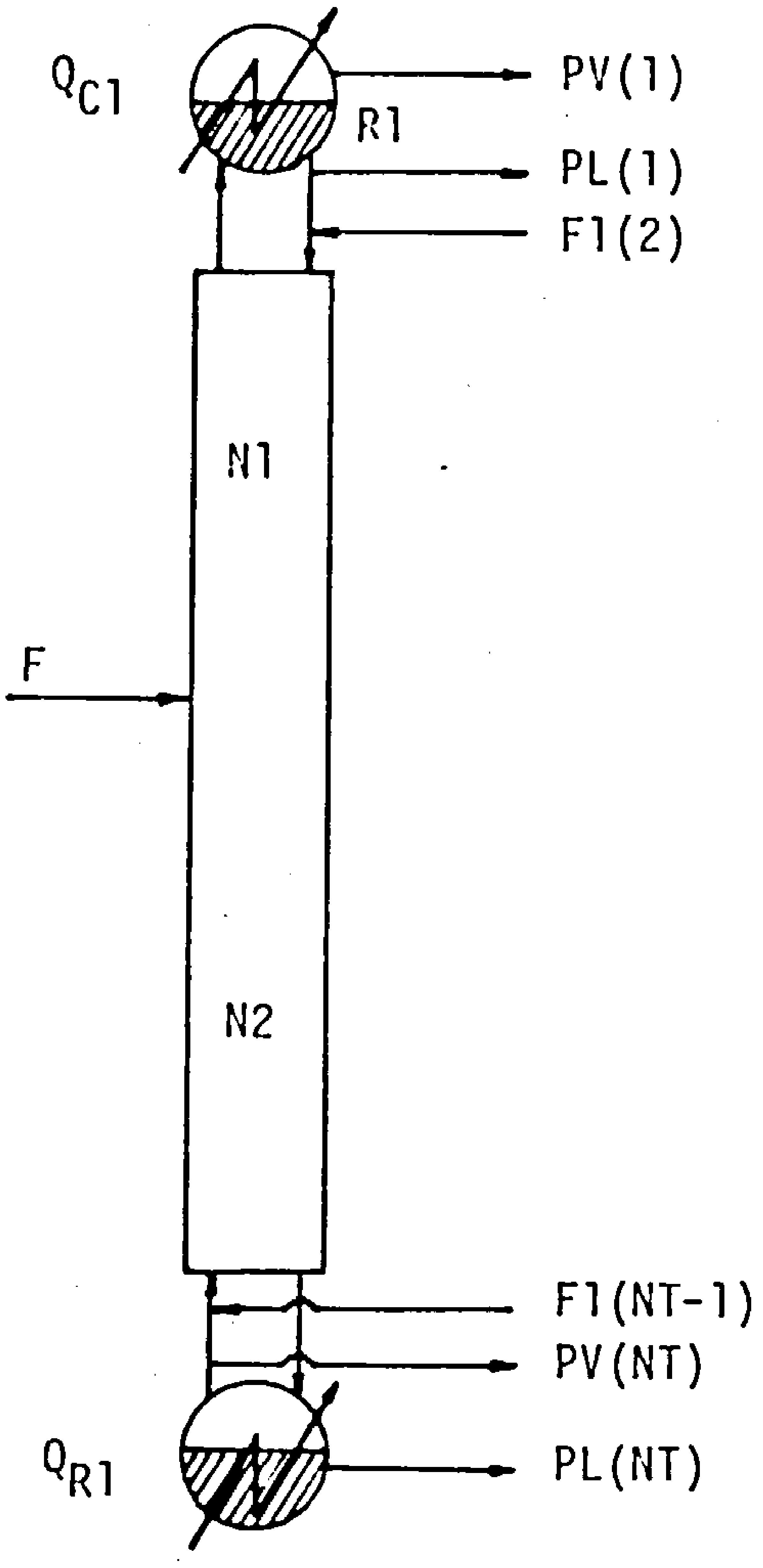

Figure 3.2

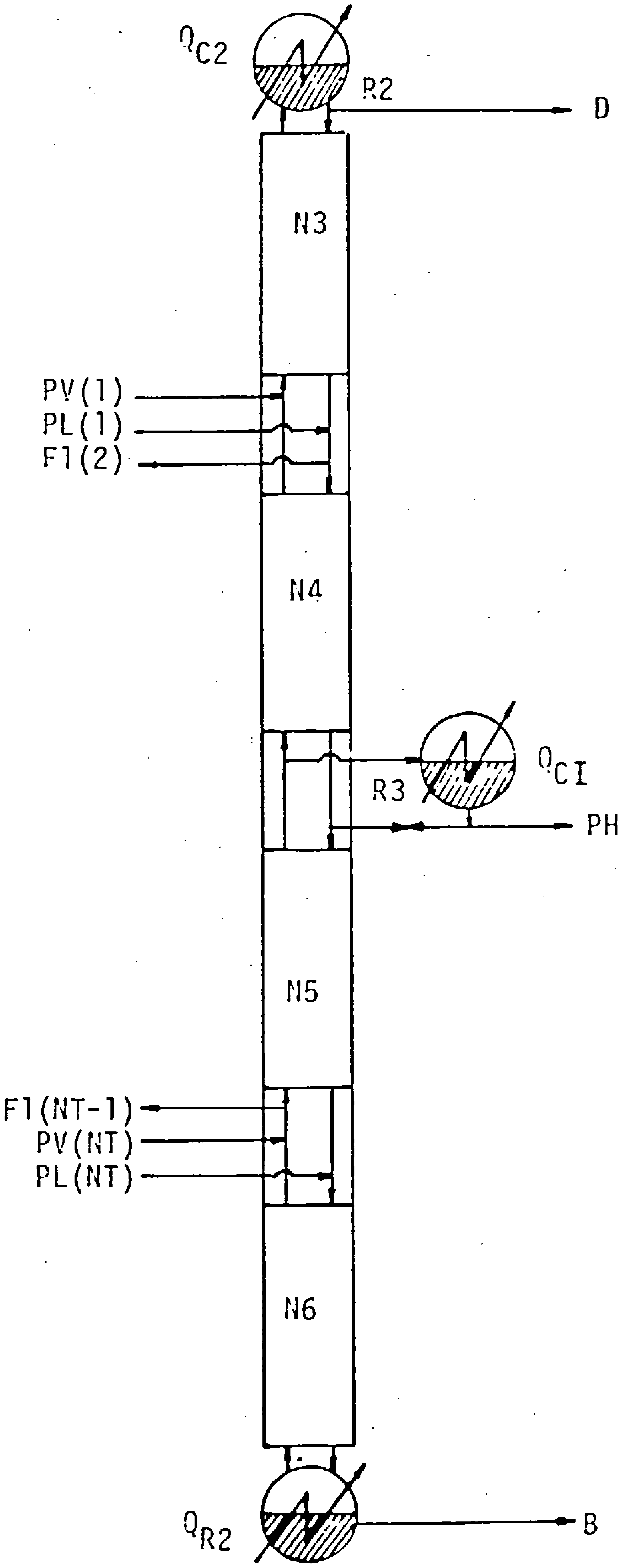

Figure 3.3

previous column can be regulated, thus increasing the degrees of freedom to eleven. The design variables chosen were:

N3, N4, N5, N6, p_2, D, PH, R2, R3, F1(2), F1(NT-1)

where p_2 is the pressure in this column.

Thus the number of degrees of freedom of the system are eighteen.

3.4 Simulation of the System

Assuming that the above eighteen design variables have been specified, the system is well determined.

Each column is solved separately and the recycle stream variables must be guessed and iterated on. To simulate the first column, the variables of the streams being fed into the first column should be specified. As the recycle stream variables are now known, some guessed values should be assigned to these variables and then updated. Enthalpy balances were not included in the simulation of a single column and will not be included in the simulation of the general configuration, therefore there is no need to calculate temperatures and enthalpies. There are two recycle streams, a liquid feed at the top and a vapour feed at the bottom of the first column. The flow rates of these two recycle streams were chosen as design variables, so that the only unknown variables are the compositions of these streams. Two component fractions in each stream could be guessed and updated and the third one calculated to satisfy that the summation of all the mole fractions add to unity. Four mole component fractions need to be guessed initially to define completely the feeds to the first column. The stage numbers were numbered from the top to the bottom of the columns.

Setting

$$NW1 = N3 \tag{3.4}$$

$$NW3 = N3+N4+N5+1 \tag{3.5}$$

the four torn variables are the liquid fractions

$$X2(1,NW1) \quad , \quad X2(2,NW1)$$

and the vapour fractions

$$Y2(2,NW3) \quad , \quad Y2(3,NW3)$$

The summation equations were used to obtain the two remaining mole fractions:

$$X2(3,NW1) = 1 - X2(1,NW1) - X2(2,NW1) \tag{3.6}$$

$$Y2(1,NW3) = 1 - Y2(2,NW3) - Y2(3,NW3) \tag{3.7}$$

A straightforward way of calculating the values of the recycle variables would be the direct substitution method. After the first simulation of the first column, the composition of the four products from the first column being fed to the second column are known and a simulation of the second column can be carried out, obtaining new values for the compositions of the recycle streams which are products from the second column. The values of the unknown recycle streams are guessed initially, the sequence of distillation columns computed through, and a new set of values for the recycle stream compositions obtained. These are compared with the values of the torn variables in the previous iteration. If they differ by less than some prescribed tolerance, the problem is solved, otherwise the cycle is repeated.

In the general configuration, capable of separating a given feed into its components, the number of torn variables can be calculated as follows:

$$n = (c-1) \sum_{i=2}^{c-1} i \tag{3.8}$$

where c is the number of components present in the feed.

3.5 Numerical Results

Particular cases of the general configuration shown for a ternary mixture are simulated in the following examples. In each case the direct substitution method is used to update the value of the torn variables. The maximum eigenvalue of the system of torn variables is estimated around the fifth iteration, using Equation C6 of Appendix C. This value (λ_{max}^{app}) is used to predict the number of direct substitution steps needed to find the solution using equation B12 of Appendix B, and checked with the number of DS steps really required to find the solution. In each example the value of the biggest eigenvalue of the system calculated at the solution using Equation C6 is reported as λ max.

Broyden's method was also used to update the compositions of the recycle streams, in the same examples as those run using the direct substitution method, to check the robustness and efficiency of both methods. Different initial guesses for the torn variables and different numbers of maximum inner iterations for the simulation of each column were tried.

A ternary mixture, described in each example, is separated into its components. Both columns are run at atmospheric pressure and the flow rate of the feed is 100 lb moles/hr in all the cases. The nomenclature used follows:

maxdis	maximum number of iterations allowed for convergence of each column simulation
it	number of iterations required to find the solution value of the torn variables
time	seconds (CDC 6500)
Z_o	initial guesses for the torn variables
Z_{sol}	solution value of the torn variables
$X_{i,F}$	molal feed compositions
$\alpha_{i,R}$	relative volatilities
minws	first iteration, in the inner loop, in which the Wegstein method is used
(nac-1)	number of direct substitution steps used between the acceleration steps.

Example I

X_F = 0.3, 0.4, 0.3 $\alpha_{i,R}$ = 4.9, 1.8, 1.0

N1	N2	N3	N4	N5	N6	R1	R2	R3	PV(1)	PL(1)	F1(2)	PV(NT)	F1(NT-1)
8	9	7	7	14	12	0.466	1.13	1.0	28.0	13.0	0	0	60.0

Z_{sol} / Y2(2,NW3) = 0.734 , Y2(3,NW3) = 0.243

Table 3.1a Direct Substitution

Changing the maximum iteration number in the column simulation

Z_o		maxdis	it	time	minws	nac	case
0.4	0.55	20	13	3.305	14	4	1
0.4	0.55	10	13	2.622	no acceleration		2
0.4	0.55	5	13	1.747	no acceleration		3
0.4	0.55	2	17	1.063	no acceleration		4
0.4	0.55	1	30	1.051	no acceleration		5

Table 3.1b Direct Substitution

Different initial guesses for the torn variables

Z_o		maxdis	it	time	minws	nac	case
0.2	0.7	5	14	1.89	no acceleration		6
0.333	0.333	5	13	1.835	no acceleration		7
0.6	0.2	5	13	1.751	no acceleration		8
0.2	0.1	5	14	1.969	no acceleration		9

Table 3.1c Broyden Method

Changing the maximum number of iterations in the column simulation

Z_o	maxdis	it	time	minws	nac	case
0.4 0.55	20	12	2.921	14	4	10
	10	15	2.699	no acceleration		11
	5	16	1.975	no acceleration		12
0.4 0.55	3	18	1.470	no acceleration		13
0.4 0.55	2	40	2.175	no acceleration		14
0.4 0.55	1	8	bounds impeding progress			15

Table 3.1d Broyden Method

Different initial guesses for the torn variables

Z_o	maxdis	it	time	case
0.2 0.7	5	22	2.605	15
0.333 0.333	5	23	2.589	16
0.6 0.2	5	17	1.965	17
0.2 0.1	5	26	2.789	18
0.2 0.1	2	bounds impeding progress		19
0.2 0.1	3	bounds impeding progress		20
0.6 0.2	3	19	1.494	21
0.333 0.333	3	38	2.833	22
0.2 0.7	3	bounds impeding progress		23

Table 3.1e Broyden Method

Z_o		maxdis	it	time	minws	nac	case
0.4	0.55	20	12	2.921	no acceleration		24
0.4	0.55	20	14	2.847	6	4	25
0.4	0.55	20	14	2.584	10	4	26

Example II

This is nearly the same as the first example, except that two design variables, the reflux ratios R2 and R3 have been changed, where R2 = 2.0 and R3 = 1.5.

Z_{sol} : Y1(2,NW3) = 0.737 , Y1(3,NW3) = 0.247

Table 3.2a Changing the maximum iteration number in the column simulation

Direct Substitution is used to update the torn variables

Z_0		minws	nac	maxdis	it	time	case
0.4	0.55	14	4	20	35	9.266	1
0.4	0.55	no acceleration		10	35	7.433	2
0.4	0.55	no acceleration		5	38	4.832	3
0.4	0.55	no acceleration		2	43	2.610	4
0.4	0.55	no acceleration		1	45	1.507	5

Table 3.2b Different initial guesses for the torn variables

Z_0		minws	nac	maxdis	it	time	case
0.2	0.7	no acceleration		10	37	7.804	6
0.2	0.7	no acceleration		5	40	5.030	7
0.333	0.333	no acceleration		5	23	2.955	8
0.6	0.2	no acceleration		5	22	2.777	9
0.2	0.1	no acceleration		5	24	3.089	10
0.2	0.7	no acceleration		1	48	1.612	11
0.333	0.333	no acceleration		1	47	1.590	12
0.6	0.2	no acceleration		1	46	1.571	13

Table 3.2c Different maximum number of iterations in each column simulation

Z_o		minws	nac	maxdis	it	time	case
0.40	0.55	14	4	20	15	4.017	1
0.40	0.55	no acceleration		10	19	3.488	2
0.40	0.55	no acceleration		5	32	3.965	3
0.40	0.55	no acceleration		2	80(*)	4.425	4

Table 3.2d Different initial values for the torn variables

Z_o		minws	nac	maxdis	it	time	case
0.200	0.100	no acceleration		5	60	bounds impeding progress	5
0.600	0.200	no acceleration		5	26	bounds impeding progress	6
0.333	0.333	no acceleration		5	41	bounds impeding progress	7
0.200	0.700	no acceleration		5	36	bounds impeding progress	8

(*) did not converge

Example III

X_F = 0.3, 0.4, 0.3 α = 6.35, 2.47, 1.0

N1	N2	N3	N4	N5	N6	R1	R2	R3	PV(1)	PL(1)	F1(2)	PV(NT)	F1(NT-1)
8	10	7	7	14	12	0.14	1.44	-1.0	30.5	41.44	20.0	0	0

Direct Substitution method used to update the torn variables

Z_{sol} / X2(1,NW1) = 0.4401 , X2 (2,NW1) = 0.5476

Table 3.3a Different maximum number of iterations in each column simulation

Z_o		minws	nac	maxdis	it	time	case
0.55	0.4	14	4	20	9	1.748	1
0.55	0.4	no acceleration		10	9	1.561	2
0.55	0.4	no acceleration		5	10	1.211	3
0.55	0.4	no acceleration		2	11	0.757	4
0.55	0.4	no acceleration		1	13	0.568	5
0.333	0.333	14	4	20	8	1.733	6
0.333	0.333	no acceleration		10	8	1.363	7
0.333	0.333	no acceleration		5	8	0.964	8
0.333	0.333	no acceleration		2	10	0.667	9
0.333	0.333	no acceleration		1	14	0.548	10

Table 3.3b Different initial values for the torn variables

Z_o		minws	nac	maxdis	it	time	case
0.7	0.2	no acceleration		5	10	1.266	11
0.2	0.6	no acceleration		5	9	1.054	12
0.1	0.2	no acceleration		5	9	1.169	13

Broyden method used to update the torn variables

Table 3.3c Different maximum number of iterations in each column simulation

Z_o	minws	nac	maxdis	it	time	case
0.333 0.333	14	4	20	15	2.95	14
	no acceleration		10	19	2.478	15
	no acceleration		5	16	1.823	16
			2	bounds impeding progress		17

Table 3.3d Different initial values for the torn variables

Changing the initial guess for the torn variables

Z_o	minws	nac	maxdis	it	time	case
0.55 0.40			20	bounds impeding progress		18
0.70 0.20			20	bounds impeding progress		19
0.20 0.60			20	24	4.781	20
0.10 0.20			20	36	8.486	21

Example IV

X_F = 0.3, 0.4, 0.3 α = 6.7, 2.4, 1.0

N1	N2	N3	N4	N5	N6	R1	R2	R3	PV(1)	PL(1)	F1(2)	PV(NT)	F1(NT-1)
8	10	8	7	15	12	0.23	1.6	-1.0	43.0	34.0	20.0	0.0	0.0

Z_{sol} / X2(1,NW1) = 0.310 , X2(2,NW1) = 0.657

Direct substitution method used to update the iterations in each column simulation

Table 3.4a Different maximum number of iterations in each column simulation

Z_o		minws	nac	maxdis	it	time	case
0.333	0.333	14	4	20	8	1.904	1
0.333	0.333	no acceleration		10	8	1.514	2
0.333	0.333	no acceleration		5	8	1.041	3
0.333	0.333	no acceleration		2	9	0.657	4
0.333	0.333	no acceleration		1	14	0.542	5

Table 3.4b Different initial values for the torn variables

Z_o		maxdis	it	time	case
0.1	0.2	5	7	0.995	6
0.2	0.6	5	7	0.877	7
0.55	0.4	5	9	1.114	8
0.7	0.2	5	9	1.190	9

Broyden method used to update the value of the torn variables

Table 3.4c Different maximum number of iterations in each column simulation

Z_o	minws	nac	maxdis	it	time	case
0.333 0.333	14	4	20	21	3.517	1
0.333 0.333	no acceleration		10	18	2.946	2
0.333 0.333	no acceleration		5	23	2.806	3
0.333 0.333	no acceleration		2	14	0.799	4
0.333 0.333	no acceleration		1	bounds impeding progress		5

Table 3.4d Different initial values for the torn variables

Z_o	maxdis	it	time	case
0.10 0.20	20	80$^{(*)}$	11.200	6
0.20 0.60	20	19		7
0.55 0.40	20	bounds impeding progress		8
0.70 0.20	20	bounds impeding progress		9

(*) did not converge

Table 3.5 Direct Substitution Convergence

Example	I	II	III	IV
Case	1	1	1	1
Iteration No.	6	6	6	5
Estimated eigenvalue	0.52	0.884	0.4	0.306
Exact eigenvalue (λmax)	0.51	0.7867	0.45	0.287
Estimated No. of additional steps ($\lambda^{\text{est}}_{\text{max}}$)	7 (6.75)	42	2 (1.6)	2.4
Exact No. of additional steps	7	29	2	3

3.6 Comments

In the first and second example, there is only a vapour recycle stream to the bottom of the first column, therefore only two vapour compositions are needed as torn variables, Y1(2,NW3) and Y1(3,NW3). In the first example, the products satisfied a minimum recovery and purity of 90 percent, while in the second example the reflux ratio in both condensers of the second column were increased in such a way that a minimum purity of 95 percent was obtained. In Table 3.5 the values of the estimated maximum eigenvalue, calculated in the fifth or sixth iteration, and the value calculated at the solution, are shown together with the predicted number of additional steps required to find the solution. The real steps needed to find the solution are shown in the same table. The approximation to the dominant eigenvalue (λ_{max}^{est}) is good enough to give an idea of the rate of convergence of the DSM and, in a general case, whether an acceleration technique or a different method altogether should be used to solve the problem.

In the first example, the estimation of the dominant eigenvalue in the sixth iteration is a very good one and the number of additional steps predicted is the same as the number of steps needed to find the solution. Allowing a maximum of 20 iterations for the simulation of the distillation columns, BM converged quicker than the DSM in cases 1 and 10, but in all the other cases DSM was more efficient and never failed to find the solution. The dominant eigenvalue in this example is 0.51 and the DSM is expected to have a good rate of convergence. In Table 3.1a, the maximum number of inner iterations for the column simulation is changed from 20 to 1, and the computing time decreases as MAXDIS decreases. Indeed, when only one iteration was allowed inside the distillation columns (case 5), the computing time required was less than a third of the time required with MAXDIS = 20 (case 1).

Table 3.1b shows the behaviour for different initial values of the torn variables. The cases were run with MAXDIS = 5 to compare them with BM, because BM is very likely to fail to find a solution if MAXDIS < 5 as can be seen in Table 3.1c and 3.1d (cases 15, 19, 20, 23). For all the different initial guesses DSM was more efficient than BM. In Table 3.1c, BM was run with different values of MAXDIS, and the computing time decreases as MAXDIS decreases up to a point in this case MAXDIS = 3 (case 13). For MAXDIS = 2 the computer time increased compared with MAXDIS = 3, and for MAXDIS = 1, BM failed to find a solution because the bounds on the variables were impeding progress. In Table 3.1d, the Steffensen method was used to accelerate the convergence in the inner loop of each distillation column, while BM was used to update the torn variables. Some reduction in the computing time can be observed (cases 25 and 26), although the reduction is less than the one obtained decreasing MAXDIS.

In the second example, the dominant eigenvalue is 0.787 and the DSM required more iterations than in the previous case to converge. In Table 3.2a, for values of MAXDIS from 20 to 5 inclusive, BM was more efficient than DSM. For MAXDIS = 2, BM did not converge in 80 iterations, but the DSM went on reducing the computing time needed as MAXDIS decreased, becoming the most efficient way of simulating the system for MAXDIS = 1. The computing time using DS and MAXDIS = 1 is less than one sixth of the computing time using DS and MAXDIS = 20. BM was used with different initial values for the torn variables and with MAXDIS = 5, it failed to find a solution in 2 cases (1 and 2) out of four trials. DSM never failed to find a solution, and the option of MAXDIS = 1 and DSM was the most effective way of solving this example.

In the third example, a different mixture is separated into its components, and in this case there is a liquid recycle stream only, being fed at the top of the first column. The dominant eigenvalue is 0.45 and the DSM was more efficient than the BM in all the cases. The robustness of the BM was poorer than in the two previous examples failing to find a solution even for MAXDIS = 20.

The last example was tried with the same feed and the same design variables as in example three, except for the two reflux ratios corresponding to both condensers in the second column that were increased. The dominant eigenvalue decreased to 0.312 and obviously the DSM performed better than the BM. Again the robustness of the BM was poor, not finding the solution for MAXDIS = 20 because bounds on the variables were impeding progress.

It can be said that the direct substitution method with MAXDIS = 1 was the best option in all the cases.

CHAPTER IV

THE OPTIMUM DESIGN OF A DISTILLATION COLUMN

4.1 Introduction

The optimum design of a distillation column is of special interest because this is the unit operation that is most frequently employed for separating products from a given mixture. Short cut methods have been widely used to design distillation columns, particularly to find the number of theoretical stages required for a given separation. Empirical equations have been proposed relating the reflux ratio and the total number of stages of a given column with two products, where the minimum reflux and the minimum number of ideal stages are known. The minimum stage number required is obtained at infinite reflux using the Fenske-Underwood (28) equation that requires constant relative volatility, though some modifications have been suggested for use in cases where the relative volatilities are not constant. The minimum reflux ratio is the maximum reflux ratio which will require an infinite amount of trys to obtain a required degree of separation and represents the minimum cost in heating and cooling utilities. The calculation of the minimum reflux for non-conventional columns has received some attention in the literature, but when the restriction imposed to simplify the model, like constant relative volatility and flow rates, are lifted the procedure can become as time consuming as a plate to plate simulation. Recently Cerda and Westerberg (29) have proposed simplified methods of determining minimum reflux conditions and have suggested the use of Edmister's "absorption factor method" to determine the number of plates and feed plate locations. However the approximations used depend heavily on the fact that the separations are sharp, which is an undesirable restriction for our approach and on the assumption that internal flow rates and relative volatiles are constant.

Given the limitations of these short-cut methods, and the underlying need to make the simplifications of constant flow rates and relative volatilities, it seemed worth examining the savings that could be made by using these simplifications in a plate by plate simulation of the column. Accordingly, the simplified algorithm described in Chapter II was developed, and this indeed proved fast enough for use in an optimum design procedure. This approach has the advantage that the model can be refined to locate the optimum more precisely - the ideal algorithm would in fact progressively increase the sophistication of the model as the optimum is approached. This of course requires the addition of energy balances to the model, but the computing time will not be greatly increased if simple expressions are used to calculate enthalpies and equilibrium-ratios.

When the number of stages in each section is known the design constraints can be added to the system of equations describing the distillation process. If the design specifications are expressed as equality constraints and their number is equal to the degrees of freedom of the system, a well determined system of nonlinear equations and variables is to be solved, and any standard algorithm may be used to find the solution of all the equations simultaneously. The need to express the design specifications as constraint equalities leaves much to be desired, especially in separation processes where the specifications will normally be a minimum recovery or purity in more than one product and it might well be that there is no feasible solution with all the constraints specified as equality constraints.

When designing a distillation column, there are frequently more degrees of freedom than design specifications, so that the design variables can be chosen to minimize or maximize a given objective function like the cost or profit of running the process. When the stage numbers are defined, a nonlinear constraint problem with a nonlinear objective function can be defined and solved with standard optimization techniques. The equations describing the column can be included as equality constraints; however, given the large number of equations representing the column, it is probably best to solve them separately. Some work has been reported lately by Boston (30) on the optimization of a distillation column where the stage numbers of each section are known. A variable metric algorithm developed by Powell (31) was used to find the design variables that would minimize a given cost function. The equations describing the distillation column were solved separately using the "inside out" algorithm (18), to reduce the search space.

When the stage numbers are included as the optimization variables in such a way that the structure of the column and the operating conditions are calculated at the same time, the problem cannot be solved with standard optimization techniques due to the inclusion of integer variables in the set of optimization variables. No technique is yet available to solve an optimization problem in which integer and real variables are solved at the same level. Sargent and Gaminibandara (5) attempted the optimum design of a distillation column in two levels: an inner optimization level in which, given the total number of stages, the feed and product distributions along the column together with the energy requirements were optimized and an outer optimization level in which the total number of stages of the column is searched to minimize a given objective function.

This approach requires that for each value of the total number of stages given by the outer level of optimization, some kind of heuristic rules should be developed to define the places in which the number of stages added or subtracted should be put in or taken away. Otherwise the initial point for each inner optimization should distribute the feed and (or) the products along the column, depending on the design specifications. This last option will not use the information obtained in the previous inner optimization and the initial point for each inner optimization will always be far from the point obtained in the previous inner optimization, thus decreasing enormously the efficiency of the optimization algorithm. The Variable Metric Projection method of Murtagh and Sargent (67) was used in the inner loop while a one-dimensional minimization method of Sargent and Sebastian (68) was used in the outer loop.

In the present work the stage numbers of each section were optimized at the same level that the other design variables as it will be seen in the following sections.

4.2 Formulation of the Optimization Problem

The mathematical description of a distillation process consists of a set of nonlinear equations containing state and design variables. The number of state variables is the same as the number of equations describing the system. If the design variables are known the values of the state variables can be determined by solving the set of equations as has been described in Chapter II. The design problem to be considered is to select the design variables in such a way that they minimize a specified objective function.

The objective function will be given by the annual cost of running the distillation column and could be written as

$$F_{obj} = C(u,s) \qquad (4.1)$$

where

S is the state variable vector of dimension m

u is the design variable vector of dimension (n-m), n being the total number of variables

The performance of the distillation process can be represented by the system of nonlinear equations

$$f(u,s) = 0 \qquad (4.2)$$

where f is the m vector of equations.

The design specifications have the general form

$$c^L < g(u,s) < c^u \qquad (4.3)$$

where g is the vector of constraint functions with lower and upper bounds c^L and C^u.

Then the general design problem consists of solving the following problem:

$$\min F_{obj} = C(u,s)$$

$$\text{st} \quad f(u,s) = 0 \qquad (P1)$$

$$c^L < g(u,s) < c^u$$

The dimension of the nonlinear vector f describing the distillation system is big. To reduce the size of the optimization problem, the equality constraints were solved separately using the simplified algorithm described in Chapter II for the simulation of a distillation column. This simplifies the optimization problem to the form

$$\min F_{obj} = C(u) \tag{4.4}$$

$$\text{st} \qquad c^L < g\,(u) < c^u \tag{4.5}$$

the dimension of the vector u now being equal to the number of degrees of freedom of the system. Some bounds on the optimization variables may also be specified as follows:

$$\nu^L < u < \nu^u \tag{4.6}$$

4.2a The Objective Function

For a given separation, the greater the reflux used the fewer the plates needed but the more steam and cooling water, the larger the diameter of the column, and the greater the size of the heat exchangers required.

There are many different systems for measuring the profitability of a process. When the plant is planned to operate at a constant annual production rate over a definite number of years, the annual annuity charge can be defined as

$$a = \frac{\pi(1+\pi)^n}{(1+\pi)^n - 1} \tag{4.7}$$

where $\pi = i + f$ (4.8)

i equals the percentage rate of interest divided by 100

f equals the percentage of inflation divided by 100

n number of years (lifetime)

The annual cost of running a distillation process was calculated as:

$$C_{annual} = a\ I_F + R_C \tag{4.9}$$

I_F = total investment in the distillation column

R_C = running costs per annum

The total installed equipment cost is made up of the cost of the distillation column and its plates and the cost of heat exchangers, condensers and reboilers. Capital costs for piping, insulation and instrumentation were estimated to add 60 percent to the installed costs of the equipment. The installed cost of the equipment was calculated following Happel and Jordan (40). These simplified costs are sufficient for preliminary design purposes. As an illustration, formulae for the cost of a single column with a liquid feed and two liquid products, one condenser and one reboiler will be developed.

The cost of a distillation column can be expressed as

$$\frac{C_o^{col} \ S^{0.77} \ N}{E \ Y} \tag{4.10}$$

where

C_o^{col} annual investment cost (£/plate yr)

S tower cross sectional area (ft^2)

N number of theoretical stages

E overall column efficiency

Y hours/year operation

The tower cross sectional areas can be calculated as:

$$S = V/G_a = D(1+R)/G_a \tag{4.11}$$

where V vapour throughput rate (lb moles/hr)

G_a allowable vapour velocity (lb moles/hr ft^2)

R reflux ratio

D product flow rate (lb moles/hr)

In most cases, the vapour velocity is limited by entrainment of liquid droplets in the rising vapour which can result in improper operation or even flooding of the column. A tower must have sufficient cross sectional area to handle the rising vapour without excessive entrainment of liquid from one plate to another.

The overall column efficiency, defined as the number of theoretical stages required to produce a given separation divided by the number of real stages necessary to produce the same separation, was used because of its simplicity.

Then, the cost of the column becomes

$$\frac{C_o^{col}\ N\ \left[(1+R)\ D\right]^{0.77}}{G_a^{0.77}\ E\ Y} = C_{col} \tag{4.12}$$

$$\text{if } C_1' = C_o^{col} \,/\, G_a^{0.77}\ E\ Y \tag{4.13}$$

$$C_{col} = C_1'\ N\ \left[(1+R)\ D\right]^{0.77} \tag{4.14}$$

The cost of the heat exchanger can be calculated as

$$C_{he} = C_o^{hex}\ A^{0.6} \,/\, Y \tag{4.15}$$

where C_o^{hex} annual investment cost of the heat exchanger

A area of heat transfer

The heat transfer area can be obtained from

$$A = Q/U\Delta T \tag{4.16}$$

Q	heat exchanged
U	overall heat transfer coefficient
ΔT	overall temperature difference

$$Q_c = (1+R)\ D\ \lambda_c \tag{4.17}$$

$$Q_R = (1+R)\ D\ \lambda_R \tag{4.18}$$

where Q_c and Q_R are the heat exchanged in the condenser and reboiler

λ_c and λ_R are the latent heat in the condenser and reboiler

then

$$A_c = (1+R)\ D\ \lambda_c / U_c \Delta T_c \tag{4.19}$$

$$A_R = (1+R)\ D\ \lambda_R / U_R \Delta T_R \tag{4.20}$$

The cost of both heat exchangers can be written as

$$C_{C+R} = (C_{2C}+C_{2R})\ (1+R)^{0.6}\ D^{0.6} \tag{4.21}$$

where $$C_{2C} = C_c \lambda_c^{0.6} \Big/ \left[U_c \Delta T_c \right]^{0.6}\ Y \tag{4.22}$$

$$C_{2R} = C_R \lambda_R^{0.6} \Big/ \left[U_R \Delta T_R \right]^{0.6}\ Y \tag{4.23}$$

It has further been assumed that the steam cost is a linear function of its condensation temperature

$$C_{steam} = (0.04\ T - 1.0)\ \text{£/tonne} \qquad (T \text{ in } {}^{o}C) \qquad (4.24)$$

The combined cost of steam and cooling water could be obtained from

$$C_{s+c} = (C_{steam} + C_{cw})\ (1+R)\ D \qquad (4.25)$$

The total cost of the distillation column is the sum of all these costs

$$\begin{aligned} C_{column} = {} & C_1'\ N\ \left[D(1+R)\right]^{0.77} + \\ & + (C_{2c}+C_{2R})\ (1+R)^{0.6}\ D^{0.6} + \\ & + (C_{steam}+C_{cw})\ (1+R)\ D \end{aligned} \qquad (4.26)$$

and $N = N1 + N2$ (4.27)

where

N1 is the number of theoretical stages above the feed

N2 is the number of theoretical stages below the feed

The optimization variables are N1, N2, R and D.

4.2b Constraints

The requirements for the separation process are expressed as constraints of the optimization problem, and will normally be specifications of the products such as purity, recoveries, temperatures, etc. It is sometimes difficult to find algebraic expressions of the constraints as functions of the optimization variables, like the stage number. In most of the cases the constraints will be inequality constraints, and in the space in which these requirements are satisfed a minimum value of the objective function will be found.

4.3 Method of Solution

The problem has been presented as an integer nonlinear problem, because some integer variables are included in the set of optimization variables. Since no explicit algorithm is yet available to solve this class of problem, the integer variables were treated as real variables and the problem posed as a nonlinear programming problem. The objective function and the constraints are normally nonlinear functions of the optimization variables and an augmented lagrangian algorithm called MINOS, of Murtagh and Saunders (38, 39) was used to find the minimum of the objective function in the feasible space defined by the constraints. MINOS requires both function and derivatives with respect to the optimization variables for the objective and constraint functions. For the objective function, there is no difficulty in obtaining the derivatives analytically, but for the constraints finite-difference approximations were used.

4.3a Objective Function Derivatives

Rewriting the cost function for a column with two products presented in Equation (4.26)

$$\begin{aligned} C = {} & C_1' \; (X1+X2) \; \left[D(1+R)\right]^{0.77} \; + \\ & + (C_{2c}+C_{2R}) \; (1+R)^{0.6} \; D^{0.6} \; + \\ & + (C_{steam}+C_{cw}) \; (1+R) \; D \end{aligned} \tag{4.28}$$

the analytical derivatives can be calculated as follows

$$\begin{aligned} \frac{\partial C}{\partial X1} &= C_1' \; \left[D(1+R)\right]^{0.77} \\ \frac{\partial C}{\partial X2} &= \frac{\partial C}{\partial X1} \end{aligned} \tag{4.29}$$

$$\begin{aligned} \frac{\partial C}{\partial R} = {} & C_1' \; (X1+X2) \; 0.77 \; (1+R)^{-0.23} \; D^{0.77} \; + \\ & + (C_{2R}+C_{2R}) \; 0.6 \; (1+R)^{-0.4} \; D^{0.6} \; + \\ & + (C_{steam}+C_{cw}) \; D \end{aligned} \tag{4.30}$$

$$\begin{aligned} \frac{\partial C}{\partial D} = {} & C_1' \; (X1+X2) \; (1+R)^{0.77} \; 0.77 \; D^{-0.23} \; + \\ & + (C_{2c}+C_{2R}) \; (1+R)^{0.6} \; 0.6 \; D^{-0.4} \; + \\ & + (C_{steam}+C_{cw}) \; (1+R) \end{aligned} \tag{4.31}$$

The real numbers X1 and X2, corresponding to the ideal stages in each section, given by the optimization algorithm as the solution values for these variables, have to be converted into real stages using the global efficiency. Having done that, the real numbers corresponding to the real stages have to be rounded up to integer numbers. The purities increase as the stage number in each section increases, so that the integer number most likely to satisfy the purity constraints is the nearest bigger integer number bracketing the real number corresponding to a given section.

4.3b Constraint Derivatives

Plate numbers have to be modified by integral amounts and an appropriate interpolation scheme was used to give data corresponding to the non-integer values of the plate number given by the optimization algorithm. The technique used is illustrated in Figure (4.1) for two integer variables. Thus if X1 and X2 are the non-integer values for the plate numbers, these are first rounded to the nearest integer number in each case to provide the base point (N1, N2+1) in Figure (4.1), then perturbations are made to the adjacent integer which brackets the corresponding non-integer value in each case, yielding points (N1+1, N2+1) and (N1,N2). For a typical dependent variable Y, the required values are then obtained as follows:

$$\begin{aligned} \frac{\partial Y}{\partial X1} &= Y(N1+1, N2+1) - Y(N1, N2+1) \\ \frac{\partial Y}{\partial X2} &= Y(N1, N2+1) - Y(N1,N2) \end{aligned} \qquad (4.32)$$

$$Y(X1, X2) = Y(N1,N2+1) + \frac{\partial Y}{\partial X1}(X1-N1) + \frac{\partial Y}{\partial X2}(X_2-(N2+1)) \qquad (4.33)$$

For the continuous variables, forward difference approximations

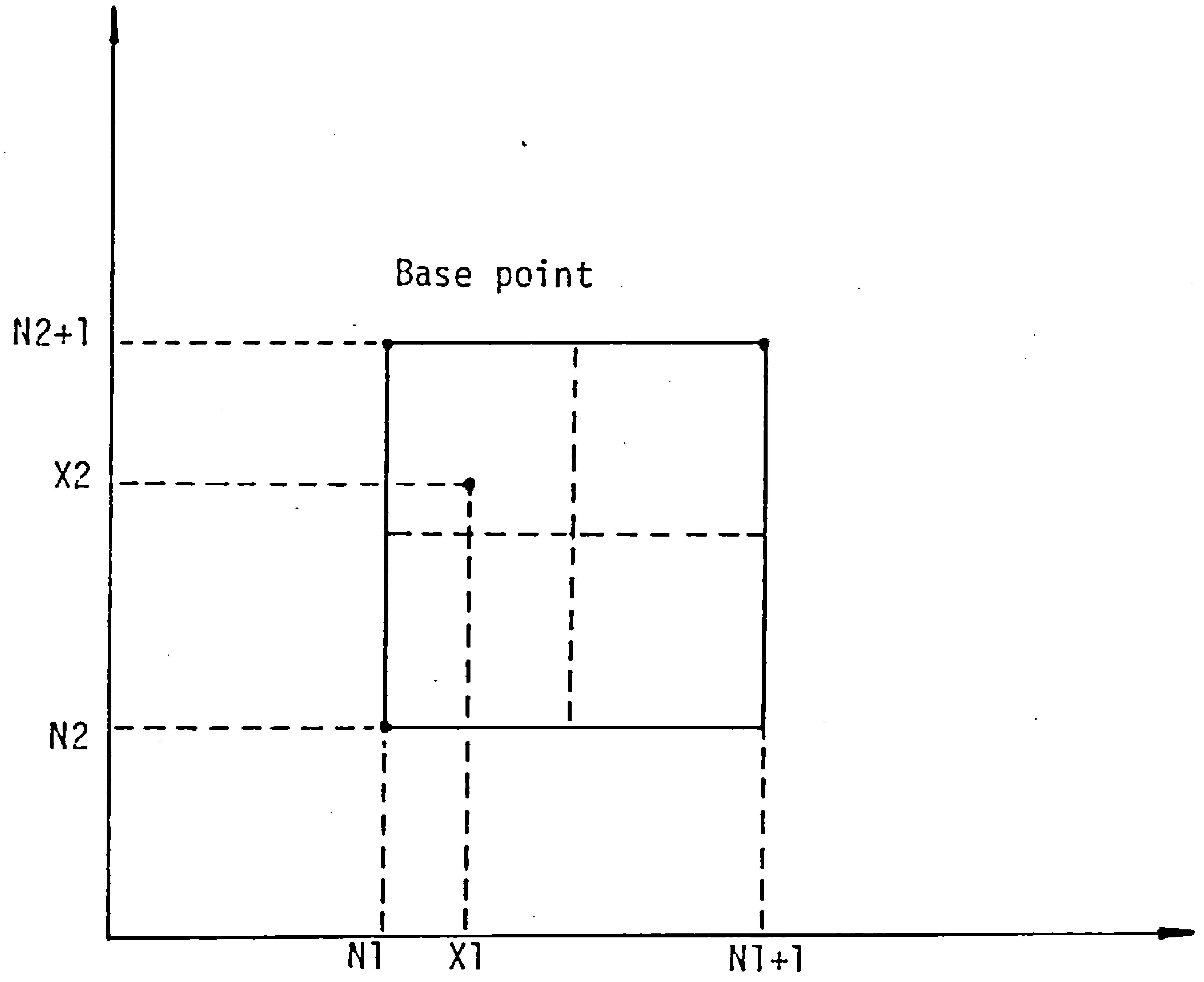

Figure 4.1

were used such as

$$\frac{\partial Y}{\partial X_i}(X) \simeq \frac{g(X+h_i)-g(X)}{h_i} \qquad (4.34)$$

to estimate the partial derivatives of the constraints. If the perturbation h_i is too small, cancellation errors could be high. On the other hand, if h_i is too large the estimation error could be excessive. Constant perturbations of 1% of the values were made. For each derivative approximation, one simulation of the column is required, and this is one of the main reasons to keep the number of the optimization variables as low as possible. As a small perturbation on each variable is required, the initial guess for the state variables in each simulation can be made equal to the solution of the previous simulation, this value being a very good initial point in most of the cases.

4.4 Numerical Examples

If steam is only available at a limited number of temperature levels, the temperature differences in the reboilers are fixed by heat sources available in the plant in consideration.

On the other hand, if the whole plant is being designed, it might well be that steam can be produced at the pressure levels required to match the temperature levels of the distillation columns. This will be the case if the cost of running the separation process is an important fraction of the total energy cost. Obviously another important variable to be optimized would be the pressure of the column in such a way that given the boundary conditions for heat to be exchanged, the pressure of the column could be chosen to use the available energy levels in the most efficient way. This problem was not attempted, but in principle, the pressure of the column could be added to the set of optimization variables and an algebraic expression should be found defining the fixed and running costs as functions of the pressure, then, the problem could be solved using the method developed in Section 4.3.

Supposing that steam was available at different pressures, the source of heating was chosen to be at a temperature 10°C higher than the liquid being vaporized and the steam cost was represented by the equation

$$\text{Steam cost} = 0.04\ T - 1.0\ \text{£/tonne} \qquad (T \text{ in } {}^{0}\text{C}) \qquad (4.35)$$

and $$T = T_{b\ell} + 10 \qquad (4.36)$$

where $T_{b\ell}$ is the bubble point of the liquid being vaporized

It was assumed that cooling water was available at 20°C and it would increase its temperature up to 10°C less than the temperature of the liquid being condensed.

The assumption of 10°C as the minimum driving force for heat transfer in the exchangers was taken to minimize the expenses in cooling and heating utilities. The steam cost is by far the most important term in the total cost of running a distillation column as can be seen in Table XVI. It seems reasonable to assume a fixed ΔT_{min}, that might well be other than 10°C, because the cost of the area needed for heat transfer in the reboilers is 20 times smaller than the steam cost. On the other hand, the cooling water cost is similar to the fixed cost of the condenser, both being small terms. For high temperature levels of the vapour to be condensed, the use of a ΔT bigger than $10^{0}C$ will decrease the fixed cost of the condenser and the total cost could be improved. The cost of water at $20^{0}C$ was £0.025/1000 gal.

For the fixed cost of heat exchangers the following relations, based on "installed costs" data given by Peters and Timmerhaus (32) were used

$$\text{Reboiler cost} = £124\ A^{0.6} \tag{4.37}$$

$$\text{Condenser cost} = £80\ A^{0.6} \tag{4.38}$$

where A (ft^2) is the heat exchange area based on appropriate heat transfer coefficients.

The cost for piping, insulation and instrumentation was estimated to be 60 percent of the cost of the installed equipment.

The installed cost of the distillation column was calculated as follows:

$$\text{Column cost} = £55\ A^{0.77} \text{ per real plate} \tag{4.39}$$

where A (ft^2) is the cross sectional area of the column.

The global efficiency used was 80 percent for all the examples.

4.4a A Single Column with Two Products

In Examples I to X, binary mixtures are separated in a distillation column at atmospheric pressure. A minimum recovery and purity of 90 percent is required in both products. Care has to be taken to specify the design constraints, specially when equality constraints are to be used. For example, if a recovery of 90 percent of the component in the largest amount is satisfied, a minimum purity of 90 percent of the component in the smallest amount in the corresponding product cannot be obtained. Similarly, supposing that a purity of 90 percent is required in both products, a recovery of 90 percent of the component in the smallest amount cannot be achieved. On the other hand, when a minimum recovery and composition are satisfied by the product corresponding to the component in the smallest amount, the same recovery and composition is satisfied by both products. Specifying constraints for one product only could make the job easier for the optimization algorithm, otherwise the four constraints should be specified as inequality constraints.

When the recovery and the purity constraints of a given product are satisfied as equality constraints, the flow rate of the product is fixed. The flow rates of the products are therefore fixed during optimization and their values are defined by the specifications of 90 percent recovery and 90 percent purity of the component in the smallest amount. Thus, only a minimum purity constraint for the smallest product is needed. This constraint is always active at the solution.

A total condenser is used because the top product was required as a liquid. The optimum column for the separation of three different mixtures, each one of them with three different feed compositions is calculated. They are fed at a rate of 500 moles lb/hr. The optimum design for the fourth binary mixture is obtained using different initial points. The annual cost of running the distillation column is minimized using MINOS. All the cases are run with the normal steam price (case A) and with the price of steam ten times cheaper than the real one (case B), to analyze the sensitivity of the solution to different prices of the most important term of the objective function. In each example the ratio of the reflux ratios and of the total stages numbers between case B and case A are shown where NT is defined as

$$NT = X1+X2$$

The values of the overall heat transfer coefficients for each example are given as U_c and U_R, where c and R refers to the condenser and reboiler respectively.

4.4b A Single Column with Three Products

In Examples XI to XV, a special case of one column with three products, shown in Figure 4.2, was optimized. A ternary mixture is separated into its components in a column at atmospheric pressure. Three liquid products are obtained from the column which has three heat exchangers, two condensers and one reboiler, so that two reflux ratios can be defined - one at the top of the column calculated as the ratio between the liquid being recycled into the column and the top product flow rate and a second reflux ratio corresponding to the intermediate condenser which can be defined as

$$R2 = PV(N1+N2+1)/PH \qquad (4.40)$$

Supposing that R2=0, then PV(N1+N2+1) = 0 and no condenser is needed. The liquid product is extracted directly from the internal liquid stream.

The optimization variables are X1, X2, X3, R, R2, D, PH, where the first three variables correspond to the stage numbers in each section of the column and PH is the molal flow rate of the intermediate product.

To calculate the fixed cost of the column, each section of the column has a different cross sectional area depending on the internal vapour flow rate going through the given section. The total annual

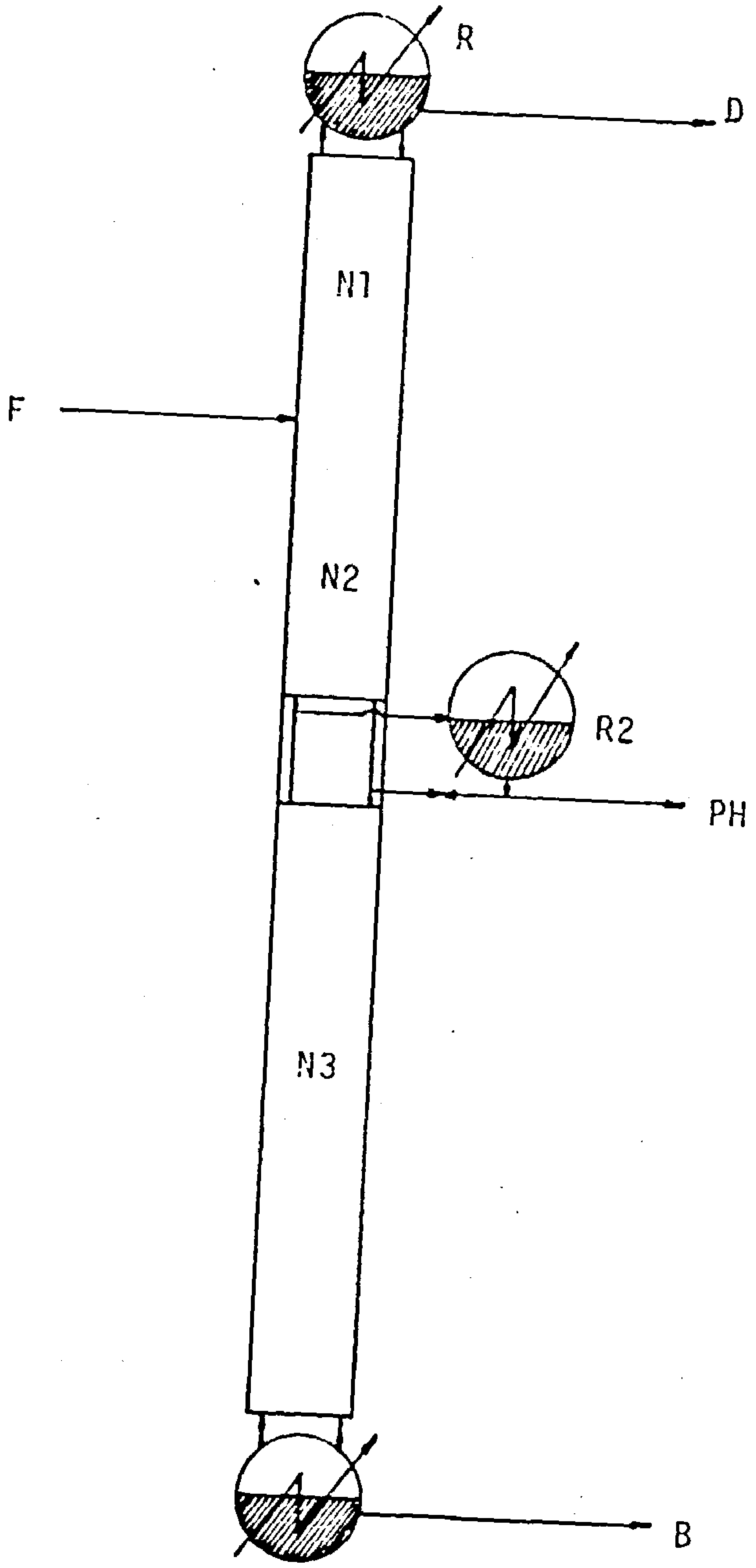

Figure 4.2

cost to be minimized is given by the following expression:

$$\begin{aligned} C_{TOTAL} = {} & C_1' \, (X1+X2) \, (D(1+R))^{0.77} + \\ & + C_1' \, X3 \, (D(1+R) + R2 \times PH)^{0.77} + \\ & + C_{2c} \, (D(1+R))^{0.6} + C_{3c} \, (R2 \times PH)^{0.6} + \\ & + C_{2R} \, (D(1+R) + R2 \times PH)^{0.6} + \\ & + C_{cwT} \, D \, (1+R) + C_{cwI} \, R2 \times PH + \\ & + C_{steam} \, (D(1+R) + R2 \times PH) \end{aligned} \tag{4.50}$$

where the first and second terms are the fixed cost of the column , the third and fourth terms, the fixed cost of the top and intermediate condensers respectively; the fifth term represents the fixed cost of the reboiler; the sixth and seventh terms the cooling water cost in the top and intermediate condensers and the last term represents the steam cost.

For a column with three sections there are three integer variables to be optimized and for each real point representing these three variables X1, X2 and X3 there are eight integer base points to choose from. As in the previous case X1, X2 and X3 are first rounded to the nearest integer number in each case to provide a base point (N1, N2, N3) and then perturbations are made to the adjacent integer number which brackets the corresponding non-integer value in each case, to approximate the partial derivatives using finite differences.

These finite approximations are required to calculate the value of any given state or dependent variable (Y) as a function of X1, X2 and X3 as shown in the following equation:

$$Y(X1,X2,X3) = Y(N1+N2+N3) + \frac{\partial Y}{\partial X1}(X1-N1) + \\ + \frac{\partial Y}{\partial X2}(X2-N2) + \frac{\partial Y}{\partial X3}(X3-N3) \qquad (4.51)$$

In addition the optimization method used requires values of the constraint derivatives, so that these finite approximations are provided as the partial derivatives of the constraints. For the real variables, constant perturbations of 1% were made.

Two different ternary mixtures with different feed compositions are processed at a rate of 500 moles lb/hr. Three liquid products were obtained, with each column being run at atmospheric pressure. The flow rates of the products are specified in each example as D and PH, corresponding to the top and intermediate product.

Example Ia

Table 4.1a Normal steam price

	X_F	α	T(^oC)	D
toluene	0.3	2.4	111	150
o-xylene	0.7	1.0	174	

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15	11.52	2.0	15.0	14.40	15.0
X2	15	15.00	2.0	15.0	18.75	19.0
R	3.3	1.919	2.0	8.0		
Cost(£/hr)	33.068	22.515				

Iterations	12
Function and gradient calls	70
Time (6500) secs	32.27
Time per column simulation (6500) secs	0.115

U_c = 100 BTU/hr ft^2 ^{o}F

U_R = 80 BTU/hr ft^2 ^{o}F

Example Ib

Table 4.1b Steam price ten times less than the real one

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	8.46	2.0	15.0	10.58	11.0
X2	15.0	10.04	2.0	15.0	12.55	13.0
R	3.0	2.005	0.2	8.0		
F_{obj} (£/hr)	5.375	3.750				

Iterations	30
Function and gradient calls	191
Time (6500) secs	72.70
Time per simulation of the column (6500) secs	0.095

Rb / Ra = 1.045

(NT)b /(NT)a = 0.698

Example II

Table 4.2a Normal steam price

	X_F	α	T(^oC)	D
Toluene	0.5	2.4	111.0	250.0
O-xylene	0.5	1.0	174.0	

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	11.01	2.0	15.0	13.76	14
X2	15.0	11.05	2.0	15.0	13.81	14
R	1.5	0.955	0.2	8.0		
Cost(£/hr)	32.065	24.978				

Iterations	10
Function and gradient calls	78
Time (6500) secs	28.96
Time per column simulation (6500) secs	0.093

U_c = 100 BTU/hr ft^2 ^oF

U_R = 80 BTU/hr ft^2 ^oF

Table 4.2b Steam price ten times less than real one

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	7.000	2.0	15.0	8.75	9.0
X2	15.0	8.002	2.0	15.0	10.00	10.0
R	1.5	1.036	0.2	8.0		
Cost(£/hr)	5.5695	3.9940				

Iterations	30
Function and gradient calls	228
Time (6500) secs	38.84
Time per column simulation (6500) secs	0.043

Rb / Ra = 1.085

(NT)b / (NT)a = 0.725

Example III

Table 4.3a Normal steam price

	X_F	α	$T(^oC)$	D
Toluene	0.7	2.4	111.0	350.0
O-xylene	0.3	1.0	140.0	

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	14.0	14.41	2.0	17.0	18.01	18
X2	16.0	12.00	2.0	17.0	15.00	15
R	1.0	0.73784	0.2	8.0		
Cost (£/hr)	35.823	31.0415				

Iterations	7
Function and gradient calls	66
Time (6500) secs	40.16
Time per column simulation (6500) secs	0.152

U_c = 100 BTU/hr ft^2 ^{o}F

U_R = 80 BTU/hr ft^2 ^{o}F

Table 4.3b Steam price ten times less than the real one

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	14.0	8.0	2.0	17.0	10.0	10
X2	16.0	8.75	2.0	17.0	10.94	11
R	1.0	0.80623	0.2	8.0		
Cost (£/hr)	6.14800	4.93085				

Iterations	19
Function and gradient calls	140
Time (6500) secs	32.23
Time per column simulation (6500) secs	0.058

Rb / Ra = 1.093

(NT)b / (NT)a = 0.634

Example IV

Table 4.4a Normal steam price

	X_F	α	T(^oC)	D
Benzene	0.3	2.79	80.0	150.0
Toluene	0.7	1.00	111.0	

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	10.0	8.99	2.0	15.0	11.24	12
X2	10.0	11.80	2.0	15.0	14.75	15
R	1.6	1.4733	0.2	8.0		
Cost(£/hr)	11.655	11.050				

Iterations	13
Function and gradient calls	90
Time (6500) secs	92.4
Time per column simulation (6500) secs	0.257

U_c = 100 BTU/hr ft^2 ^oF

U_R = 80 BTU/hr ft^2 ^oF

Table 4.4b Steam price ten times less than the real one

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	10.0	7.0	2.0	15.0	8.75	9
X2	10.0	8.4	2.0	15.0	10.50	11
R	1.6	1.601	0.2	8.0		
Cost(£/hr)	2.555	2.390				

Iterations	29
Function and gradient calls	184
Time (6500) secs	29.9
Time per column simulation (6500) secs	0.041

Rb / Ra = 1.087

(NT)b / (NT)a = 0.741

Example V

Table 4.5a Normal steam price

	X_F	α	T(°C)	D
Benzene	0.5	2.79	80.0	250.0
Toluene	0.5	1.0	111.0	

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	14.0	8.0	2.0	20.0	10.0	10
X2	15.0	9.153	2.0	20.0	11.44	12
R	0.8	0.710	0.4	8.5		
Cost(£/hr)	13.6255	12.5125				

Iterations	24
Function and gradient calls	96
Time (6500) secs	39.6
Time per column simulation (6500) secs	0.103

U_c = 100 BTU/hr ft^2 °F

U_R = 80 BTU/hr ft^2 °F

Table 4.10b Normal steam price

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	10.0	8.7	2.0	17.0	10.86	11
X2	10.0	10.6	2.0	17.0	13.25	14
R	1.6	1.57407	1.0	8.0		
Cost(£/hr)	6.12695	6.0440				

Iterations	10
Function and gradient calls	40
Time (6500) secs	15.468
Time per column simulation (6500) secs	0.09668

Table 4.10c Steam price ten times less than real one

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	10	7.0	2.0	17.0	8.75	9
X2	10	8.31	2.0	17.0	10.39	11
R	1.6	1.69186	0.2	8.0		
F_{obj}(£/hr)	2.3143	2.0855				

Iterations	23
Function and gradients calls	297
Time (6500) secs	67.08
Time per column simulation (6500) secs	0.06

Example XI

Table 4.11a Normal steam price

	X_F	α	T(°C)	D	PH
Dichloromethane	0.3	4.9	41.0	150.0	
Chloroform	0.4	1.8	61.0		200.0
Carbon tetrachloride	0.3	1.0	76.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	13.01	2.0	30.0	16.26	17
X2	10.0	7.98	2.0	30.0	9.96	10
X3	10.0	9.03	2.0	30.0	11.29	12
R	12.0	6.8067	3.0	30.0		
R2	0.05	0.01	0.01	4.0		
F_{obj}(£/hr)	38.148	23.080				

Iterations	9
Function and gradient calls	30
Time (6500) secs	52.5

U_c = 175 BTU/hr ft^2 °F

U_R = 150 BTU/hr ft^2 °F

Table 4.11b Steam price ten times less than real price

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	10.19	2.0	30.0	12.74	13
X2	10.0	8.36	2.0	30.0	10.45	11
X3	10.0	8.50	2.0	30.0	10.63	11
R	12.0	6.955	3.0	30.0		
R2	0.02	0.01	0.01	4.0		
Cost(£/hr)	12.315	7.4245				

Iterations	11
Function and gradient calls	92
Time (6500) secs	114.51

Rb / Ra = 1.022

Example XII

Table 4.12 Normal steam price

	X_F	α	T(°C)	D	PH
Dichloromethane	0.2	4.9	41.0	100.0	
Chloroform	0.4	1.8	61.0		200.0
Carbon tetrachloride	0.4	1.0	76.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	12.32	2.0	30.0	15.40	16
X2	10.0	6.97	2.0	30.0	8.71	9
X3	10.0	7.94	2.0	30.0	9.93	10
R	12.0	10.1856	3.0	30.0		
R2	0.5	0.01	0.01	4.0		
Cost(£/hr)	25.945	21.870				

Iterations	9
Function and gradient calls	32
Time (6500) secs	52.383

U_c = 175 BTU/hr ft^2 °F

U_R = 150 BTU/hr ft^2 °F

Example XIII

Table 4.13 Normal steam price

	X_F	α	T(°C)	D	PH
Dichloromethane	0.1	4.9	41.0	50.0	
Chloroform	0.4	1.8	61.0		200.0
Carbon tetrachloride	0.5	1.0	76.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	14.00	2.0	20.0	17.50	18
X2	9.0	6.41	2.0	15.0	8.01	8
X3	9.0	6.50	2.0	15.0	8.31	9
R	22.0	20.098	10.0	30.0		
R2	0.02	0.01	0.01	4.0		
Cost(£/hr)	22.970	20.650				

Iterations	3
Function and gradient calls	9
Time (6500) secs	18

U_c = 175 BTU/hr ft^2 °F

U_R = 150 BTU/hr ft^2 °F

Example XIV

Table 4.14a Normal steam price

	X_F	α	(T^oC)	D	PH
Benzene	0.3	6.7	80.0	150.0	
Toluene	0.4	2.4	111.0		200.0
O-xylene	0.3	1.0	174.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	15.4	2.0	20.0	19.25	20
X2	10.0	4.0	2.0	15.0	5.00	5
X3	10.0	5.0	2.0	15.0	6.25	7
R	8.0	6.23	3.0	30.0		
R2	0.02	0.01	0.01	4.0		
Cost(£/hr)	69.62	57.25				

Rb / Ra = 1.054

U_c = 80 BTU/hr ft^2 ^oF

U_R = 100 BTU/hr ft^2 ^oF

Table 4.14b Steam price ten times less than the normal price

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	10.987	2.0	20.0	13.73	14
X2	10.0	4.172	2.0	15.0	5.22	6
X3	10.0	5.96	2.0	15.0	7.45	8
X4	8.0	6.5677	4.0	30.0		
X5	0.02	0.01	0.01	4.0		
Cost(£/hr)	12.320	9.445				

Iterations	29
Function and gradients calls	105
Time (6500) secs	126

Rb / Ra = 1.054

Example XV

Table 4.15 Normal steam price

	X_F	α	$T(^oC)$	D	PH
Benzene	0.2	6.7	80.0	100.0	
Toluene	0.4	2.4	111.0		200.0
O-xylene	0.4	1.0	174.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	13.0	2.0	20.0	16.25	17
X2	6.0	4.0	2.0	15.0	5.00	5
X3	6.0	5.0	2.0	15.0	6.25	7
X4	12.0	9.71	5.0	30.0		
X5	0.02	0.01	0.01	4.0		
Cost(£/hr)	66.472	54.500				

Iterations	10
Function and gradient calls	62
Time (6500) secs	85.3

U_c = 80 BTU/hr ft^2 ^{o}F

U_R = 100 BTU/hr ft^2 ^{o}F

Example XVI

Table 4.16 Normal steam price

	X_F	α	T(^oC)	D	PH
Benzene	0.1	6.7	80.0	50.0	
Toluene	0.4	2.4	111.0		200.0
O-xylene	0.5	1.0	174.0		

	Initial point	Sol point	Lower bound	Upper bound	Real plates	Rounded sol
X1	15.0	13.0	2.0	20.0	16.25	17
X2	10.0	3.0	2.0	15.0	3.75	4
X3	10.0	5.0	2.0	15.0	6.25	7
X4	24.0	19.3	10.0	30.0		
X5	0.02	0.01	0.01	4.0		
Cost(£/hr)	64.70	51.65				

Iterations	13
Function and gradient calls	153
Time (6500) secs	166

U_c = 80 BTU/hr ft^2 ^oF

U_R = 100 BTU/hr ft^2 ^oF

Table 4.17a Percentage of total cost of the separation process

Costs (%)	Example II	Example V	Example VIII	Example X
Column	3.80	5.27	11.395	11.28
Condenser	0.74	1.62	1.760	3.47
Reboiler	2.38	4.11	3.430	5.10
Cooling water	0.53	1.37	3.365	10.63
Steam	92.55	87.63	80.050	69.52

Table 5.17b Temperatures of the light and heavy pure components at atmospheric pressure

^oC	Example II	Example V	Example VIII	Example X
$T_{light\ comp}$	111	80	61	41
$T_{heavy\ comp}$	174	111	76	61

Table 4.18a Percentage of the total cost of the column

Costs (%)	Example XI	Example XIV
Column	10.06	3.21
Condenser	1.82	0.65
Reboiler	4.19	2.72
Cooling water	8.52	0.80
Steam	75.42	92.62

Table 4.18b Temperature at the top and bottom of the column

^oC	Example XI	Example XIV
Lighest component	41	80
Heaviest component	76	174

4.5 Comments

From Tables 4.17a, 4.17b, 4.18a and 4.18b, it can be observed that the higher the temperature level at which the steam has to be provided, the bigger the contribution of the steam cost to the total cost, varying from 93 percent in Examples II and XIV to 70 percent in Example X. In Examples VIII, X and XI, the cost of cooling water is much higher than in the other examples, due to the low temperature level at which cooling is needed ($41^{o}C$), contributing to the total cost with more than 10 percent in Example X. Thus, the steam and cooling water costs together represent more than 80 percent in all the cases, while the fixed cost of the column varies from 3.2 percent in case XIV to 11.4 percent in case VIII. It is clear that the dominant factor in the total cost of a distillation column is the temperature levels at which heat is exchanged, so that to find the optimal operating conditions of a given column, this factor should be taken into account.

4.5a Two Product Cases

In all the examples in which a binary mixture was separated into its components the system was first designed with the steam price given by Equation (4.35) and then with a steam price ten times less than the real one. As expected, an increase in the value of the reflux ratio (R) was observed in all the cases when cheap steam was used and at the same time a decrease of the number of ideal stages needed to obtain the same degree of separation. The increment of the reflux ratio ranged from 4.5 percent in Example I to 12.7 percent in Example VI, the average being 9.6 percent in the first ten examples while the decrease on the total number of ideal stages (NT) ranges from 25 to 40 percent. Thus, one can expect savings of the order of 10 percent in the cost of running a column by adjusting properly the reflux ratio and the number of ideal stages needed for a given separation. The computing time required to find the minimum of the cost function ranges from 15 secs to 117 secs in the CDC 6500, the average being 55 secs. So, it is clear that anyone designing a distillation column need spend less than a couple of minutes of computing time to find the optimum design of the column. As mentioned previously, different boundary conditions can easily be considered in the cost function. For instance, if for any reason steam is available and has no real cost in the context of a specific plant, then there will be no steam cost in the total cost of the column. If steam is available only at a given pressure, then the steam cost is constant regardless of the temperature level of the reboilers in the column, but the size of the reboiler will be dependent on the temperature level.

In Example X, two different initial points were used to check the sensitivity of the solution point to different initial points. The solutions obtained are slightly different, but when the real variables corresponding to ideal stages are transformed into real stages and then rounded up to integer numbers, the solution becomes the same. The reflux ratio is the same in both cases up to the third decimal.

4.5b Three Product Cases

A single column, in which the intermediate component is removed as a sidestream, the lighest component is taken overhead and the heaviest product removed as bottom product, is designed optimally in the last six examples. Although this sidestream column is unlikely to become the optimum configuration for the separation of a ternary mixture, as has been shown by Doukas and Luyben (27), it was chosen as an example to check the algorithm with non-conventional columns. They studied this column, between different alternatives, to separate a mixture of benzene, toluene and o-xylene and found that this single sidestream column is only the most economical configuration for low concentrations of benzene in the feed (less than 10 percent).

In the optimal solution, for all the cases, the intermediate product is removed from the internal liquid stream, so that no intermediate condenser is needed. This is due to the fact that the concentration of the intermediate component in the liquid is bigger than the vapour concentration.

Two cases, XI and XIV, were run at normal steam price and at a steam price ten times less than the normal one. When the cost was minimized using cheap steam, the reflux ratio increased only 2.2 and 4.5 percent for Examples XI and XIV, respectively. The minimum is less sensitive to cost prices than in the previous examples of one column with two products, probably because this column consumes more energy than the column with two products.

The efficiency and the robustness of this algorithm to find the optimum design of a single column is good. The tighter the bounds on the optimization variables the higher the efficiency and robustness of the method. As the objective function depends strongly on the reflux ratio, MINOS will tend to decrease the reflux ratio reaching non-feasible points and, then, increase the stage numbers to reach a feasible point. For normal steam prices, the reflux ratio may reach the lower bound and the real variables representing the stage numbers in each section may reach the upper bounds. Supposing that an approximation to the minimum value of the reflux ratio is known, by using an appropriate value for the lower bound on the reflux ratio the efficiency of the algorithm could be greatly increased. These values were not assumed known, but in practical cases it is likely that the designer will know, from the information already available of processes in operation, the order of magnitude of the reflux ratio and the stage number in each section. Therefore good approximations of the lower bound on the reflux ratio and upper bounds on the stage numbers could be expected, thus allowing for improvements in the efficiency of the algorithm.

CHAPTER V

OPTIMUM DESIGN OF A SYSTEM OF DISTILLATION COLUMNS

5.1 Introduction

Process systems are defined by the interconnections between the process units and the capacities and operating conditions of these process units. Therefore, the optimal design of process systems requires a search over the space of configurational alternatives as well as over the design variable space for each particular process configuration. Three reviews of the field have recently been published by Hlavacek (42), Westerberg (43) and Nishida et al (44). The major problem of synthesizing a particular process from the available units is the high dimensionality of the design problem. In the optimum design of a distillation system, the processes are restricted to distillation columns and heat exchangers, though there are many possible alternatives to choose from. The number of possible sequences rises rapidly as the number of products increases as shown by Thompson and King (45). Because of the large combinatorial problem resulting when many products are to be obtained, they proposed a simplification of the screening procedure by incorporating some heuristic rules.

Hendry and Hughes (46) used a dynamic programming technique to locate the optimum path through a tree of possible separation sequences. If the components of a mixture are ranked according to a particular physical property, then it is possible to derive all possible sub-groups of components which may result during the separation of the mixture using that particular property. This is done by first deriving all the sub-groups which may results from one splitting of the original list, and then continuing to repeat this with the sub-group so derived until no more sub-groups may be generated.

To avoid the need to search over the entire space of all possible separators, branch and bound algorithms were proposed by Westerberg and Stephanopoulos (47), Rodrigo and Seader (48) and Gomez and Seader (49). Supposing that the tree of all possible separations is developed as proposed by Hendry and Hughes (46), all the possible sequences can be found by following the branches of the tree. The cost of any of these sequences can be calculated and used as the upper bound (UB) on the optimal sequence. Any complete or partially developed separation sequence which has a lower bound larger than the UB cannot be optimal and it is disregarded. The UB can be updated as sequences are found whose total cost is smaller than the UB. The nearer the UB is to the solution the more effective the search, therefore heuristics can be used to choose the sequence whose cost is the UB initially.

Evolutionary synthesis refers to the synthesis of new processes by modifications of previously generated processes. King, Gantz and Barnes (50) applied this technique to demethanizer towers in ethylene plants. No theoretical guidance was given for the selection of process modifications; these were drawn from considerable engineering experience in this particular process unit. The use of an evolutionary approach to the synthesis of distillation columns was presented by Stephanopoulos and Westerberg (51) where four basic ideas were used: an initial flowsheet, rules to make systematic and small changes to the flowsheet, and an effective strategy to apply these rules, and means to compare the original flowsheet to any of its neighbours. Seader and Westerberg (52) examined the use of heuristic rules to aid in the first and third ideas proposed by Stephanopoulos and Westerberg (51). Often the heuristics are in conflict and in order to give different priorities to different heuristic rules, these could be ranked as proposed by Seader and Westerberg (52), but as they pointed out the ordering of the heuristics is itself an heuristic. Nath and Motard (53) further developed the combined use of the evolutionary approach and heuristic rules. Eighteen heuristic rules are reported by Nishida et al (44). The use of such rules has been of much help in developing initial structures for the branch and bound and the evolutionary approach.

The idea of determining an optimal system structure from an enlarged system structure, which is directly optimized was first introduced by Umeda, Hirai and Ichikawa (54). Streams were split into alternatives routes and when the optimized splitting ratios are close to 0 or 1, one of the alternatives streams can be eliminated. Box's complex method was used for the optimization. The advantages being that the structured variables (split ratios) and the optimal design parameters of each subsystem are obtained simultaneously. The structure solved with this approach (54) consisted of two reactors, two distillation columns and several heat exchangers. Osaka and Fan (55) optimized the same structure, coupling the approach of Umeda et al (54) with a multilevel technique.

The basic limitations of all these methods is the restriction of considering successive splits between components which excludes many potentially interesting configurations. Only conventional schemes for separating an n-component mixture in (n-1) two sectional columns have been considered. Petlyuk, Plotonov and Slavinskii (56) studied thermally coupled columns and their performance was compared with conventional columns using the following criteria: total specific amount of liquid to be vaporized for all the columns and the overall specific work of separation which takes into consideration the temperature levels at which heat is exchanged. The savings achieved with the coupled columns on the amount of liquid being vaporized is as high as 50 percent compared with the conventional scheme; only ternary mixtures were considered. Petlyuk, Plotonov and Avetlyan (57) considered various examples with more than three components and pointed out that the new arrangement would require n(n-1) different sections for a given mixture of n component to be separated into its components. This led Sargent and Gaminibandara (5) to tackle the synthesis problem by proposing a general system of interlinked columns from which most configurations of interest could be obtained by deletion of parts of the more general configuration. This general configuration for a ternary mixture was developed from the coupled system presented by Petlyuk _et al_ for a ternary mixture, in such a way that the conventional arrangements and all the cases presented by Petlyuk _et al_ (56) were included in the general configuration. The general configuration was extended to n products and is made up of n(n-1) different sections. The cost of separating various ternary mixtures into its components in eight different configurations was studied by Tedder and Rudd (58). The separation of a ternary mixture into three product streams was studied by Doukas and Luyben (59) using four different configurations including a single column with sidestream product.

The general configuration presented by Sargent and Gaminibandara (5) provides for some thermal linking between the columns, but does not deal with the general heat integration problem. Here the first work was done by Rathore _et al_ (60, 61) who considered possible matches of hot and cold streams in heat exchangers along with the separation splits, extending the algorithm of Hendry and Hughes (46) to deal with the problem. Umeda _et al_ (62) used an _ad hoc_ method based on effective use of the overall heat content-temperature diagram for the system. The branch and bound algorithm has been pursued by Sophos et al (63) and Morari and Faith (64). Again, however, the separation possibilities in all these papers are limited to sharp splits between adjacent components, and the kinds of heat integration considered are severely limited. Moreover, as pointed out by Dunford and Linnhoff (65), there are dangers in considering heat integration only in relation to the separation system; the solutions adopted may prevent wider heat integration in the rest of the process.

In this work we develop further the approach of Sargent and Gaminibandara (5) in optimizing a general configuration. A slightly more general configuration is proposed and a greatly simplified model is used in the early stages of the optimization to economize in computing effort. No attempt is made to solve the general heat integration problem, but as pointed out earlier, the configuration itself provides for some thermal coupling between columns. In addition, the objective function can reflect the cost of provision and removal of heat at various specified temperature levels, so that an optimal selection of appropriate heat sources and sinks in made. Thus heat integration with the rest of the process can be effected through adjustment of temperature levels and costs of the heat sources and sinks.

5.2 The General System Configuration

The general configuration used is illustrated in Figure 5.1 for the separation of a ternary mixture into its constituents and in Figure 5.2 for a mixture of four components. The extension to a larger system should be obvious. This general configuration differs slightly from the one proposed by Gaminibandara and Sargent (5). For example in the case of a ternary mixture, liquid and vapour feeds from the condenser and reboiler of the first column into the second column

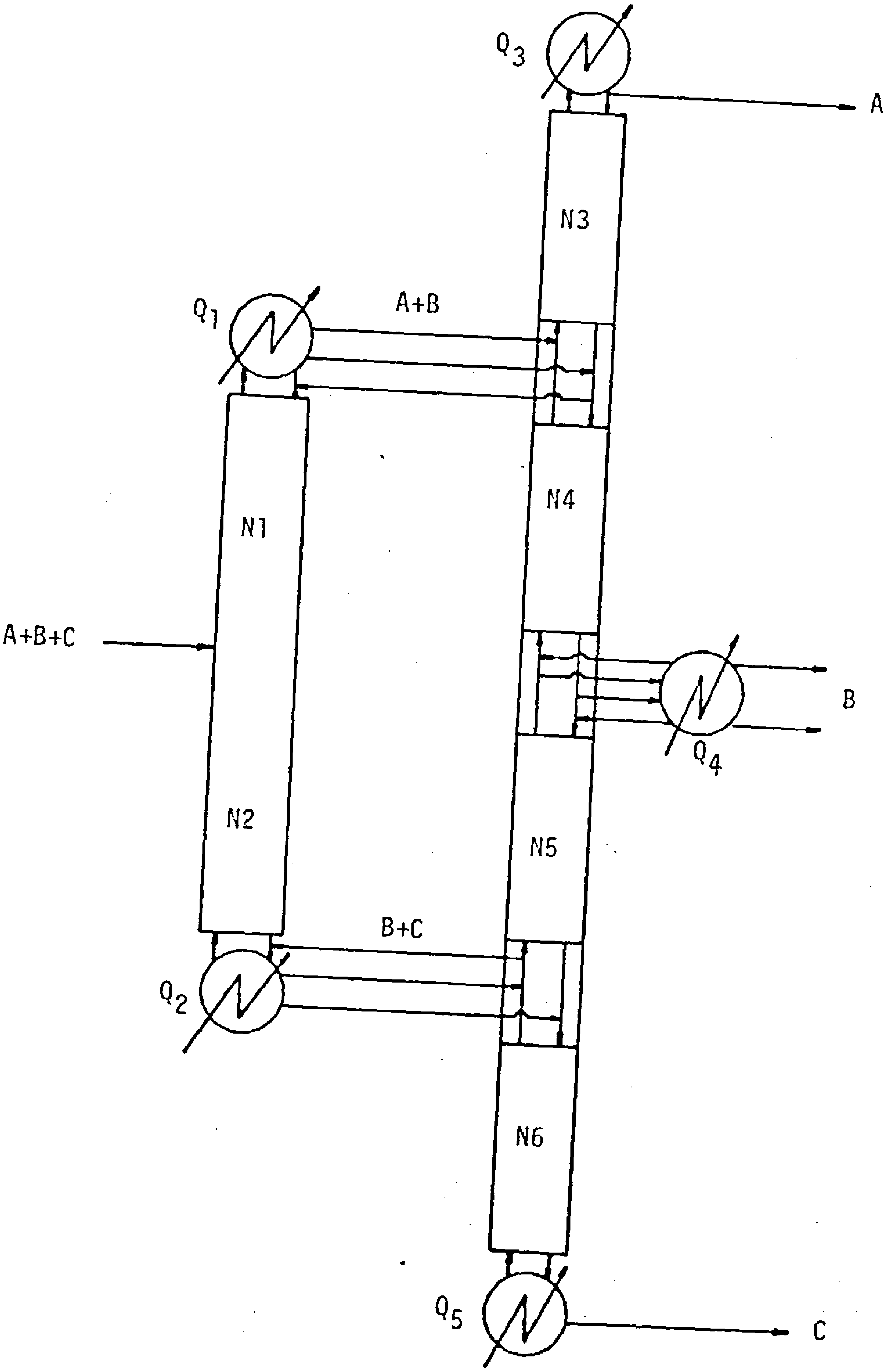

Figure 5.1

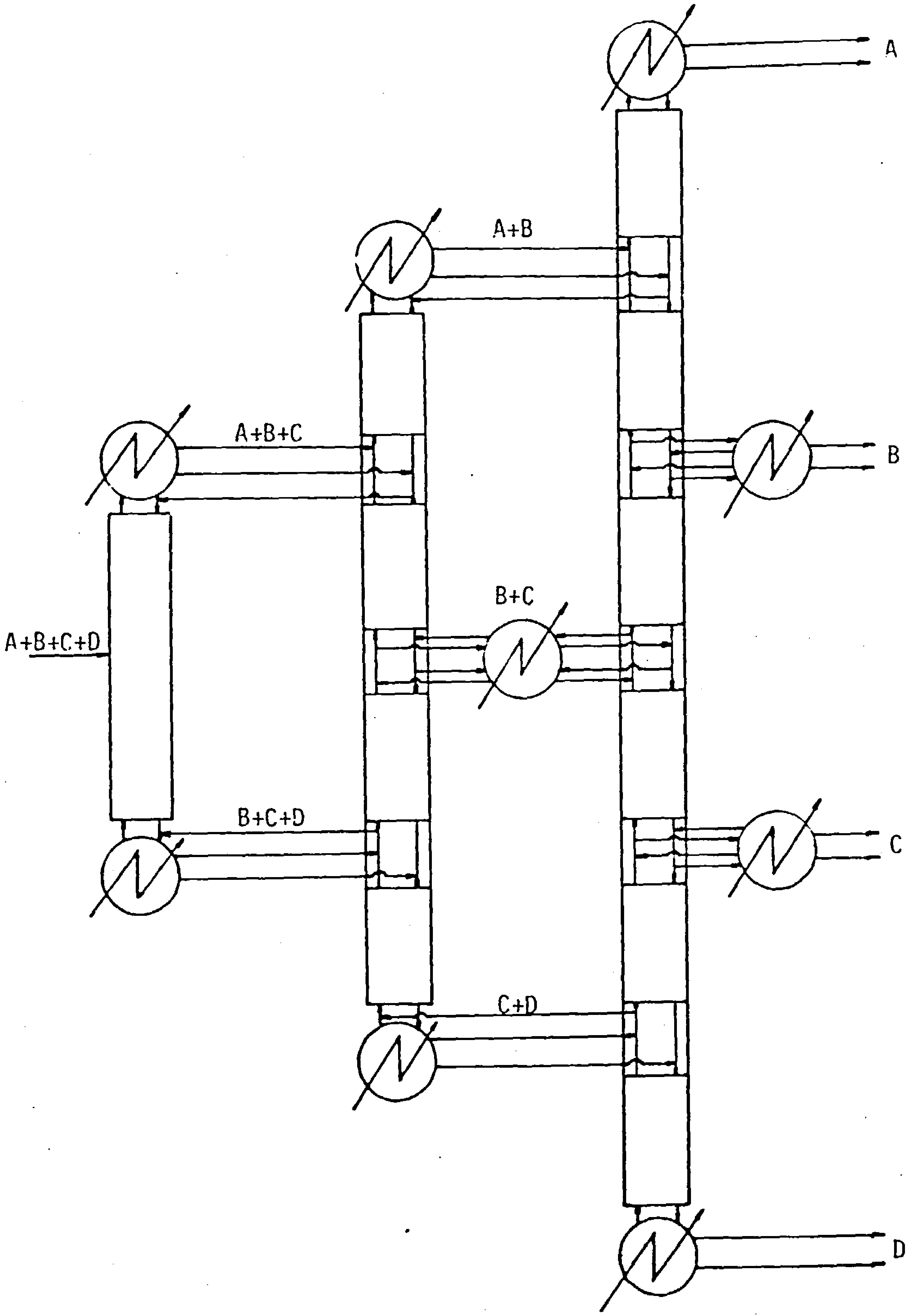

Figure 5.2

are included while in the previous general structure (5) only vapour from the condenser and liquid from the reboiler were taken into consideration as feeds into the second column. A general configuration of this type yields most of the particular configurations known, as can be observed in Figure 5.3. The conventional configuration shown in Figure 5.3a can be obtained from the general configuration shown in Figure 5.1 by deleting sections N5, N6 and obviously Q_5, the one shown in Figure 5.3b by deleting sections N3, N4 together with Q_3. The Petlyuk configuration shown in Figure 5.2c can be obtained if $Q_1 = Q_2 = Q_4 = 0$.

The top section of the second column, section 3(N3), in Figure 5.1 receives vapour from section 1(N1) and section 4(N4) and provides liquid to both sections, therefore it can be said that section 3 is shared by both columns and can be located at the top of the first column to perform the same function as the one performed at the top of the second column. The same is true for the bottom section of the second column, section 6(N6), in Figure 5.1 which provides vapour to section 5(N5) and section 2(N2) and receives liquid from both sections, therefore it can be located at the bottom of the first column. Figure 5.3d can be obtained if sections 3 and 6 of Figure 5.1 are transferred to the first column. Supposing that $n_4 = Q_1 = Q_2 = 0$ in Figure 5.3d, then the configuration shown in Figure 5.3e is obtained while Figure 5.3f is obtained if $n_5 = Q_1 = Q_2 = 0$.

These are only some of the possibilities and they are far from exhaustive but they show how any particular configuration can be obtained from the general system by deletion of column sections, heat transfer devices or interconnecting streams. Since these deletions correspond to setting variables to zero, the optimum configuration

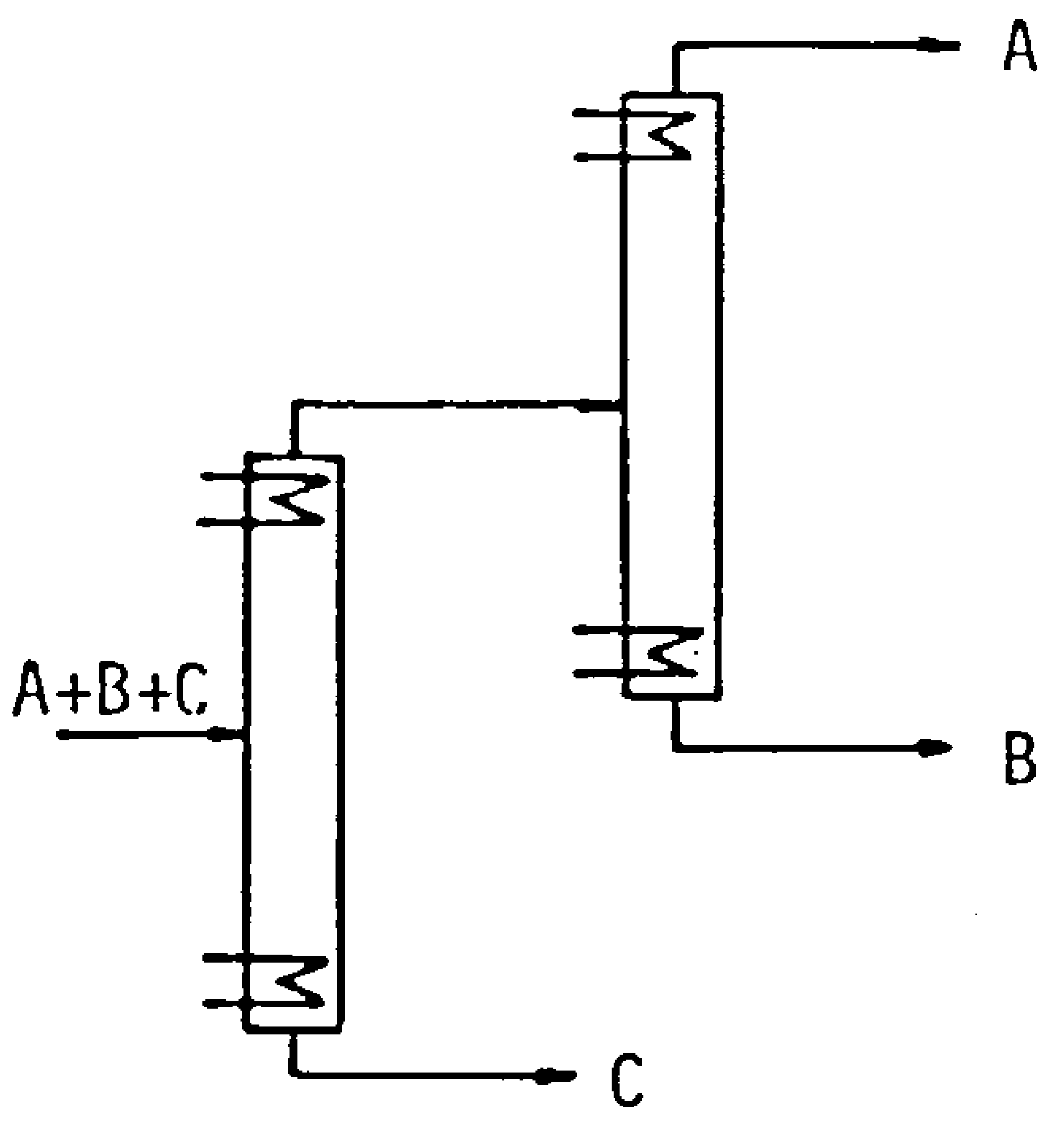

Figure 5.3a

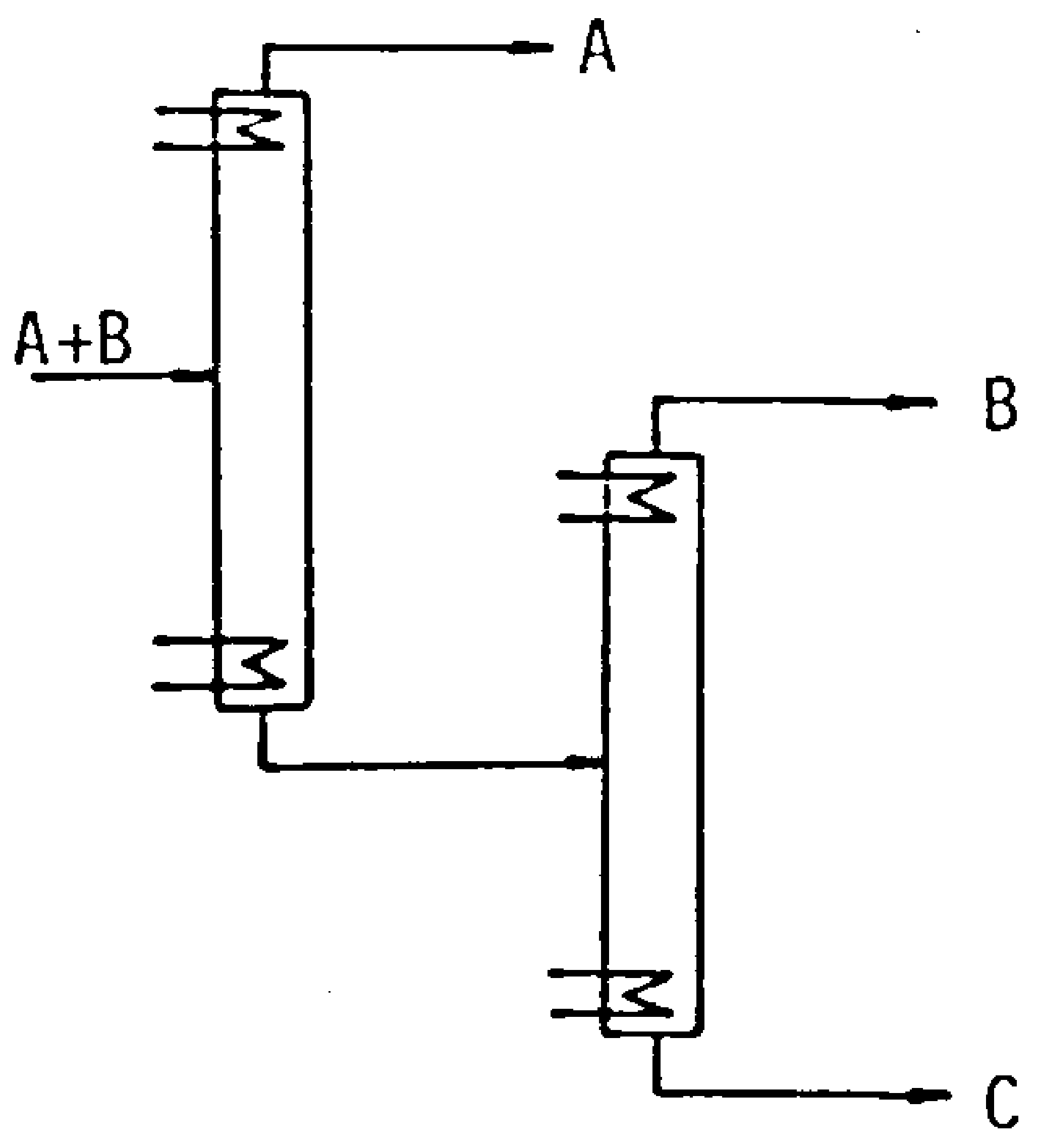

Figure 5.3b

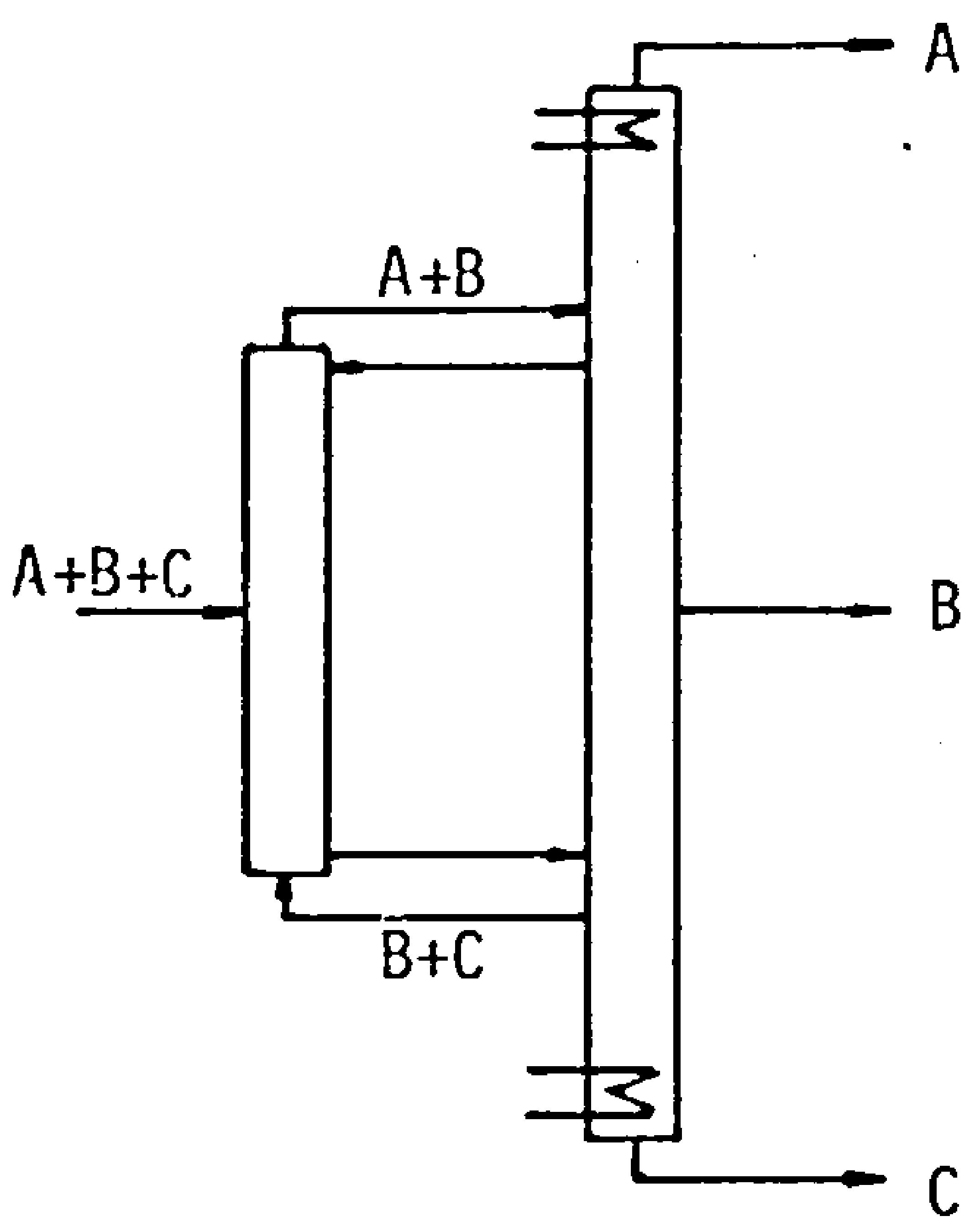

Figure 5.3c

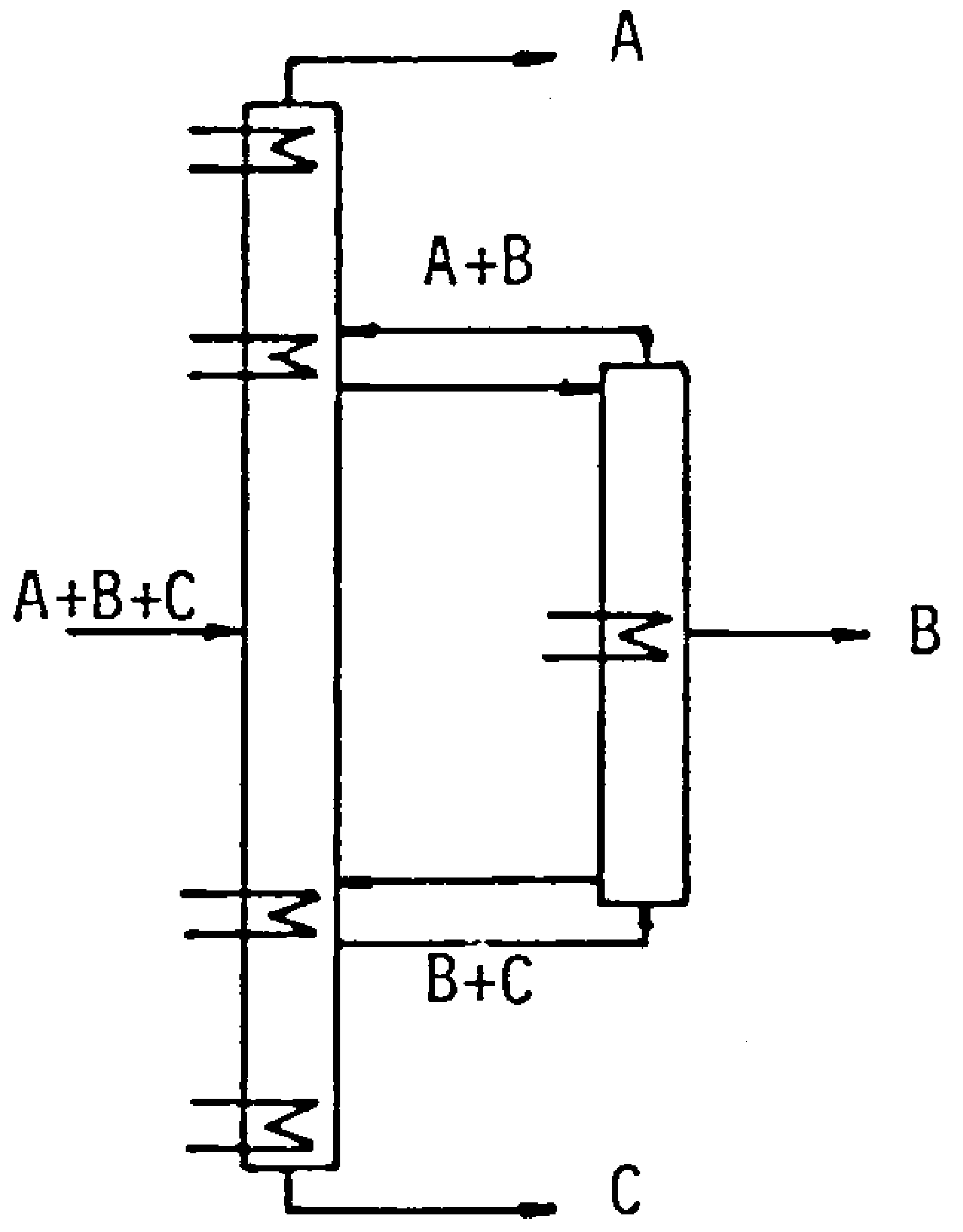

Figure 5.3d

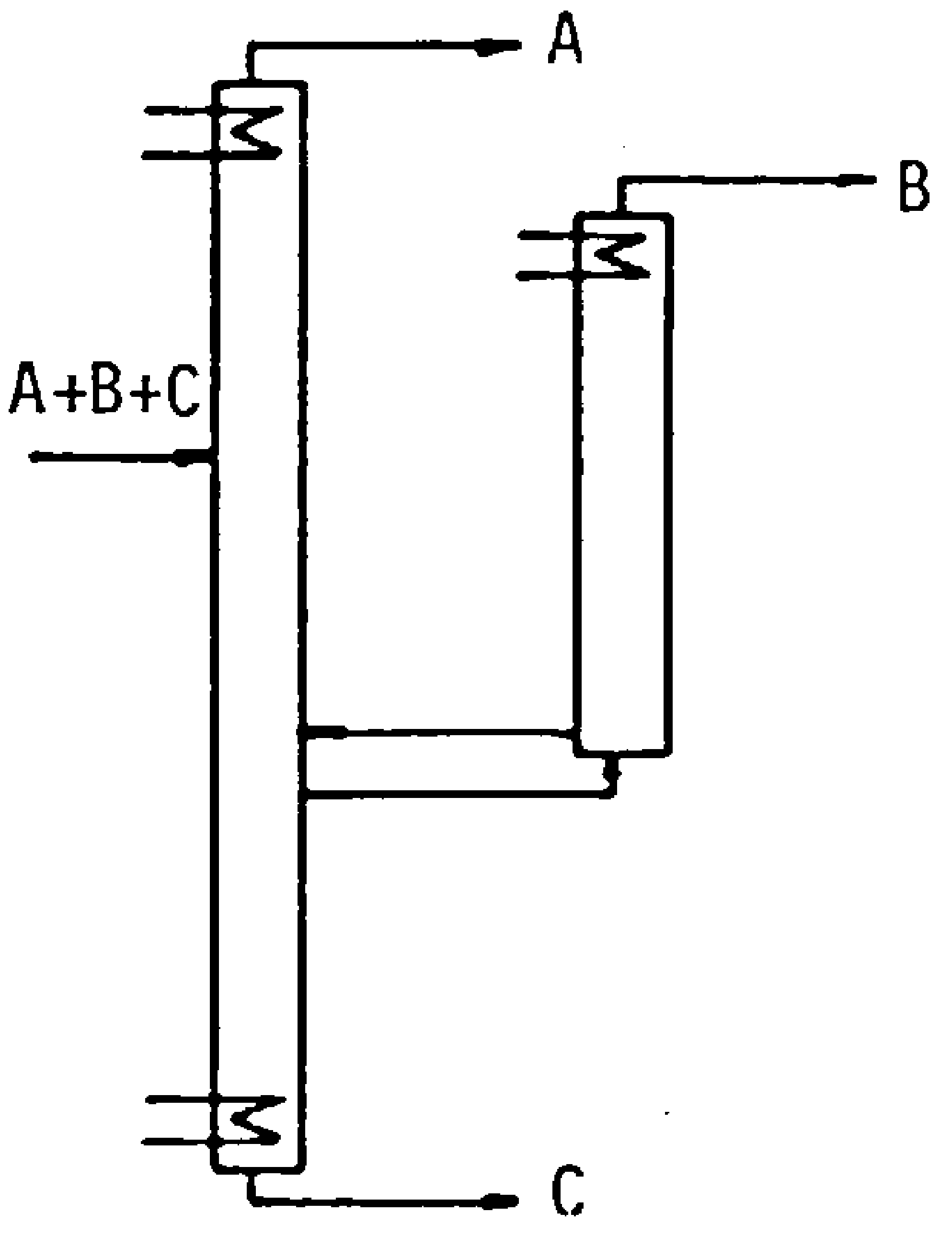

Figure 5.3e

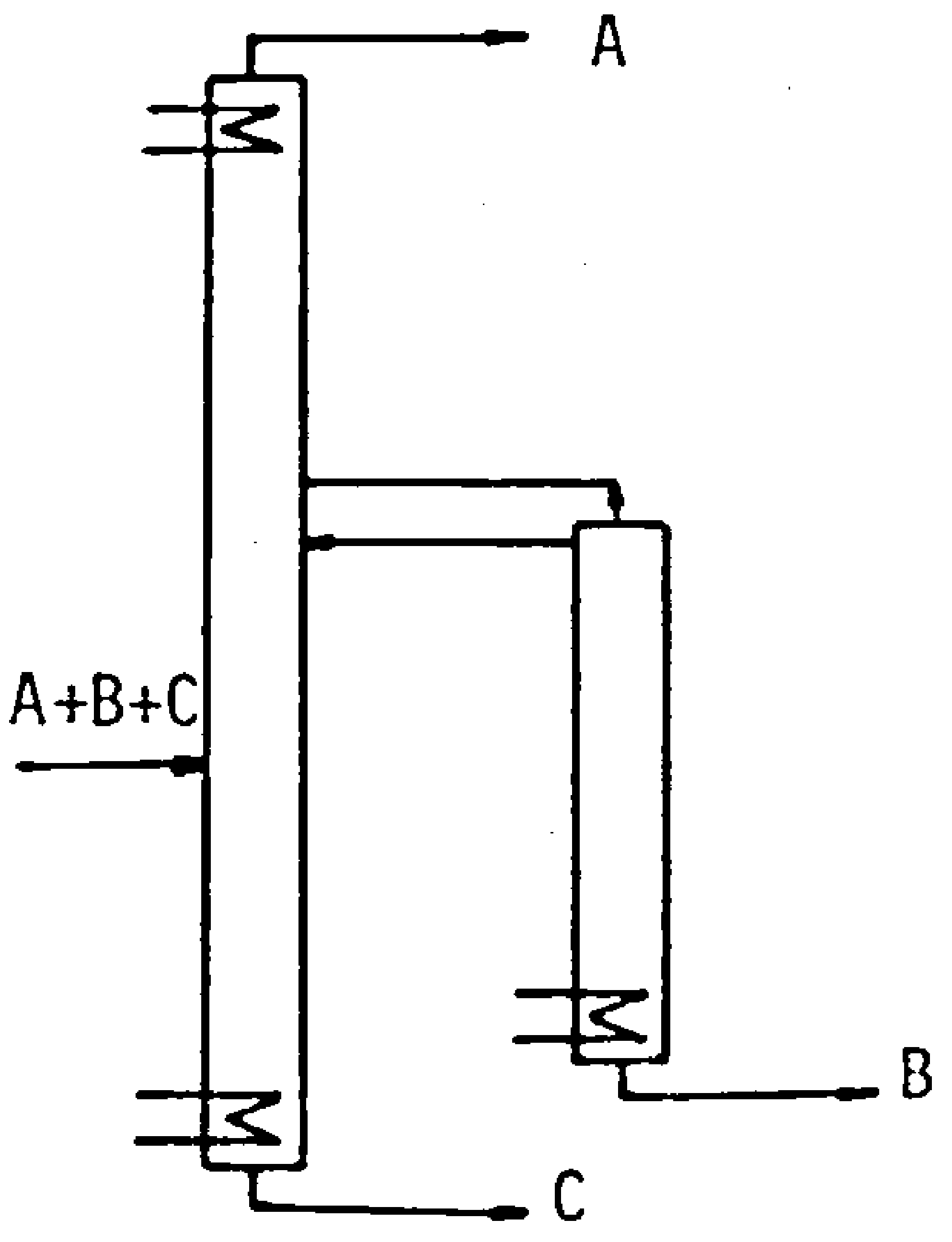

Figure 5.3f

can in principle be found by optimum design of the general system.

5.3 Optimum Design of the General Configuration for a Ternary Mixture

5.3a Formulation of the Optimum Design Problem

The general design problem consists of finding the configuration that will satisfy the required specifications on the products, with the minimum annual cost. Therefore the optimum design can be posed as an optimization problem as follows:

$$\begin{aligned} & \min F_{obj} = C(u,s) \\ \text{st} \quad & h(u,s) = 0 \\ & c^L < g(u,s,) < c^u \end{aligned} \qquad 5.P1$$

where the vector h describes the general system of distillation columns and the vector g represents the design and operational constraints. To reduce the size of the optimization problem the vector h of equality constraints is solved separately using the simulation algorithm presented in Chapter III, and the problem 5.P1 can be rewritten as:

$$\begin{aligned} & \min F_{obj} = C(u) \\ \text{st} \quad & c^L < g(u) < c^u \\ & b^L < u < b^u \end{aligned} \qquad 5.P2$$

where the dimension of the vector u is equal to the degrees of freedom of the general system of interlinked columns. Bounds on the variables were added in problem 5.P2 because the optimization algorithm, used to solve the problem, MINOS (38, 39) requires bounds on the optimization variables. The set of optimization variables include the stage numbers in all the sections, the size of the heat exchangers and the flow rates of the streams connecting the columns.

The objective function and its derivatives and the constraint functions and their derivatives are required by MINOS, and their calculation will be analyzed in the following sections.

5.3b The Objective Function

The calculate the annual cost of running the distillation system the procedure presented in Chapter IV is followed. Supposing that C_i^c is the annual cost of each section of a given column and that C_i^h includes the fixed and running cost of each heat exchanger, then the total cost of the system is defined as

$$C_{TOTAL} = \sum_{i=1}^{n} C_i^c + \sum_{i=1}^{m} C_i^h \tag{5.1}$$

where n is the number of different sections in the system

and m is the number of heat exchangers

Each column section has a different cost per plate depending on the diameter of the given section which is proportional to the internal vapour flow rate going through the section.

The final product flow rates are likely to be tightly bound by the specifications of purity and recovery, and their inclusion as variables to be chosen by the optimization algorithm is not expected to cause a considerable change in the solution especially for high recoveries and purities; therefore the flow rates of the final products were fixed to deal with a smaller number of optimization variables. For the same reason both columns were operated at atmospheric pressure, though the inclusion of the pressure in each column as an optimization variable should be included in any future work.

The objective function to be minimized is given in the following equation, where all the final products were liquid:

$$
\begin{aligned}
F = {} & C1 \times (X1 + X2) \; ((1 + X8)\,(X9 + X10))^{0.77} + \\
& + C1 \times X3 \times ((1 + X7) \times D)^{0.77} + \\
& + C1 \times X4 \times \big((1+X7) \times D - X9 \big) + \\
& + C_1 \times X5 \times \big((1+X7)\, D - X9 + X14 \times PH \big) + \\
& + C_1 \times X6 \times \big((1 + X7) \times D - X9 + X14 \times PH - X12 \big) + \\
& + C2F \times \big(X8 \times (X9 + X10) + X10 \big)^{0.6} + \\
& + C2R \times \big(X8 \times (X9 + X10) + X10 \big) + \\
& + C3F \times \big((1 + X8)\,(X9 + X10) + X12 - X13 \big)^{0.6} + \\
& + C3R \times \big((1 + X8)\,(X9 + X10) + X12 - X13 \big) + \\
& + C4F \times ((1+X7)\, D)^{0.6} + C4R \times (1 + X7)\, D + \\
& + C5F \times (X14 \times PH)^{0.6} + C5R \times X14 \times PH + \\
& + C6F \times \big((1 + X7) \times D + X14 \times OH + X13 - X9 - X12 \big) + \\
& + C6R \times \big((1 + X7) \times D + X14 \times PH + X13 - X9 - X12 \big)
\end{aligned}
\tag{5.2}
$$

where X1 is the real variable representing the number of stages in section 1
X2 is the real variable representing the number of stages in section 2
X3 is the real variable representing the number of stages in section 3
X4 is the real variable representing the number of stages in section 4
X5 is the real variable representing the number of stages in section 5
X6 is the real variable representing the number of stages in section 6
X7 = R2
X8 = R1
X9 = PV1(1)
X10 = PL1(1)
X11 = F1(2)
X12 = PV1(NT)
X13 = F1(NT-1)
X14 = PV2(N3+N4+1) / PH

where the nomenclature is the same as in Chapter III.

The first term in Equation (5.2) represents the cost function of the first column, the second, third, fourth and fifth terms represent the cost of the four sections of the second column, the sixth and seventh terms, the fixed and running costs of the top condenser of the first column, the eighth and ninth, the fixed and running costs of the reboiler of the first column, the tenth and eleventh, the fixed and running costs in the top condenser of the second column, the twelfth and thirteenth, the fixed and running costs in the intermediate condenser and the fourteenth and fifteenth terms, the fixed and running costs of the reboiler in the second column.

The fourteen partial derivatives of the objective function can be calculated analytically.

5.3c The Constraints

For given feeds the design variables available are the number of plates in each column section, the flow rates of interconnecting streams and the heat transfer rates to the exterior. The temperatures, compositions and flow rates of all streams leaving any of the plates in the system can be calculated using the performance algorithm given in Chapter III. From these state variables, any other property required for the design specifications can be obtained. Partial derivatives of the design specifications in relation to the optimization variables are needed. To calculate the finite-difference approximations to the partial derivatives, perturbations of 1 percent of the values of the continuous variables were made. But for the stage number the interpolation scheme presented in the previous chapter was used, where a base point is chosen and then perturbations are made to the adjacent integer number which brackets the corresponding non-integer number given by the optimization algorithm.

In addition to the design specification,physically realizable design and operating conditions impose further constraints. In particular the internal flow rates in each section of a given column must be non-negative, some of these extra constraints are also nonlinear. They are algebraic expressions of the design variables and therefore the partial derivatives can be calculated algebraically. Some of these operational constraints are the following expressions to avoid negative internal flow rates:

$$(1 + X7)\ D - X9 \geqslant 0 \qquad (5.3)$$

The vapour feed from the top of the first column into the second column cannot be bigger than the vapour internal flow rate in the top section of the second column, otherwise the vapour flow rate in the section immediately below is negative.

$$(1 + X8) \times (X9 + X10) - X13 \geqslant 0 \qquad (5.4)$$

The vapour fed at the bottom of the first column cannot be bigger than the internal vapour flow rate in the top section of the first column, considering that the intermediate feed into the first column is liquid.

$$(1 + X7)\ D - X9 + (1 + X14)\ PH + X13 - X12 \geqslant 0 \qquad (5.5)$$

The internal vapour flow rate in the bottom section of the second column has to be positive.

$$X7 \times D + X10 - X11 + (X14 - 1)\ PH \geqslant 0 \qquad (5.6)$$

The internal liquid flow rate in the section below the intermediate condenser, in the second column, cannot be negative.

$$X9 + X10 + X12 - X11 - X13 - F \geqslant 0 \qquad (5.7)$$

The liquid product from the reboiler of the first column cannot be negative. F is the flow rate of the feed to the system.

The optimization algorithm, MINOS, will search for the minimum of the annual cost of running the distillation system in the feasible space defined by the design and operational constraints. The optimization variables were scaled in such a way that all their values are between 0 and 10.

5.4 Numerical Examples

A liquid ternary mixture of 500 lb moles/hr is separated in each one of its components in the following examples, where a minimum purity of 90 percent in the three products is required.

As the steam price is the dominant term in the total cost, the optimization of the general configuration is carried out at normal steam prices in the first example and at steam prices ten times less than the normal one in the second example. The third example is run at normal steam price, but changing the rate of interest and the rate of inflation, using as the initial point the solution of the first example. The information that follows is common to the three first examples:

Component	Mole fraction	Relative volatility	Temperature (°C)
Dichloromethane	0.3	4.9	41
Chloroform	0.4	1.8	61
Carbon tetrachloride	0.3	1.0	76

U_c = 175 BTU/(^oF hr ft^2)

U_R = 150 BTU/(^oF hr ft^2)

D = 150 moles lb/hr

PH = 200 moles lb/hr

In the fourth example a ternary mixture composed of benzene, toluene and o-xylene is separated into its components, where the temperature at which heat has to be exchanged is higher than in the previous examples.

The values of the flow rates given in the examples as X9, X10, X11, X12 and X13 are given in moles lb/hr based on a feed to the system of 100 moles lb/hr.

Example I

i = f = 0.0 lifetime = 10 years

Table 6.1a Normal steam price

	Initial values	Sol point	Lower bound	Upper Bound
X1	5	8	2.0	20.0
X2	5	9	1.0	25.0
X3	7	9	2.0	18.0
X4	7	8	1.0	20.0
X5	12	16	1.0	25.0
X6	12	15	2.0	25.0
X7	2.0	1.035	0.01	4.0
X8	0.8	0.460	0.01	3.0
X9	32.0	27.0	0.10	90.0
X10	8.0	15.0	0.10	70.0
X11	10.0	0.1	0.10	60.0
X12	20.0	0.1	0.10	100.0
X13	10.0	61.32	0.10	90.0
X14	1.8	1.0	0.01	4.0
Total cost (£/hr)	15.6865	12.000		

Iterations	25
Function and gradient calls	115
Time (CDC 6500) secs	825

Table 6.1b Costs (£/hr)

Columns	1.098
Exchangers	0.595
Steam	9.507
Cooling water	0.800
Total	12.000

Example II

i = f = 0.0 lifetime = 10 years

Table 6.2 Steam price ten times less than normal price

	Initial values	Sol values	Lower bound	Upper bound
X1	8.0	7.0	2.00	20.0
X2	9.0	12.0	2.00	25.0
X3	15.0	-	2.00	18.0
X4	12.0	-	1.00	20.0
X5	16.0	14.0	1.00	25.0
X6	14.0	12.0	2.00	25.0
X7	1.355	0.01	0.01	4.0
X8	0.510	1.3951	0.01	3.0
X9	40.0	0.1	0.10	90.0
X10	0.2	30.0	0.10	70.0
X11	0.2	0.1	0.10	60.0
X12	0.2	0.1	0.10	100.0
X13	60.0	71.85	0.10	90.0
X14	1.07	2.15	0.01	4.0

Iterations	9
Function and gradient calls	16
Time (CDC 6500) secs	112

Example III

i = 0.1 f = 0.1 lifetime = 10 years

Table 6.3 Normal steam price

	Initial values	Sol values	Lower bound	Upper bound
X1	8	7.5	2.0	20.0
X2	9	9.5	2.0	25.0
X3	8	6.7	2.0	18.0
X4	8	7.3	1.0	20.0
X5	16.0	14.0	1.0	25.0
X6	15.0	12.0	2.0	25.0
X7	1.1	1.130	0.01	4.0
X8	0.5	0.466	0.01	3.0
X9	27.0	28.00	0.10	90.0
X10	15.0	13.00	0.10	70.0
X11	0.02	0.01	0.10	60.0
X12	0.02	0.01	0.10	100.0
X13	0.6	0.60106	0.10	90.0
X14	1.0	1.03	0.01	0.01
Cost(£/hr)	14.130	13.550		

Iterations 6

Function and gradient calls 23

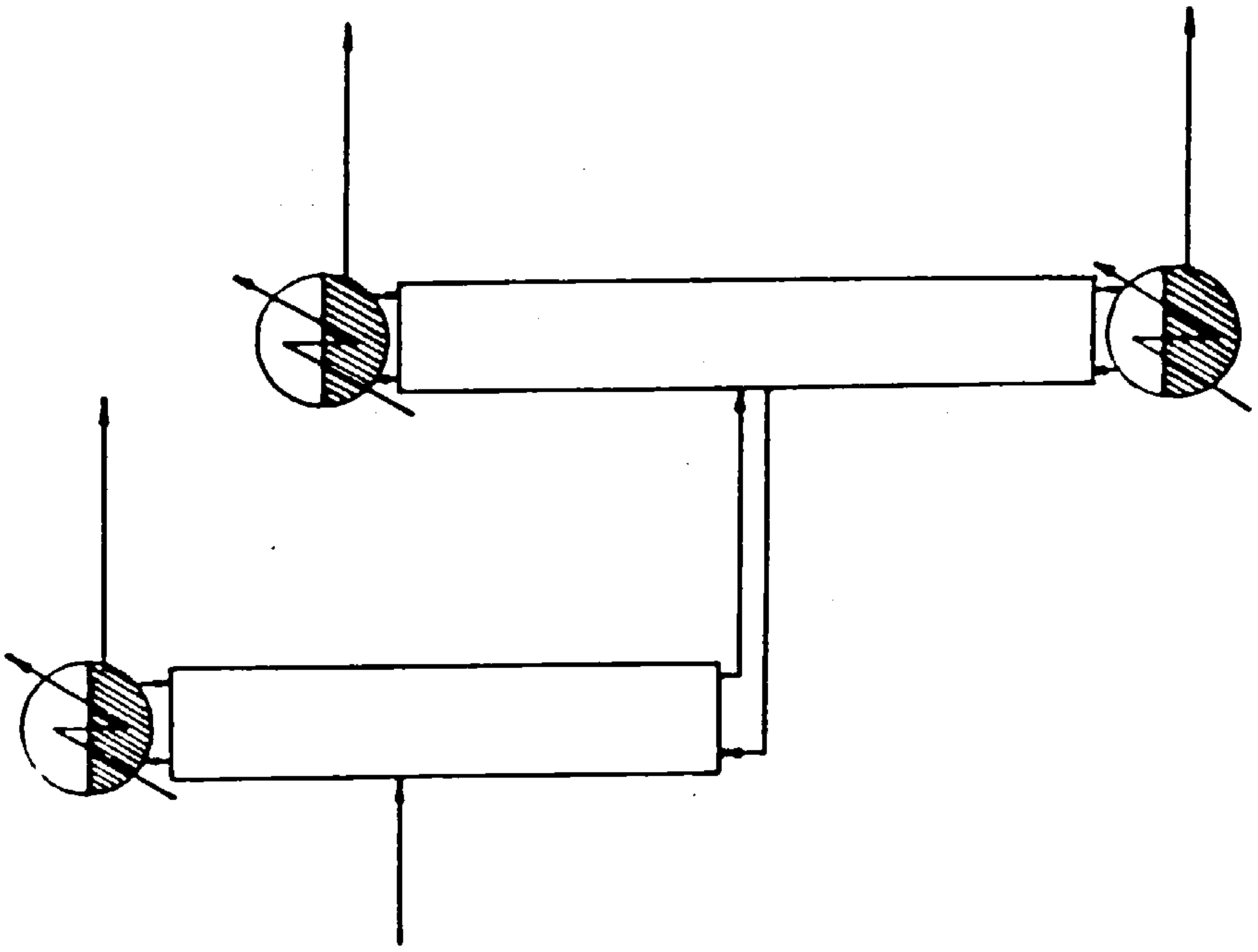

Figure 5.b - Example II

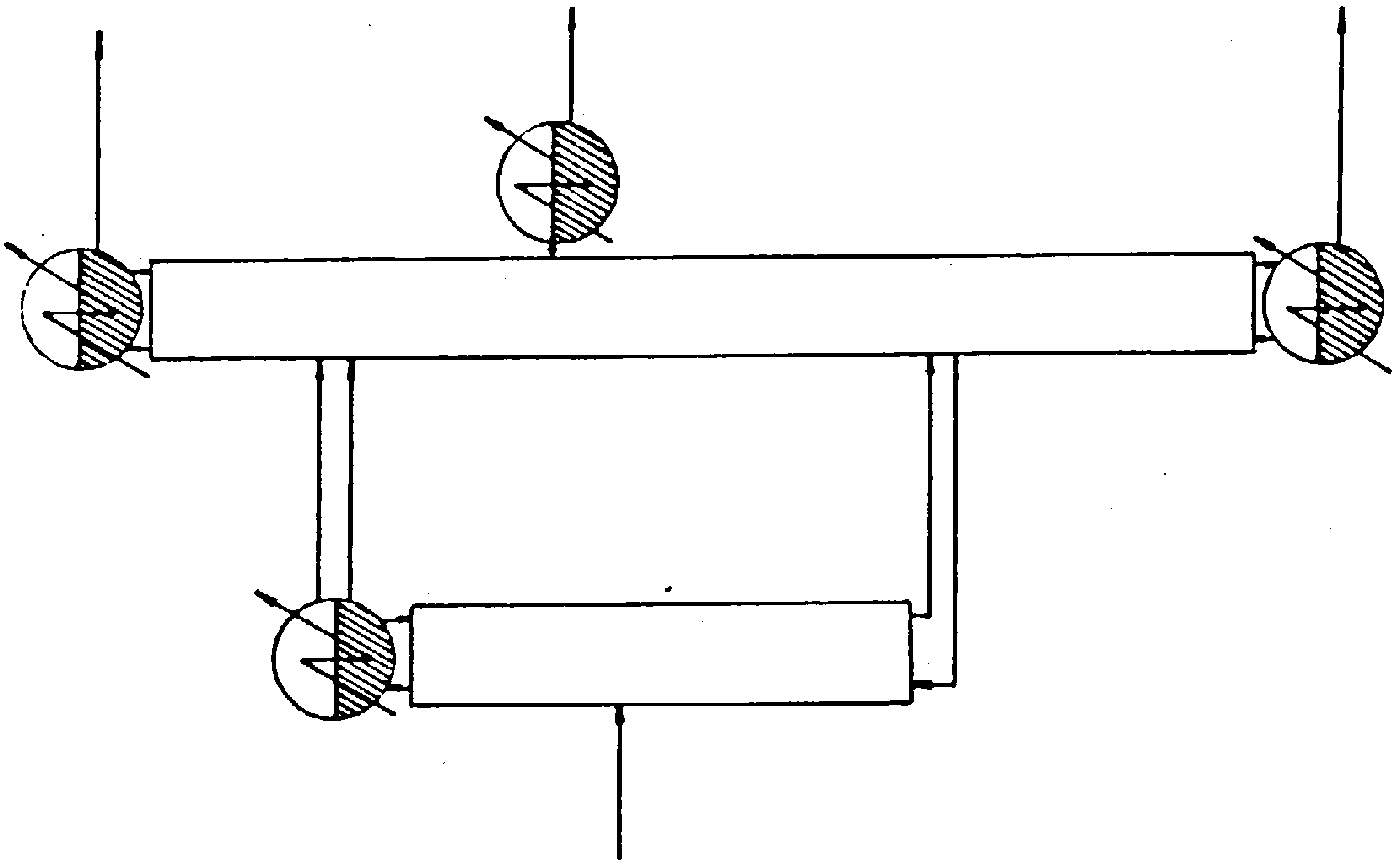

Figure 5.a - Examples I and III

Example IV

Component	Mole fraction	Relative volatilities	Temperature (^{o}C)
Benzene	0.3	6.7	80
Toluene	0.4	2.4	111
O-xilene	0.3	1.0	174

i = 0.1 f = 0.1 lifetime = 10 years

Table 6.4 Normal steam price

	Initial values	Sol values	Lower bound	Upper bound
X1	8	8	2.00	20.00
X2	10	10	3.00	25.00
X3	7	8	2.00	18.00
X4	7	7	1.00	20.00
X5	14	15	1.00	25.00
X6	12	12	2.00	25.00
X7	0.8	1.68	0.01	4.00
X8	0.3	0.3	0.01	3.00
X9	27.0	43.0	0.10	90.00
X10	23.0	14.0	0.10	70.00
X11	5.0	0.1	0.10	60.00
X12	5.0	0.1	0.10	100.00
X13	5.0	0.1	0.10	90.00
X14	1.0	0.01	0.01	4.00
Cost(£/hr)	30.275	24.650		

Iterations	14
Function and gradient calls	58
Time (CDC 6500) secs	487.5

U_c = 100 BTU/(^{o}F hr ft^2)

U_R = 80 BTU/(^{o}F hr ft^2)

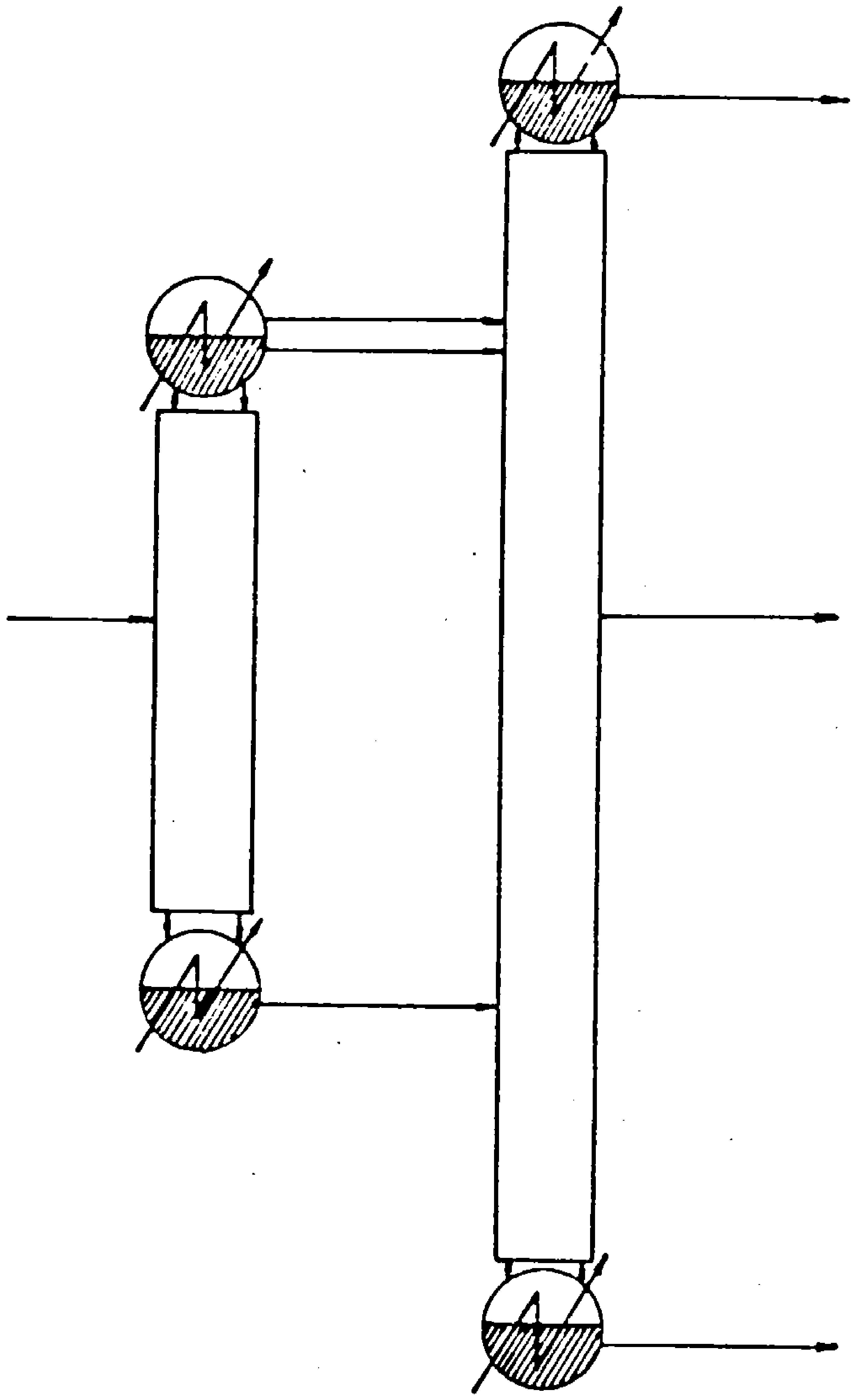

Figure 5.c - Example IV

5.5 Comments

On the calculation of the partial derivatives of the purities in relation to the optimization variables fifteen simulations of the distillation system are required to obtain the finite-difference approximations, considering that there are fourteen optimization variables. In the simulation of the distillation system Broyden's method was used to update the four torn variables. In the first simulation the Jacobian was approximated using finite-differences but in the following fourteen simulations, the Jacobian of the previous solution was used as the initial Jacobian of the current simulation. Convergence was normally obtained in less than the 5 iterations which are required for the finite-difference approximation of the Jacobian, thus increasing the efficiency of the method. This result is not surprising given the fact that the solutions of the fifteen simulations are very near due to the fact that the steps on the optimization variables are small.

Using realistic cost, it can be seen that steam costs dominate and economy indicates the coupled system. In the first example where the annuity is 0.1 and the normal steam price is used, a coupled system is obtained as the optimum design. The reboiler at the bottom of the first column is deleted because the constraint expressed in Equation (5.4) is active, therefore all the internal vapour in the first column is provided by the vapour feed at the bottom of the first column. Indeed this extraction of vapour from the second column improves the separation. By providing vapour to the bottom of the first column the internal vapour flow rate in the bottom section of the second column increases, thus the slope of the operating line (L/V) decreases and the distance between the operating line and the equilibrium line increases. Therefore the change of composition from plate to plate is bigger in this section of the column, where the most difficult part of the separation is carried out ($\alpha = 1.8$). That is why vapour is produced at the highest level of temperature when it could have been produced at a cheaper cost at the bottom of the first column. One also has to consider that in this example the bubble point (BP) of the heaviest product is $76^{o}C$, and hence the temperature of the bottom plate of the first column is an intermediate value between $61^{o}C$ and $76^{o}C$. Clearly there cannot be much difference in price for this specific example between heating in one or the other reboilers, thus the intermediate flow rates are chosen to improve the separation.

On the other hand, it is advantageous to provide cooling at the top of both columns, given the fact that the temperature level of the lightest component ($41^{o}C$) is very near the temperature level of the cooling water, thus requiring a considerable amount of cooling water and area for heat to be exchanged in the top section of the second column. Therefore the possibility of cooling at the other condenser at higher temperature levels is used; the intermediate condenser in the second column serves solely to condense the chloroform product.

Drastic reduction of steam costs, ten times less than the real one, as shown in Example II, tips the balance towards saving capital costs and a more conventional two columns system results. All the steam is supplied at the bottom of the second column to decrease the slope of the operating line below the feed in the second column which carries out the more difficult separation. This simplified configuration satisfies one of the most popular heuristic rules: leave the most difficult separation for last.

The third example is run at normal steam prices, where the annuity is 0.24,therefore the fixed costs are more important than in the first example and the total number of stages at the solution is less than in the first example.

In the fourth example a mixture of benzene, toluene and o-xylene is separated into its components. In the initial guess an intermediate condenser in the second column is included to condense the toluene product. In this example the BP of the three pure components at atmospheric pressure are 80^{0}C, 111^{0}C and 174^{0}C. In the optimum design, the intermediate condenser is deleted and the toluene product obtained directly from the internal liquid stream. This liquid is provided by increasing the amount of vapour being condensed at the top of the second column, as can be seen for the increase in the variable X7 representing the reflux ratio in the top condenser. Steam price is more expensive than in the previous case due to the higher temperature levels at which steam has to be provided, therefore it has to be used in an efficient way. It is unlikely that the savings in cooling water that could be expected if cooling were carried out in the intermediate condenser could compensate for the decrease in the slope of the operating lines (L/V) in the top of the second column, whereby the operating line is nearer to the equilibrium line and the change in the composition from step to step is smaller.

When the reflux ratio approaches the minimum reflux ratio the stage numbers might increase very quickly. In the case of a single column, as pointed out in the previous chapter, the use of a reasonable lower bound on the reflux ratio can increase the efficiency of the optimization algorithm. In the general configuration, the reflux ratios should be able to approach zero because one doesn't know in advance which heat exchangers and column sections might be deleted. For these reasons, in some cases the optimization algorithm might fail to find a solution because the reflux ratio reaches the lower bound value, zero, and the stage numbers increase up to the upper bound values, getting stuck at these values.

It might well be the case that the temperature level of the condensers is high enough to provide heat to other processes in the plant as pointed out by Dunford and Linnhoff (65). Siirola (66) has evaluated the possibility of steam generation from the condensers of distillation systems. Steam could be available from other processes and its price is likely to be much less than the normal steam price. These options have not been tried but only a change in the cost function of the general configuration is required.

CHAPTER VI

CONCLUSIONS

The aim of this work has been to develop an algorithm for the optimal design of systems of distillation columns for the separation of mixtures into several fractions. Given the limitations on the type of distillation columns and their interconnections allowed in most of the published work on the synthesis of such systems, the approach presented in Chapter V is followed. A general structure is proposed, which contains most of the known arrangements between columns and the optimal design is obtained by optimizing this general structure.

To make this approach practicable, a very fast method is needed to compute the performance of the system under given conditions. Shortcut methods were therefore investigated, but in the proposed method, described for a single column in Chapter II, a plate to plate simulation is carried out under the assumption that internal flow-rates and relative volatilities remain constant. With these simplifications, the computing time required for the optimum design of a single distillation column is not excessive, as can be seen in the examples presented in Chapter IV and there are no restrictions on the type of columns to be designed optimally; any number of feeds, products and heat exchangers can be dealt with. The extension of this approach to deal with the simulation of the general configuration is described in Chapter III, and again it can be seen from the results that the method is very efficient.

Release of the simplifying assumptions requires only minimal modifications of the simulation subroutine, with inclusion of calls to an appropriate physical property package, but of course, accuracy must be paid for in extra computing time.

The interpolation scheme developed to deal with the integer variables, presented in Chapter IV, permits the simultaneous optimization of all the design variables of a single distillation column, a design problem that has not been solved up to now, despite the importance that the optimum design of distillation columns has had in the chemical engineering literature. This approach makes it possible to use any standard nonlinear programming algorithm for the optimization. In the present work, the MINOS package was used, which proved adequate for single column problems, but failed rather frequently when applied to the general configuration.

Thus, overall the approach is completely satisfactory for the optimal design of single distillation columns. If the flowsheet is chosen on other grounds, and the cost of the utilities is known, the algorithm presented in Chapter IV can be used to design each of the columns separately. Therefore this approach is a very useful tool for a designer in need of a fine-tuning optimization method. In the examples presented in Chapter IV, the annual cost of running the distillation column was minimized but, in principle, any other objective function could be used.

Most of the work on synthesis of distillation columns does not include mass recycle between columns except when an external substance to the mixture being separated is used. It has been shown in Chapter V that the use of recycle streams from the second column into the first column improves the performance of the second column thereby increasing the efficiency of the distillation system. It is therefore important to develop synthesis methods which allow such coupling, but will also generate simpler systems when these are more economic, as in the approach proposed here. There has been significant progress in the nonlinear programming field during the last several years, and if the theory and algorithms developed are incorporated in good software packages, the robustness and efficiency of the simultaneous optimization of general structures is also likely to improve.

The magnitude of the computing task rapidly increases with the number of fractions to be produced from the given mixture, and the size of the system to be optimized will be limited by the storage and time required by the simulation of the system. However, the capacity of computers is constantly increasing with time, allowing a corresponding increase in the complexity of the system to be optimized.

Thus the basic approach seems well justified by the results obtained so far, and it will be worth exploring other facets of the problems.

Work needs to be done on the flexibility, operability and controllability of the coupled system of distillation columns. It is reassuring that the control studies done by Doukas and Luyben (27) have shown that satisfactory control schemes can be devized and that the coupled systems are stable under moderate disturbances.

To deal with the problem of energy integration with the rest of the process, it is also clearly necessary to include the pressures at which the columns are operated as design variables. As far as the numerical techniques are concerned, this is a trivial extension, which merely increases the size of the optimization problem. However, running columns at different pressures has implications in terms of pumps or compressors, and hence possibly of layout, which in turn complicates formulae for costs. Allowing for direct thermal coupling between columns and the use of heat-pump cycles causes further complications. The problem is then to devize a configuration sufficiently general to include most useful solutions, yet sufficiently simple to yield a tractable optimization problem.

From these considerations it seems unlikely that the approach can be extended to deal comprehensively with the heat integration problem, but with the inclusion of variable pressures the algorithm should again prove a useful tool to assist the designer in developing solutions.

REFERENCES

1. Friday, J.R. and B.D. Smith, "An Analysis of the Equilibrium Stage Separation Problem. Formulation and Convergence", 10, 698 (1964).
2. Amundson, N.R. and A.J. Pontinen, "Multicomponent Distillation Calculation on a Large Digital Computer", Ind.Eng.Chem., 50, 730 (1958).
3. Wang, J.C. and G.E. Henke, "Tridiagonal Matrix for Distillation", Hydrocarbon Process, 45, 155 (1966).
4. Murtagh, B.A. and R.W.H. Sargent, "The Design of Plate Distillation Columns for Multicomponent Mixtures", Trans.Instn.Chem.Engrs., 47, T85, (1969).
5. Gaminibandara, K. and R.W.H. Sargent, "Optimum Design of Plate Distillation Columns", in L.C.W. Dixon (Ed), "Optimization in Action", (Academic Press, London 1976).
6. Stainthorp, F.P. and P.A. Whitehouse, "General Computer Programs for Multi-Stage Counter-Current Separation Problems", "Part I : Formulation of the Problem and Method of Solution", I.Chem.E. Symposium Series No. 23, p.181 (1967).
7. Stainthorp, F.P., P.A. Whitehouse and H. Kandela, "Part II: Description of the Computer Program", I.Chem.E. Symposium Series No. 23, p.189 (1967).
8. Tierney, J.W. and J.L. Yanosik, "Simultaneous Flow and Temperature Correction in the Equilibrium Stage Problem", A.I.Ch.E.J., 15, 897 (1969).
9. Orbach, O., C.M. Crowe and A.I. Johnson, "Multicomponent Separation Calculations by the Modified Form of Newton's Method", The Chem. Eng. J., 3, 176 (1972).

10. Tomich, J.F., "A New Simulation Method for Equilibrium Stage Processes", A.I.Ch.E.J., 16, 229 (1970).

11. Broyden, C.G., "A Class of Methods for Solving Nonlinear Simultaneous Equations", Math.Comput., 19, 577 (1965).

12. Naphtali, L.M. and D.P. Sandholm, "Multicomponent Separation Calculation by Linearization", A.I.Ch.E.J., 17, 148, (1971).

13. Ishii, Y. and F.D. Otto, "A General Algorithm for Multistage Multicomponent Separation Calculations", The Can.J.Chem.Eng., 51, 601 (1973).

14. Shewchuk, C.F. and H.P. Hutchison, "A Computational Method for Multiple Distillation Towers", Trans.Instn.Chem.Engrs., 52, 325 (1974).

15. Shewchuk, C.F., "Extension of the Quasi-Linear Method for Non-Ideal Distillation Calculations", Trans.Instn.Chem.Engrs., 55, 130 (1977).

16. Barret, A. and J.J. Walsh, "Improved Chemical Process Simulation using Local Thermodynamic Approximations", presented at CACE '79, Montreaux, 1979.

17. Boston, J.F. and S.L. Sullivan, "An Improved Algorithm for Solving the Mass Balance Equations in Multistage Separation Processes", Can.J.Chem.Eng., 50, 663 (1972).

18. Boston, J.F., "Inside-Out Algorithms for Multicomponent Separation Process Calculations", in R.G. Squires and G.V. Reklaitis (Eds), "Computer Applications to Chemical Engineering", ACS Symposium Series No. 124, pp.135 (1980).

19. Hlavacek, V. and P. Van Rompay, "Simulation of Countercurrent Separation Processes via Global Approach". Submitted for the 14th European Symposium CHEM CONTROL 81 (Austria, Vienna, Sept., 1981).

20. Bending, M.J. and H.P. Hutchinson, "The Calculation of Steady State Incompressible Flows in Large Networks of Pipes", Chem. Eng.Sci., 28, 1857 (1973).

21. Kuricek M., V. Hlavacek and F. Prochaska, "Global Modular Newton-Raphson Technique for Simulation of Interconnected Plant Applied to Complex Rectification Columns", Chem.Eng.Sci., 31, 177 (1976).

22. Browne, D.W., Y. Ishii and F.D. Otto, "Solving Multicomponent Equilibrium Stage Operation by Total Linearization", The Can.J. Chem.Eng., 55, 307 (1977).

23. Holefing, B.S. and J.D. Seader, "A Modified Naphtali-Sandholm Method for General Systems of Interlinked Multistage Separators", A.I.Ch.E.J., 24, 1131 (1978).

24. Hidalgo, R.A., Correa, V.A., A. Gomez and J.D. Seader, "An Optimal Arrangement of Simultaneous Linearized Equations for General Systems of Interlinked Multistaged Separators", A.I.Ch.E.J., 26, 585 (1980).

25. Schubert, L.K., "Modification of a Quasi-Newton Method for Nonlinear Equations with a Sparse Jacobian", Math.Comp., 24, 27 (1970).

26. Curtis, A.R., M.J.D. Powell and J.K. Reid, "On the Estimation of Sparse Jacobian Matrices", J.Inst.Math.Applics., 13, 117 (1974).

27. Doukas, N.P. and W.L. Luyben, "Control of an Energy-Conserving Prefactionator/Sidestream Column Distillation System", Ind.Eng. Chem.Process.Des.Dev., 20, 147 (1981).

28. Fenske, M.R., Ind.Eng.Chem., 24, 482 (1932).

29. Cerda, J. and A.W. Westerberg, "Short-Cut Methods for Complex Distillation Columns", "Part I - Minimum Reflux", Report No. DRC-06-10-79, "Part II - Number of Stages and Feed Tray Location", Report No. DRC-06-11-79, Design Research Center, Carnegie-Mellon University, Pittsburgh, 1979.

30. Boston, J.F., "Algorithm for Distillation Calculations with Bounded-Variable Design Constraints and Equality or Inequality Constrained Optimization", presented at the A.I.Ch.E. Distillation Symposium, Houston (1979).

31. Powell, M.J.D., paper presented at 1977 Dundee Conference on Numerical Analysis.

32. Peters, M.S. and K.D. Timmerhaus, "Plant Design and Economics for Chemical Engineers", McGraw-Hill (1980).

33. Aitken, A.C., "On Bernoulli's Numerical Solution of Algebraic Equations", Proc. Royal Soc. Edinburgh, 46, 289 (1925).

34. Seffensen, J.F., "Remarks on Iterations", Skand.Aktuar.Tidskr., 16, 64 (1933).

35. Wegstein, J.H., "Accelerating Convergence on Iterative Processes", Comm. ACM, 1, 9 (1958).

36. Orbach, O. and C.M. Crowe, "Convergence Promotion in the Simulation of Chemical Processes with Recycle - The Dominant Eigenvalue Method", Can.J.Chem.Eg., 49, 509 (1971).

37. Perkins, J.D. and S.R. Metcalfe, "Information Flow in Modular Flowsheeting System", Trans.I.Chem.E., 56, 210 (1978).

38. Murtagh, B. and M.A. Saunders, "Large Scale Linearly Constrained Optimization", Math.Prog., 14, 41 (1978).

39. Murtagh, B.A. and M.A. Saunders, "The Implementation of a Lagrangian-based Algorithm for Sparse Nonlinear Constraints", System Optimization Laboratory Report SOL 80-1, Stanford University, California.

40. Happel and Jordan, "Chemical Process Economics", Marcel Dekker, (1975).

41. Shaham, M. and R.L. Motard, "Applications of the Theory of Linear Recycle Systems", presented at the 78th A.I.Ch.E. National Meeting, Salt Lake City, Utah (1974).

42. Hlavacek, V., "Journal Review: Synthesis in the Design of Chemical Processes", Computers and Chemical Engineering, 2, 67-75 (1978).

43. Westerberg, A.W., "A Review of Process Synthesis", in R.G. Squires and G.V. Reklaitis (Eds), "Computer Applications to Chemical Engineering", ACS Symposium Series No. 124, pp.53-87 (1980).

44. Nishida, N., G. Stephanopoulos and A.W. Westerberg, "A Review of Process Synthesis", Report No. DRC-06-17-80, Design Research Center, Carnegie-Mellon University, Pittsburgh 1980.

45. Thompson, R.W. and C.J. King, "Systematic Synthesis of Separation Schemes", A.I.Ch.E.J., 18, 941 (1972).

46. Hendry, J.E. and R.R. Hughes, "Generating Separation Process Flowsheets", Chem.Eng.Prog., 68, 71-76 (1972).

47. Westerberg, A.W. and G. Stephanopoulos, "Branch and Bound Strategy with List Techniques for the Synthesis of Separation Schemes", Chem.Engng.Sci., 30, 963 (1975).

48. Rodrigo, F.R. and J.D. Seader, "Synthesis of Separation Sequences by Ordered Branch Search", A.I.Ch.E.J., 21, 885 (1975).

49. Gomez, M.A. and J.D. Seader, "Separation Sequence Synthesis by a Predictor Based Ordered Search", A.I.Ch.E.J., 22, 970 (1976).

50. King, C.J., D.W. Gantz and F.J. Barnes, "Systematic Evolutionary Process Synthesis", Ind.Eng.Chem.Proc.Des.Dev., 11, 271 (1972).

51. Stephanopoulos, G. and A.W. Westerberg, "Studies in Process Synthesis - II. Evolutionary Synthesis of Optimal Process Flowsheets", Chem.Eng.Sci., 31, 195 (1976).

52. Seader, J.D. and A.W. Westerberg, "A Combined Heuristic and Evolutionary Strategy for Synthesis of Simple Separation Sequences", A.I.Ch.E.J., 23, 951 (1977).

53. Nath, R. and R.L. Motard, "Evolutionary Synthesis of Separation Processes", A.I.Ch.E.J., 27, 578 (1981).

54. Umeda, T. A. Hirai and A. Ischikawa, "Synthesis of Optimal Processing System by an Integrated Approach", Chem.Engng.Sci., 27, 795 (1972).

55. Osaka, K. and L.T. Fan, "Synthesis of an Optimal Large-Scale Interconnected System by Structural Parameter Method Coupled with Multilevel Technique", Can.J.Chem.Eng., 51, 94 (1973).

56. Petlyuk, F.B., U.M. Plotonov and D.M. Slavinskii, "Thermodynamically Optimal Method for Separating Multicomponent Mixtures", International Chem.Eng., 5, 555 (1965).

57. Petlyuk, F.B., V.M. Plotonov and U.S. Avetlyan, "Optimum Arrangements in the Fractionation of Multicomponent Mixtures", Khim.Prom., 42, 865 (1966).

58. Tedder, D.W. and D.F. Rudd, "Parametric Studies in Industrial Distillation", "Part I - Design Comparisons", p.303, "Part II - Heuristic Optimization", p.316, "Part III - Design Methods and their Evaluation", p.323, A.I.Ch.E.J., 24 (1978).

59. Doukas, N. and W.L. Luyben, "Economics of Alternative Distillation Configurations for the Separation of Ternary Mixtures", Ing.Eng. Chem.Process.Des.Dev., 17, 272 (1978).

60. Rathore, R.N.S., K.A. Van-Wormer and G.J. Powers, "Synthesis Strategies for Multicomponent Separation Systems with Energy Integration", A.I.Ch.E.J., 20, 491 (1974).

61. Rathore, R.N.S., K.A. Van Wormer and G.J. Powers, "Synthesis of Distillation Systems with Energy Integrations", A.I.Ch.E.J., 20, 940 (1974).

62. Umeda, T., K. Niida and K. Shiroko, "A Thermodynamic Approach to Heat Integration in Distillation Systems", A.I.Ch.E.J., 25, 423 (1979).

63. Sophos, A., G. Stephanopoulos and M. Morari, "Synthesis of Optimum Distillation Sequences with Heat Integration Schemes", presented at 71st National Meeting, A.I.Ch.E., Miami, 1978.

64. Morari, M. and D.C. Faith, "The Synthesis of Distillation Trains with Heat Integration",

65. Dunford, H.A. and B. Linnhoff, "Energy Savings by Appropriate Integration of Distillation Columns into Overall Processes", presented at the I.Chem.E. Symposium Series No. 61, Leeds, July 1981.

66. Siirola, J.J., "Energy Integration in Separation Processes", Vol. II, "Foundations of Computer-Aided Chemical Process Design", S.H. Mah and W.D. Seider, pp.573 (American Institute of Chemical Engineers, 1980).

67. Sargent, R.W.H. and B.A. Murtagh, "Projection Methods for Nonlinear Programming", Math. Prog. 4, 245 (1973).

68. Sargent, R.W.H. and D.J. Sebastian, "The Minimization of Single-Variable Functions", presented at a meeting on "Optimization Problems in Engineering and Economics", Naples, December 1974.

APPENDIX A

Triangular Factorization

Given $GX = f$

with G non-singular and tridiagonal

i.e. $G_{ij} = 0$ for $j < i-1$ and $j > i+1$

G can be factorized

$$\begin{bmatrix} g_{11} & g_{12} & & & \\ g_{21} & g_{22} & g_{23} & & \\ & & & & \\ & & & & g_{n-1,n} \\ & & & g_{n,n-1}, & g_{n,n} \end{bmatrix} = \begin{bmatrix} \ell_{11} & & & & \\ \ell_{21} & \ell_{22} & & & \\ & \ell_{23} & \ell_{33} & & \\ & & & & \\ & & & \ell_{n,n-1} & \ell_{n,n} \end{bmatrix} \begin{bmatrix} u_{11} & u_{12} & & & \\ & u_{22} & & & \\ & & & & \\ & & & & u_{n-1,n} \\ & & & & u_{n,n} \end{bmatrix}$$

whence

$$g_{i,i-1} = \ell_{i,i-1}\, u_{i-1,i-1}$$

$$g_{i,i} = \ell_{i,i-1}\, u_{i-1,i} + \ell_{i,i}\, u_{i,i}$$

$$g_{i,i+1} = \ell_{i,i}\, u_{i,i+1}$$

choosing $\ell_{ii} = 1$

$$\ell_{i,i-1} = g_{i,i-1} / u_{i-1,i-1}$$

$$u_{i,i} = g_{i,i} - \ell_{i,i-1}\, u_{i-1,i}$$

$$u_{i,i+1} = g_{i,i+1}$$

If $UX = \nu$

then $L\nu = L\,UX = GX = f$

$L\,U = G$

$\ell_{11}\ u_{11} = g_{11}$ where $\ell_{11} = 1$

for i=1 $u_{11} = g_{11}$

and from $L\,\nu = f$

$\ell_{11}\ \nu_1 = f_1$ again $\ell_{11} = 1$

then $\nu_1 = f_1$

$\ell_{21}\ \nu_1 + \ell_{22}\ \nu_2 = f_2$

$\nu_2 = (f_2 - \ell_{21}\ \nu_1) / \ell_{22}$

$\nu_i = f_i - \ell_{i,i-1}\ \nu_{i-1}$ for $i > 1$

To obtain X from $UX = \nu$

for i=m $x_n = \nu_n/u_{n,n}$

$x_{n-1} = (\nu_{n-1} - u_{n-1,n}\ x_n) / u_{n-1,n-1}$

$x_i = (\nu_i - u_{i,i+1}\ x_{i+1}) / u_{i,i}$

$x_i = (\nu_i - g_{i,i+1}\ x_{i+1}) / u_{i,i}$ $i < n$

Partial pivoting was used to reduce the effects of round-off errors

APPENDIX B

Convergence properties of the direct substitution method in recycle systems.

When using the direct substitution method for a recycle system one assumes the calculated value of the recycle variable g(x) to be the new estimation of X. Then

$$X_{k+1} = g(X_k) \tag{B1}$$

where K is the iteration number

A recycle system is shown in the following figure:

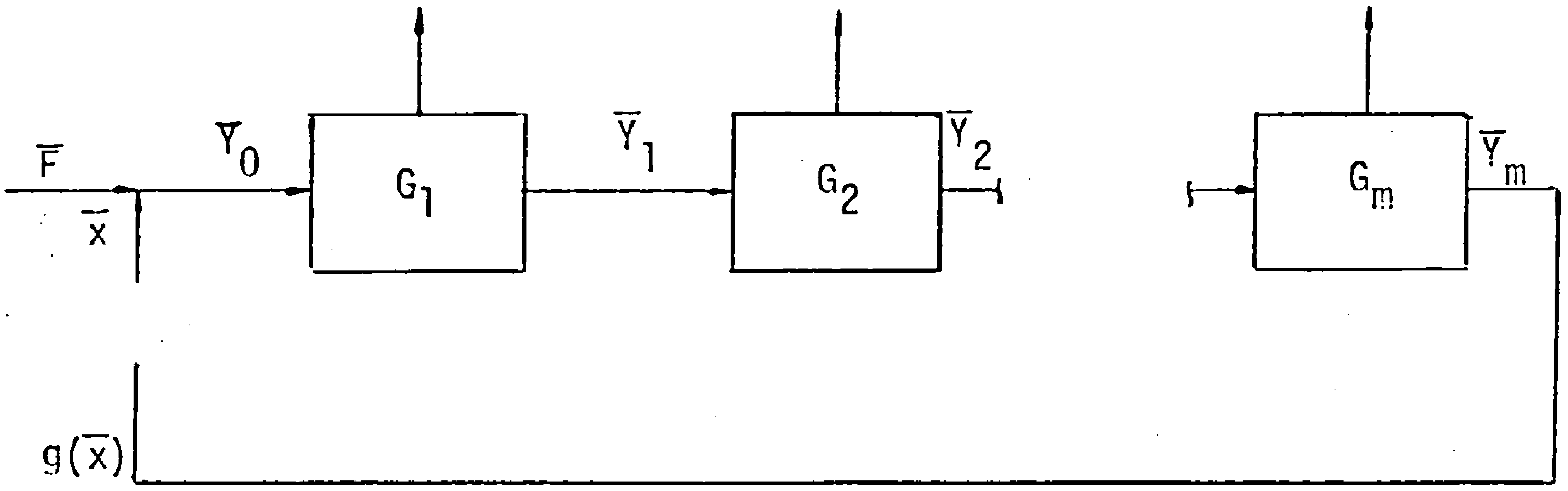

Figure B1 - A recycle system

where F is the feed

X is the assumed recycle value

G_i is the equipment matrix for unit i

y_i is the set of output variables for unit i

Given $y_o = y_m + F$ (B2)

$y_i = g_i(y_{i-1})$ $i = 1,2, \ldots m$ (B3)

To solve by direct substitution

$$\left.\begin{aligned} y_{o,k+1} &= y_{m,k} + F \\ y_{i,k+1} &= g_i(y_{i-1,k+1}) \end{aligned}\right) \quad k = 1,2,3,\ldots \tag{B4}$$

From (B4) $y_{i,k+1} - y_{i,k} = g_i(y_{i-1,k+1}) - g_i(y_{i-1,k})$

By Mean-value Theorem

$$y_{i,k+1} - y_{i,k} = G_{i,k}\,(y_{i-1,k+1} - y_{i-1,k}) \tag{B5}$$

where G_{ik} is the matrix of partial derivatives of $g_i(\cdot)$ with each row evaluated at an appropriate point between $y_{i-1,k}$ and $y_{i-1,k+1}$ (ie. at $\bar{y}_{i-1,k} = \alpha\, y_{i-1,k} + (1-\alpha)\, y_{i-1,k+1}$ for some α with $0 \leqslant \alpha \leqslant 1$).

Write $\Delta y_{i,k} = y_{i,k} - y_{i,k-1}$ (B6)

From (B5) and (B6) $\Delta y_{i,k+1} = G_{i,k}\, \Delta y_{i-1,k+1}$ (B7)

whence $\Delta y_{m,k+1} = R_k\, \Delta y_{o,k+1}$ (B8)

where $R_k = G_{m,k}\, G_{m-1,k} \cdots G_{2,k}\, G_{1,k}$

Now from (B4) and (B6) $\Delta y_{o,k+1} = y_{o,k+1} - y_{o,k}$

$$= (y_{m,k} + F) - (y_{m,k-1} + F)$$

$$= y_{m,k} - y_{m,k-1} = \Delta y_{m,k} \tag{B9}$$

From (B8) and (B9) $\Delta y_{m,k+1} = R_k\, \Delta y_{m,k}$ (B10)

Let λ_k be the eigenvalue of largest magnitude for the matrix R_k. Then if there is a positive scalar $r < 1$ such that $|\lambda_k| \leqslant r$ for all k, we have $\Delta y_{mk} \to 0$ as $k \to \infty$ and $\sum_{k=1}^{\infty} \Delta y_{m,k}$ is convergent.

But from (B6) : $$y_{m,k} = y_{m,0} + \sum_{j=1}^{k} \Delta y_{m,j} \tag{B11}$$

so the sequence $\{y_{m,k}\}$ then converges.

Note that if R* at the solution y_m^* has all eigenvalues of magnitude less than unit, and the functions $g_i(\cdot)$ are continuously differentiable, then $|\lambda_k| \leqslant r \leqslant 1$ for some r and for all $y_{m,k}$ sufficiently close to y_n^*. Thus direct substitution then converges from starting points $y_{m,0}$ sufficiently close to y^*_m.

The rate of convergence is a function of the maximal eigenvalue of the recycle matrix. The maximum eigenvalue can be estimated from the ratio of changes in two successive iterations as has been shown by Orbach and Crowe (36). The number of direct substitution steps needed before a good estimation of λ_{max} can be obtained, as the ratio of successive changes, is a function of the difference between the maximal and the second largest eigenvalue. Once, an approximated value of the maximal eigenvalue has been found, the number of additional iterations required to obtain the solution can be estimated. The number of iterations (k) required to reduce an error vector to a fraction δ of its initial value is

$$k \underline{\vee} \log \delta \,/\, \log \lambda_k \tag{B12}$$

If all the eigenvalues have an absolute value less than unity, the iterative procedure is convergent and the rate of convergence is governed by the biggest eigenvalue in absolute value, the nearer to unity, the slower the convergence. For an eigenvalue near to unity, the convergence will be very slow and what is worse, the change in the computer value from iteration to iteration will be so small that the computations may appear to have converged before a correct solution is reached.

Though the direct substitution method may have a slow rate of convergence, Shacham and Motard (41) have shown that the direct substitution method must converge for every physically stable linear recycle system. Assuming the linear recycle system is in steady state, then

$$X^s = R\,X^s + b \tag{B13}$$

Supposing that a small disturbance (δX) is introduced in the recycle stream, as the disturbance passes through the system a number of times:

$$X_1 = R(X^s + \delta X) + b = X^s + R \quad \delta X$$

$$X_2 = R\,X_1 + b = R(X^s + R\,\delta X) + b$$

$$= X^s + R^2\,\delta X$$

$$X_k = X^s + R^k\,\delta X \tag{B14}$$

Then the disturbance will die out on condition that $|\lambda_k| < 1$. If $|\lambda_k| > 1$, the disturbance will grow indefinitely. The same conclusions are valid for a general nonlinear recycle system in the vicinity of the solution. In this case R will be the Jacobian of the system near the solution, therefore, the direct substitution method must converge for every physically stable system, from an initial point which is close enough to the solution.

APPENDIX C

Acceleration of the direct substitution method

When an iteration

$$X^{k+1} = g(X^k) \qquad k=0,1, \quad n \qquad (C1)$$

leads to a linearly convergence sequence $\{X^k\}$, one method of improving the convergence is to use extrapolation in the form of Aitken's δ^2-acceleration (33). The method was originally proposed for one equation and the sequence $\{X^k\}$ was defined by

$$X^s = q^k X^{k-1} + (1-q^k) X^k \qquad (C2)$$

where

$$q^k = (X^k - X^{k-1})/(X^k - 2X^{k-1} + X^{k-2}) \qquad (C3)$$

and

$$X^{k-1} = g(X^{k-2})$$

$$X^k = g (X^{k-1})$$

The method was extended to multivariable problems by Steffensen (34) and is better known in the Chemical Engineering literature as Wegstein method (35). In its application to a system of nonlinear equations, the assumption that the system is uncoupled is made, and in this case q^k is a diagonal matrix defined as

$$q^k_{ij} = 0 \qquad i \neq j$$

$$q^k_{ij} = (X^k_j - X^{k-1}_j)/(X^k_j - 2X^{k-1}_j + X^{k-2}_j) \qquad \text{if } i=j \qquad (C4)$$

Rewriting equations C2 for each variable

$$X_j^s = X_j^{k-1} + (X_j^k - X_j^{k-1})/(1-S_j) \qquad \text{(C5)}$$

where the slope S_j is obtained from two successive trials

$$S_j = \frac{X^k - X^{k-1}}{X^{k-1} - X^{k-2}} = \frac{\varepsilon^k}{\varepsilon^{k-1}} \qquad \text{(C6)}$$

For one of the variables, the value of S_j could be a good approximation to the biggest eigenvalue, as has been shown by Orbach and Crowe (36). In their Dominant Eigenvalue Method, they used the maximum eigenvalue of the system to accelerate the convergence of the direct substitution method, using the following formula:

$$X_j^s = X_j^{k-1} + \alpha^k(X_j^k - X_j^{k-1})/(1-\lambda_{max}) \; , \forall \; j \qquad \text{(C7)}$$

with $0 < \alpha^k \leq 1$ included as a damping factor to suppress oscillation and λ_{max} being the maximum eigenvalue of the system. Orbach and Crowe (36) noted that a few iterations using direct substitution are almost always needed before each new extrapolation is made.

The extrapolation applied could be too large, and the system would then become unstable. Considering that

$$q_j = S_j \; / \; (S_j - 1) \qquad \text{(C8)}$$

then

$$\lim_{S_j \to 1} |q_j| = \infty \qquad \text{(C9)}$$

Moreover, the acceleration procedure is most need when $S_j \to 1$.

Some lower bounds on q_j could then be used to limit the extrapolation. The introduction of some delay, in the DS method, before the acceleration procedure is applied, will stabilize the acceleration. In this way, the first acceleration step is used after m direct substitution steps and from then on acceleration each n direct substitution steps.

The best values of m and n depend on the specific problem to be solved, where n is likely to have a value between 4 and 8.

APPENDIX D

The Method of Broyden

Consider a set of n nonlinear equations

$$f_j\ (X_1, X_2\ \ldots\ , X_n) = 0 \quad , \quad j=1,2\ \ldots\ n \qquad \text{(D1)}$$

that can be written more concisely as

$$f(X) = 0$$

Where X is the column vector of independent variables and f is the vector of functions f_j. If X^k is the kth approximation to the solution of equation (D1) and f^k is written for $f(X^k)$, then Newton's method is defined by

$$X^{k+1} = X^k - A_k^{-1}\ f^k \qquad \text{(D2)}$$

where A_k is the Jacobian matrix evaluated at X^k.

One disadvantage of the Newton's method is the evaluation of the Jacobian matrix each iteration. If the partial derivatives cannot be calculated analytically, an approximation to the Jacobian matrix must be obtained numerically. Broyden (11) described a class of methods in which an approximation of the Jacobian or the inverse of the Jacobian is used and the approximation is updated after each step using the information generated during the step. No additional function evaluation is required, thus being much simpler to perform than the evaluation of the complete Jacobian. Supposing that

$$p^k = -\ B_k^{-1}\ f^k \qquad \text{(D3)}$$

where B_k is some approximation to the Jacobian matrix evaluated at X^k, then

$$X^{k+1} = X^k + t\ p^k \qquad \text{(D4)}$$

where t is a scalar multiplier chosen to prevent the process diverging.

An approximation to the negative increase of the Jacobian matrix is named as

$$H^k = B_k^{-1} \tag{D5}$$

The value of the matrix H is updated using the following equation

$$H^{k+1} = H^k - \frac{(p^k + H^k y^k)\, p^{kT} H^k}{p^{kT} H^k y^k} \tag{D6}$$

where

$$y^k = f(X^{k+1}) - f(X^k) \tag{D7}$$

and $$p^k = X^{k+1} - X^k \tag{D8}$$

To begin with, values of X^o and H^o are needed. Given values of X^o, the initial Jacobian can be obtained using finite differences. Alternatively, the identity matrix can be used as the initial approximation. For sparse Jacobian matrices, the knowledge of the constant elements in a Jacobian matrix can be used to reduce the work required to estimate the remaining elements by finite differences as shown by Curtis, Powell and Reid (26). Shubert (25) developed a formula to update the Jacobian for sparse systems.

Broyden (11) proposed to adjust t to guarantee norm reduction, such that

$$\| F^{k+1} \| < \| F^k \| \tag{D9}$$

where $$F^k = f^{kT} f^k \tag{D10}$$

This normal reduction impair the efficiency of the Broyden method, therefore Perkins and Metcalfe (37) suggested the unity for the parameter b, unless $\| F^{k+1} \| > 10 \| F^k \|$ in which case t = 0.1 should be used. This version of Broyden was used.