A simple, reliable and fast algorithm for the simulation of multicomponent distillation columns

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A B S T R A C T

This work has developed a simple, reliable and fast algorithm for the simulation of multicomponent distillation columns, where any equilibrium stage can accept a feed-stream and/or a vapor-side-stream and/or liquid-side-stream. The new scheme considers internal molar overflows and constant relative volatilities to avoid the need of heat balances and vapor–liquid equilibrium calculations. The solution scheme is founded on a Newton-based formulation in block algebra, which relies in a simple, reliable and fast algorithm. Although the proposed calculation scheme can be classified as an approximate method, it is very useful when accurate phase equilibrium and enthalpy data are lacking. Numerical experimentations show good agreement with the results obtained with well-known rigorous simulation approaches.

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1. Introduction

In the literature, a number of well proven rigorous methods are available for the simulation of multicomponent vapor–liquid distillation columns (c.f. King, 1980; Holland, 1981; Kister, 1992; Seader and Henley, 2005). Most of these methods rely on stage-by-sage calculations or simultaneous solution of the material, equilibrium, summation and heat equations, usually identified as MESH equations that describe the separation process. The solution of these equations can be cumbersome and time consuming even with the current availability of computing capabilities.

Nowadays, there are two approaches to approximate the product compositions in multistage vapor–liquid separations of a given mixture: (1) grouping the equations that describe the separation process, denoted as group methods (c.f. work by Kamath et al., 2010), or (2) based on the Fenske (Fenske, 1932), Underwood (Underwood, 1948) and Gilliland (Gilliland, 1940) equations, denoted as FUG methods (c.f. work by Gadalla et al., 2003).

On the other hand, when accurate phase equilibrium and enthalpy data are lacking it is often not convenient to resort to rigorous thermodynamic models, since in many cases one would like to obtain a reliable estimate of the composition of the products, as those obtained with a rigorous simulation approach, without sacrificing rigorousness of computation to the speed of calculation.

This work aims at providing a simple, efficient, reliable and fast algorithm to the problem of simulation of multicomponent vapor–liquid distillation columns, for the separation of ideal and nearly ideal mixtures, based on constant relative volatilities and constant molar overflow, where any equilibrium stage can accept a feed stream and/or vapor side-stream and/or liquid-side streams. The simple characteristic is due to the use of only material and equilibrium equations. The efficient characteristic is provided due to the proposed approach rely in a simultaneous solution of all the equations involved through a Newton iteration scheme based in block algebra. The reliable feature is due to the assumptions of constant molar overflow and constant relative volatilities. These assumptions are well accepted for modeling separation processes involving hydrocarbon mixtures; they have been used in dynamical, control behavior (see for instance, Skogestad and Morari, 1988) and even in steady state

Nomenclature

A	elements of the Jacobian Matrix
B	elements of the Jacobian Matrix
C	elements of the Jacobian Matrix
F	molar flow rate of a feed stream
F	vector of nonlinear functions
L	molar flow rate of a liquid stream
M	material balance equation
N	number of stages
N_C	number of components
SL	molar flow rate of a liquid side stream
SV	molar flow rate of a vapor side stream
V	molar flow rate of a vapor stream
x	mole fraction of a component in the liquid phase
x	vector of unknowns
y	mole fraction of a component in the vapor phase
z	mole fraction of a component in the feed stream

Greek letters

α	relative volatility of a component
δ	Kronecker delta

Subscripts

i, j, k, m, n integer indices

studies (see for instance, Huss and Westerberg, 1996; Vaca et al., 2007). However, the structure and convergence characteristics of the problems involved have not been explicitly presented or exploited, being these two things the principals of this work. Numerical experimentation shows that the results obtained with the proposed algorithm are in good agreement with those obtained with rigorous well accepted methods like the presented by Naphtali and Sandholm (1971) either for simple and complex distillation columns configurations. Note that in this work we consider as simple configurations, those columns with one feed stream and two product streams (one top product and one bottom product), and for complex configurations those columns with one or more feed streams and/or two or more product streams.

This work is organized as follows. The mathematical model of the proposed algorithm is described in Section 2. Some computational aspects related to the numerical solution of the proposed model are discussed in Section 3. Numerical experimentations of different column configurations are presented in Section 4. Some issues regarding the mathematical approach are presented in Section 5. The work is closed with some concluding remarks in Section 6.

2. Mathematical model

Let us consider a system with N stages and N_C components, where the stages are numbered from top to bottom. The material balance around a j-th stage yields:

$$M_{i,j} = V_{j+1}y_{i,j+1} + L_{j-1}x_{i,j-1} + F_jz_{i,j} - (SV_j + V_j)y_{i,j}$$
$$- (SL_j + L_j)x_{i,j} = 0 \tag{1}$$

where $x_{i,j}$, $y_{i,j}$, $z_{i,j}$ are the mole fractions of component i, on stage j, respectively, in the liquid phase, vapor phase and feed stream; F_j is the feed stream flow rate entering stage j; L_j, V_j, SL_j, and SV_j are the liquid, vapor, liquid side stream and vapor side stream flow rates leaving stage j, respectively.

Assuming that the exit streams are in theoretical equilibrium, and that the ratio of the equilibrium constants are near independent of the temperature (see for instance, Doherty and Malone, 2001), we can expect that the following relationship holds:

$$y_{i,j} = \frac{\alpha_i x_{i,j}}{\sum_{k=1}^{N_C} \alpha_k x_{k,j}} \tag{2}$$

where α_i is the relative volatility of component i, then (1) can be cast as:

$$M_{i,j} = V_{j+1}\frac{\alpha_i x_{i,j+1}}{\sum_{k=1}^{N_C} \alpha_k x_{k,j+1}} + L_{j-1}x_{i,j-1} + F_jz_{i,j}$$
$$- (SV_j + V_j)\frac{\alpha_i x_{i,j}}{\sum_{k=1}^{N_C} \alpha_k x_{k,j}} - (SL_j + L_j)x_{i,j} = 0 \tag{3}$$

The set of $N \cdot N_C$ nonlinear equations (3) involves the following variables: $2N$ internal mole flow, that is, the flow rates of liquid and vapor streams leaving the equilibrium stage, $2N$ flow rates of liquid and vapor side streams, N flow rates of feed streams, $N \cdot N_C$ liquid mole fractions, $N \cdot N_C$ feed stream mole fractions, and N_C relative volatilities. The total number of variables is $2N \cdot N_C + 5N + N_C$. Then, the degrees of freedom are $N \cdot N_C + 5N + N_C$. For a simulation problem, the degrees of freedom are fulfilled assigning N feed flows and $N \cdot N_C$ feed stream mole fractions; $2N$ flow rates of liquid and vapor side streams; $2N$ flow rates of liquid and vapor streams leaving the equilibrium stage, and N_C relative volatilities. Note that the relative volatilities are calculated at the average feed streams conditions and the internal mole flow rates V_j and L_j are known using the constant molar overflow assumption (Seader and Henley, 2005). After, the degrees of freedom are satisfied, the unknowns are the liquid mole fractions, providing a system of $N \cdot N_C$ equations (that is, $M_{i,j} : i = 1, \ldots, N_C; \ j = 1, \ldots, N$) with $N \cdot N_C$ unknowns (that is, $x_{i,j} : i = 1, \ldots, N_C; \ j = 1, \ldots, N$).

In this approach two things have to be considered:

1. The assumption of constant molar overflow fixes the global balance around the equilibrium stage, and the fact that the summation of the feed composition is unity, in those equilibrium stages where a feed stream is present, forces that the summation of liquid compositions in all the equilibrium stages are unity when the solution is reached, that is, the summation restriction $\sum_{i=1}^{i=Nc} x_{i,j} - 1 = 0$ is implicitly satisfied and not have to be considered.
2. Since the relative volatilities are assumed constant along all the stages, provides that phase equilibrium calculation are avoided (*i.e.* bubble point at each stage, see for instance, Monroy-Loperena, 2003).

The next section describes the solution method for the system of equations (3).

3. Solution of the proposed mathematical model

Solving system (3) means finding the set of independent variables **x** which satisfies the relationships, **F(x)** = 0, where **F** is
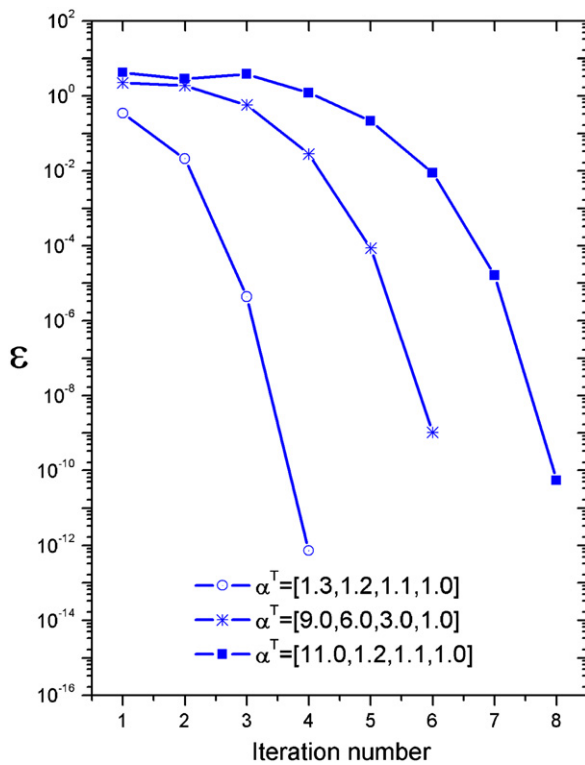
Fig. 1 – Convergence patterns with different constant relative volatilities sets for the fed mixture.

the vector of functions (3) and $\mathbf{x}$ is the vector of liquid mole fractions, both arranged by stage. To carry out this situation we propose to use the well known Newton iteration scheme. Denoting k as an iteration index, in Newton iteration, a new set of values of functions $\mathbf{F}^{k+1}$ are generated from a previous estimate in the following fashion

$$\mathbf{F}(\mathbf{x}^{k+1}) = \mathbf{F}(\mathbf{x}^k) + \left[\frac{\partial \mathbf{F}}{\partial \mathbf{x}}\right]_{\mathbf{x}=\mathbf{x}^k}(\mathbf{x}^{k+1} - \mathbf{x}^k) = 0 \qquad (4)$$

This equation is used to estimate $\mathbf{x}^{k+1}$. When $\mathbf{x}^{k+1} - \mathbf{x}^k$ is sufficiently small, the correct set of values of $\mathbf{x}$ has been found, and the iteration stops.

The Jacobian Matrix $[\partial\mathbf{F}/\partial\mathbf{x}]$ is here very large, but its evaluation is greatly facilitated by the fact that the conditions on stage j only are directly influenced by the conditions on stage $j-1$ and $j+1$. As a result, the Jacobian becomes block-tridiagonal in structure, which permits rapid solution by block elimination. The equations are solved by Thomas elimination (see for instance, Seader and Henley, 2005), until the Euclidian norm of the functions is smaller than a given tolerance, ε, say 10^{-6}.

The elements of each block that conform the Jacobian Matrix are:

$$A_{j,m,n} = \frac{\partial M_{m,j}}{\partial x_{n,j-1}} = \delta_{m,n}L_{j-1} \qquad (5)$$

$$B_{j,m,n} = \frac{\partial M_{m,j}}{\partial x_{n,j}} = -(V_j + SV_j)\left[\frac{\alpha_m}{\sum_{k=1}^{N_C}\alpha_k x_{k,j}}\right]$$

$$\times \left[\delta_{m,n} - \frac{\alpha_n x_{m,j}}{\sum_{k=1}^{N_C}\alpha_k x_{k,j}}\right] - \delta_{m,n}(L_j + SL_j) \qquad (6)$$

$$C_{j,m,n} = \frac{\partial M_{m,j}}{\partial x_{n,j+1}} = V_{j+1}\left[\frac{\alpha_m}{\sum_{k=1}^{N_C}\alpha_k x_{k,j+1}}\right]$$

$$\times \left[\delta_{m,n} - \frac{\alpha_n x_{m,j+1}}{\sum_{k=1}^{N_C}\alpha_k x_{k,j+1}}\right] \qquad (7)$$

where $\delta_{m,n}$ is the Kronecker delta.

4. Numerical experimentation

First to show the reliability of the proposed method to handle different kinds of mixtures, that is, close boiling and wide

| Table 1 – Characteristics of the cases of study.				
	Case 1		Case 2	
Column pressure [kPa]	2000		1650	
Stages	31		16	
Condenser type	Total		Partial	
Vapor top product [kmol/h]	–		15.0	
Liquid top product [kmol/h]	50.0		5.0	
Bottom product [kmol/h]				
Liquid side stream [kmol/h]	–		3.0	
Stage number	–		3	
Vapor side stream [kmol/h]	–		37.0	
Stage number	–		13	
Reflux ratio	6		7.5	
Feeds	1	1	1	2
Ethane (C$_2$) [kmol/h]	–	2.5		0.5
Propane (C$_3$) [kmol/h]	50.0	14.0		6.0
N-butane (n-C$_4$) [kmol/h]	50.0	19.0		18.0
Isobutane (i-C$_4$) [kmol/h]	50.0	–		–
N-pentane (n-C$_5$) [kmol/h]	50.0	5.0		30.0
N-hexane (n-C$_6$) [kmol/h]	–	0.5		4.5
Temperature [K]	370.8	339.7		386.3
Pressure [kPa]	2000	1650		1650
Stage number	13	6		9
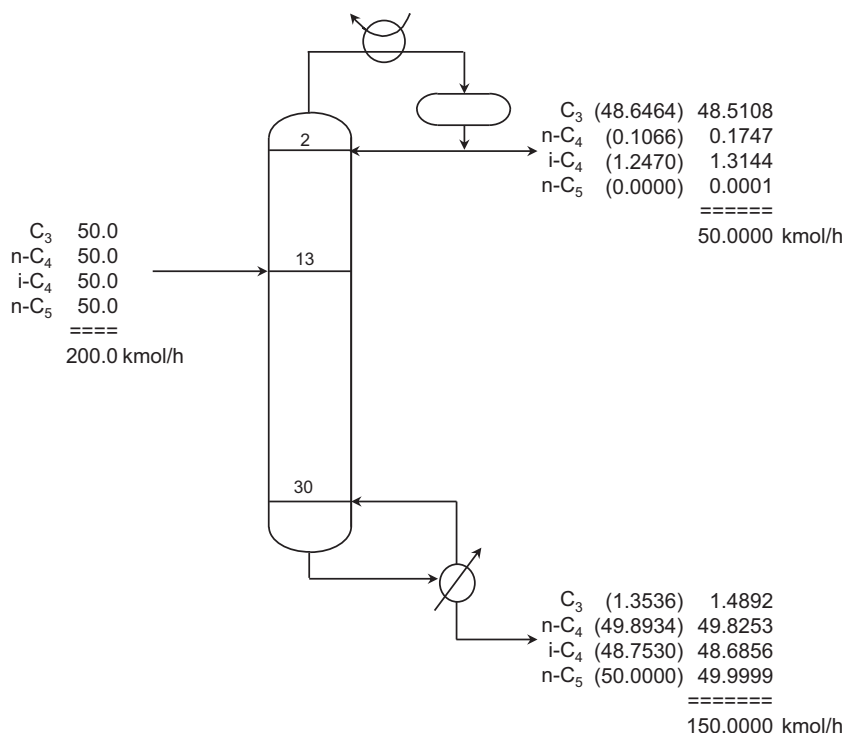
Fig. 2 – Specifications and results for the case of study 1.

boiling mixtures, let us assume an equimolar four component mixture with a flow of 100 kmol/h that will be introduced at stage 5 for a column with 10 equilibrium stages and a reflux ratio of 3. The distillate and bottom flow rates are specified as 50 kmol/h. Three cases are considered, the former consists of a mixture with constant relativity volatilities of $\boldsymbol{\alpha}^T = [1.3, 1.2, 1.1, 1.0]$ that can be viewed as a close boiling mixture, a second one with constant relativity volatilities of $\boldsymbol{\alpha}^T = [9.0, 6.0, 3.0, 1.0]$ arriving to an intermediate boiling mixture, and a last one with constant relativity volatilities of $\boldsymbol{\alpha}^T = [11.0, 1.2, 1.1, 1.0]$ that can be viewed as a wide boiling mixture. In all these cases the initialization of the liquid mol fractions in all the stages is taken as 0.25 (equimolar mole fractions). Referring to Fig. 1, we can see that a close boiling mixture need less iterations to achieve the convergence than a wide boiling mixture, part of this is because the initialization composition profiles that are more *ad hoc* with the final results obtained in the close boiling mixture, but what we want to show is that the method is capable to solve all kinds of mixtures without any problem (see for instance, Friday and Smith, 1964).

Now we will show the proposed method in two benchmark cases of simulation studies. As the first case of study, consider a depropanizer, with 31 equilibrium stages including the reboiler and a total condenser (see Table 1 and Fig. 2). Since the distillate composition will be basically propane, the
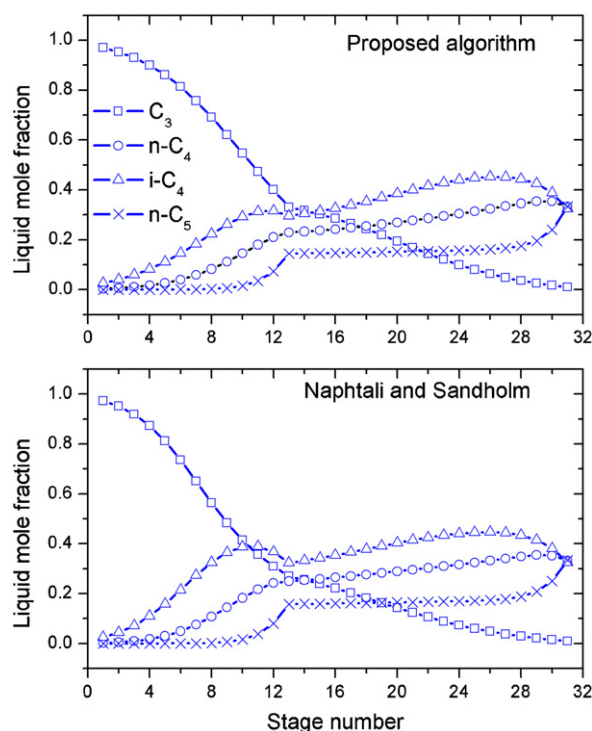

Fig. 3 – Liquid composition profiles for the case of study 1.

calculation of this column would normally be expected to be difficult (see for instance, Orbach and Crowe, 1971; Monroy-Loperena and Flores-Sánchez, 2003). For this case the relative volatilities used are shown in Table 2, and there were calculated from the global composition fed to the column at its bubble and dew points at the average pressure of the column, using the thermodynamical model based in the Soave–Redlich–Kwong equation of state (Soave, 1972). The pure-component physical properties required in the calculation were taken from Reid et al. (1987). The complete set
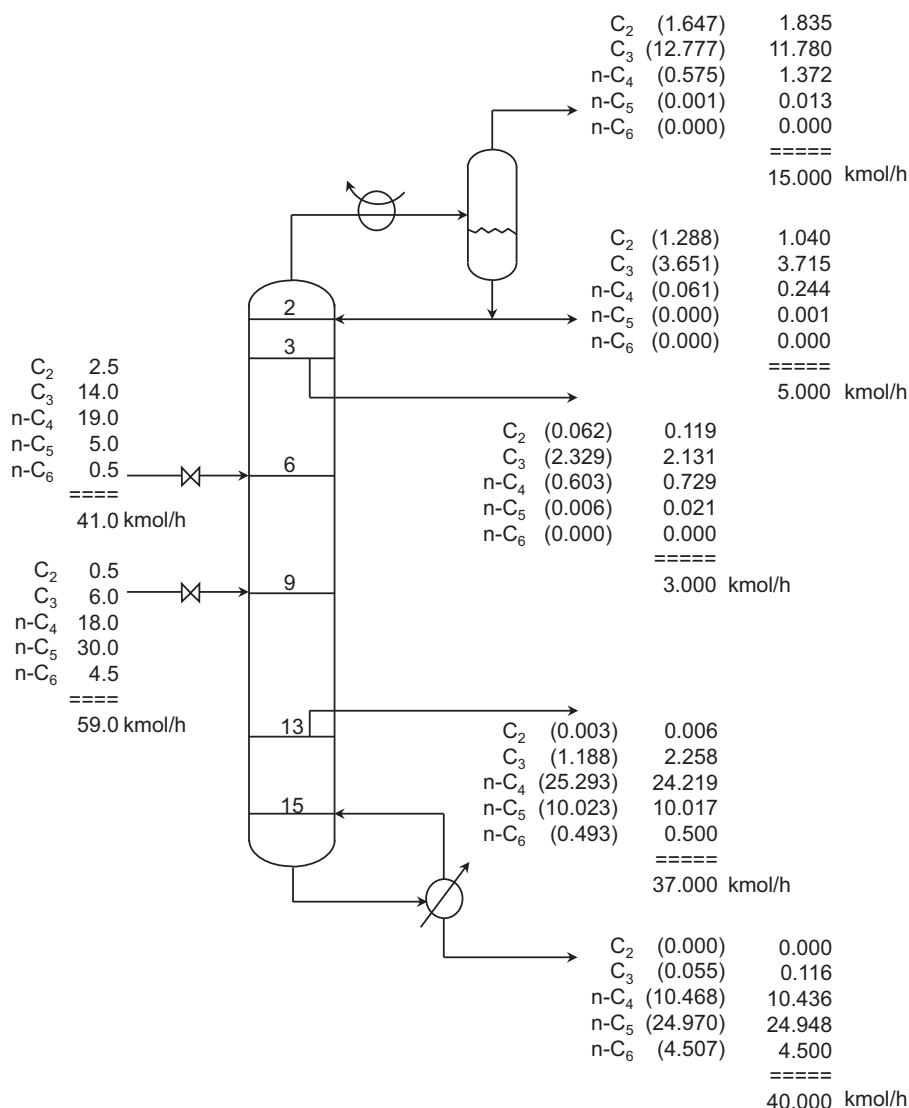
Table 2 – Constant relative volatilities used in the cases of study.		
	Case 1	Case 2
Ethane (C$_2$)		9.56
Propane (C$_3$)	3.35	5.31
N-butane (n-C$_4$)	1.81	2.99
Isobutane (i-C$_4$)	2.12	
N-pentane (n-C$_5$)	1.00	1.73
N-hexane (n-C$_6$)		1.0

Fig. 4 – Specifications and results for the case of study 2.

of results is presented in Fig. 2. Initial values for the liquid mole fractions were set as 0.25 (equimolar mole fractions) in all the stages. For the sake of comparison also in Fig. 2, the results are compared with those obtained using the Naphtali and Sandholm (Naphtali and Sandholm, 1971) iteration scheme, using the thermodynamical model based in the Soave–Redlich–Kwong equation of state. Fig. 3 shows the liquid composition profiles obtained with the proposed calculation and those obtained with the Naphtali and Sandholm iteration scheme (values in parenthesis). Note the good agreement between the two liquid composition profiles.

As a second benchmark case of study, consider a more complex distillation configuration; with two feeds, two liquid side-streams and one vapor-side stream (see Table 1 and Fig. 4). For this case the relative volatilities are presented in Table 2, and are calculated as in the first example, from the average feed composition. Initial values for the liquid mole fractions were set as 0.20 (equimolar mole fractions) in all the stages. The complete set of results is presented in Fig. 4. Also in Fig. 4, the results obtained by the Naphtali and Sandholm iteration scheme are presented. Fig. 5 shows the liquid composition profiles obtained with the proposed calculation scheme and those obtained with the Naphtali and Sandholm iteration scheme (values in parenthesis). As in the previous case,

a good agreement between the product compositions and liquid composition profiles is obtained, although the presence of multiple feeds and side-streams.

Finally, Fig. 6 shows the evolution of the convergence for the last case assuming three different initial sets of independent variables (liquid mole fractions), as follows: (1) initial liquid mole fractions profile were assumed as $x_{i,j} = \alpha_i / \sum_{k=1}^{k=N_C} \alpha_k$ for $j = 1, \ldots, N$, (2) liquid mole fractions profiles have been set as an equimolar mixture, and finally, (3) all mole fractions in the column were fixed to 1×10^{-6}. Note that in all the initializations the presented scheme is stable, because its ability of convergence by approaching to the ultimate solution is monotonic and without either oscillations or divergence.

5. Some comments regarding the merits of the algorithm presented above

(i) Because of the large dimensionality of the original system the ill-conditioning is avoided by means of the solution through the block-tridiagonal system.
(ii) The solution of the block-tridiagonal system requires less computing and is more stable than that of the full system.
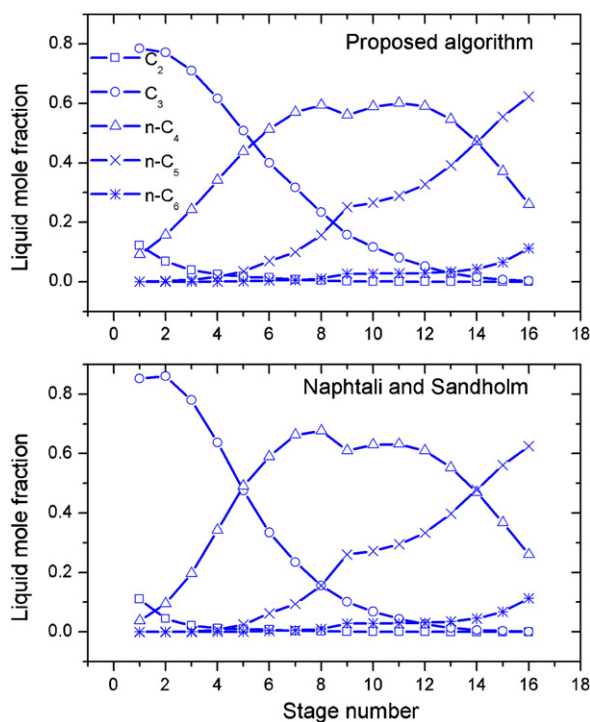
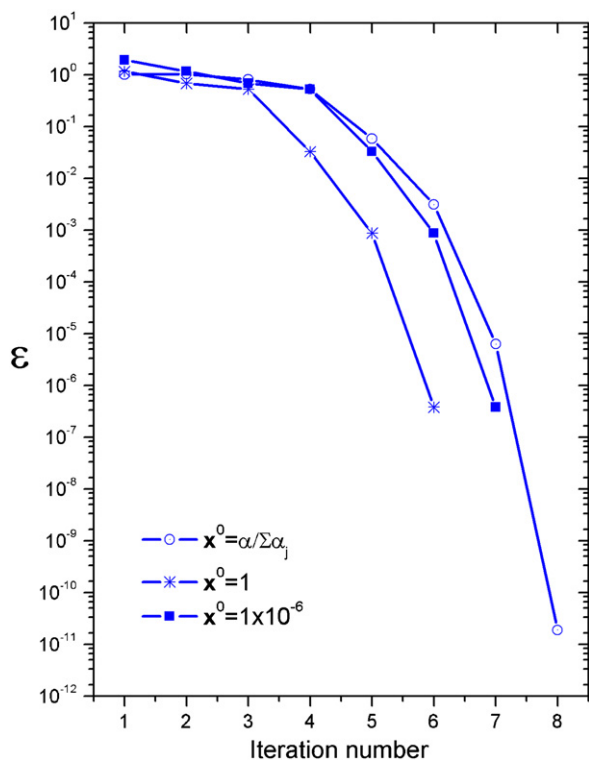**Fig. 5 – Liquid composition profiles for the case of study 2.**

Fig. 6 – Convergence patterns for the case of study 2 with different initial sets of the liquid mole fractions profiles.

(iii) The convergence characteristics of the proposed iteration scheme do not depend on the configuration of the distillation column.

(iv) The proposed iteration scheme can be considered reliable, because the use of simple initialization procedures, for the liquid composition profile, ensuring a stable approach to the solution.

(v) The relative volatilities are truly assumed constant because they are calculated only one time with the information of the feeds, bringing the effect that the vapor–liquid equilibrium calculations are eliminated.

(vi) Because the method is based on the approach of stage to stage material balances, the internal profiles of composition are consistent.

(vii) The proposed algorithm, compared to the well known methods, based on groups or FUG equations, in addition to the compositions of the products gives the internal composition profiles, even in the case of columns with multiple feeds streams and/or side streams, which gives more versatility for the study of separation processes (see for instance, Fruehauf and Mahoney, 1993).

(viii) By modifying Eq. (1) to accept streams from or to a non-adjacent stage and replacing the Thomas elimination procedure, results in that the proposed scheme can handle interlinked configurations without any problem (see for instance, Browne et al., 1977; Hofeling and Seader, 1978).

(ix) The fact that relative volatilities are almost independent of temperature allows the proposed calculation procedure provides good mole fraction profiles without considering the stage temperatures as iteration variables. Other well known methods, that use liquid mole fractions or liquid molar flows as iteration variables, have to consider the stage temperature as an iteration variable due to vapor–liquid equilibrium coefficients are strong functions of temperature (see for instance, Fredenslund et al., 1977a,b; Block and Hegner, 1976; Cairns and Furzer, 1990). However, once the nonlinear system of equations has been solved, if it is required, the mole fractions of the vapor streams and the stage temperatures can be calculated applying bubble point calculations (see for instance, Monroy-Loperena, 2003).

(x) The proposed calculation procedure have to be seen as the most simple, fast and a reliable method for the simulation of multicomponent distillation ideal and nearly ideal mixtures, when accurate phase equilibrium and enthalpy data are lacking.

6. Conclusions

In this work the well-known constant molar overflow and constant relative volatility assumptions are used to develop a simple, reliable and fast algorithm for the simulation of multicomponent distillation columns, where any equilibrium stage can accept a feed-stream and/or a vapor-side-stream and/or liquid-side-stream.

The proposed algorithm relies on a Newton-based formulation in block algebra which provides simple, reliable and fast computational scheme, even if approximately.

With reference to others well-known approximate methods as those denoted as group methods or FUG methods, the proposed calculation procedure gives the product composition coupled with the liquid composition profile, even in the case where multiple feeds streams and/or side streams are used.

Comparison of the obtained results with those obtained by a rigorous procedure (that is, Naphtali and Sandholm iteration scheme) shows the reliability of the proposed algorithm. The accuracy appears to be satisfactory also in those distillation cases with many feed-streams and side-streams.

Finally, it must be remarked that the proposed calculation scheme is not a substitute of well known rigorous methods but is very usefully when accurate phase equilibrium and enthalpy data are lacking.

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