

Improvement of the Relaxation Method for Multicomponent Distillation

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A new method is proposed which improves the stability and the rate of convergence of the relaxation method for a multicomponent distillation. The relaxation factor is treated as dependent on each component on each plate at each iteration step, and is determined by the molar flow rate of streams to and from the plate. The proposed method is applicable to an ideal and non-ideal distillation column with any number of feeds and side-cut streams, and to multiple distillation columns interlinked by recycle streams. The applicability of the proposed method is demonstrated by the excellent convergence for several examples.

1. Introduction

Several calculation methods for a multicomponent distillation are available in the literature, but there are still needs for a more efficient and general method which may be applicable to all types of columns and mixtures, even a complex unit with multiple feeds and a highly non-ideal mixture, e.g. an extractive distillation.

Among the methods, the basic relaxation method of Rose et. al.⁹⁾ has recently been recognized as one of the most advantageous methods. Many attempts^{1,3,4,5)} have been made to improve its stability and rate of convergence. Ball¹⁾, Ishikawa and Hirata³⁾, and Jelinek et. al.⁵⁾ modified the working equations of the procedure. Ishikawa and Hirata⁴⁾ further modified the relaxation factor by treating it empirically dependent on individual component at each iteration step, but not on each plate of the column.

In this investigation, it is intended to develop a method of the determination of the value of the relaxation factor. This factor is considered as component, plate and iteration step dependent, and is determined by the molar flow rate of streams to and from the plate at each iteration step. It is expected that the resulting method increases the rate and stability of convergence, and is applicable to wide variety of distillations for practical purposes.

2. Model Column and Assumptions

The basic model column under consideration is a general complex distillation column shown in Fig.

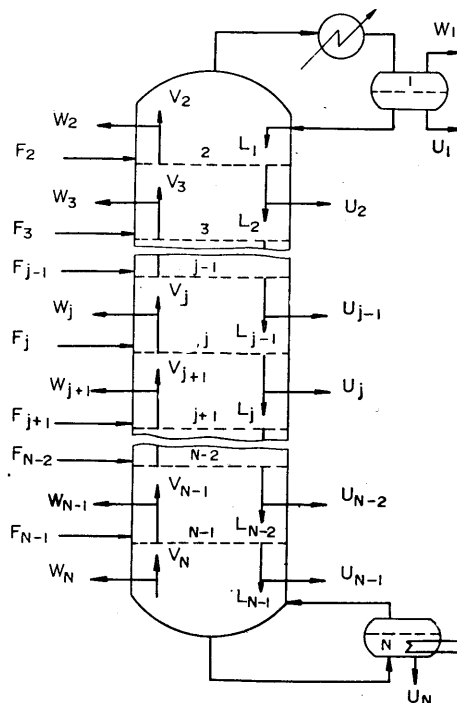


Fig. 1 Schematic diagram of the model column

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1. The top end of the column may have no condenser or any one of three types, namely, a total, partial, or two-product condenser. The bottom end of the column may have no reboiler or a partial reboiler.

The standard assumption of theoretical equilibrium plates is made. Enthalpy balances are not considered in this investigation for the simplification, taking the unreliability of enthalpy predictions of non-ideal mixtures into account. The constant molar flow process is under consideration.

3. Working Equation

The basic equation of the relaxation method is that of the transient mass balance around a plate. When usual simplifying assumptions of

- a) negligible vapor hold-up
- b) constant liquid hold-up
- c) composition of liquid hold-up is identical with that of liquid leaving the plate

are made, the equation is represented by

$$H_j^L \frac{dx_{i,j}}{dt} = \{V_{j+1}y_{i,j+1} + L_{j-1}x_{i,j-1} + F_j z_{i,j}\} - \{(V_j + W_j)y_{i,j} + (L_j + U_j)x_{i,j}\}, \quad (1)$$

in which H_j^L refers to the total moles of hold-up on j -th plate. Integration of Eq.(1) by using the explicit Euler method yields

$$x^{(n+1)}_{i,j} = x^{(n)}_{i,j} + \frac{\Delta t}{H_j^L} [\{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n)}_{i,j-1} + F_j z_{i,j}\} - \{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}\}], \quad (2)$$

where Δt refers to a small time increment. Eq.(2) is the working equation of Rose et. al.⁹⁾ Ishikawa and Hirata³⁾ improved it to give

$$x^{(n+1)}_{i,j} = x^{(n)}_{i,j} + \mu [\{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_j z_{i,j}\} - \{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}\}], \quad (3)$$

where the relaxation factor is defined by

$$\mu \equiv \Delta t / H_j^L. \quad (4)$$

Eq.(3) is used as the working equation in this investigation.

4. Methods of Determinating the Value of Relaxation Factor

In cases where the value of μ is too small, convergence of $x^{(n+1)}_{i,j}$ obtained by Eq.(3) is monotonous but slow. On the other hand, in cases where the value is too large, the over correction provides

the phenomenon of vibration or divergence. This suggests the presence of an optimum value of it. The examination by numerical computations indicates that the optimum value depends strikingly on component on each plate at each iteration step. The relaxation factor can then be considered as

$$\mu^{(n)}_{i,j} = \Delta t^{(n)}_{i,j} / H_j^L. \quad (5)$$

A large time increment, $\Delta t^{(n)}_{i,j}$, is taken for a stable component, and a small time increment for an unstable component. Eq.(3) indicates that the composition change in the hold-up during the time interval of $\Delta t^{(n)}_{i,j}$ is due to the net quantity of the component added and removed as the result of streams to and from the plate.

Therefore, it must be reasonable to determine the value of $\Delta t^{(n)}_{i,j}$ by the following physical ground, taking the stability of the component change in the hold-up into account.

"Moles of component i added to and/or removed from j -th plate during the time interval of $\Delta t^{(n)}_{i,j}$ must not exceed moles of component i in the hold-up on the plate."

The above principle can be written as

$$H_j^L x^{(n)}_{i,j} \geq \Delta t^{(n)}_{i,j} g^{(n)}_{i,j}, \quad (6)$$

where $g^{(n)}_{i,j}$ is molar flow rate of component i added to and/or removed from j -th plate. Then, from the definition, the value of $\mu^{(n)}_{i,j}$ is restricted in the range:

$$\mu^{(n)}_{i,j} \leq x^{(n)}_{i,j} / g^{(n)}_{i,j}. \quad (7)$$

Introducing a parameter ξ ($0 < \xi \leq 1$), Eq. (7) is rewritten as

$$\mu^{(n)}_{i,j} = (1 - \xi) x^{(n)}_{i,j} / g^{(n)}_{i,j}. \quad (8)$$

In the definition of $g^{(n)}_{i,j}$, four cases (methods) can be considered as follows:

Method I $g^{(n)}_{i,j} \equiv$ total molar flow rate of component i leaving j -th plate at n -th iteration.
 $= (V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}$ (9)

Then,

$$\mu^{(n)}_{i,j} = (1 - \xi) \frac{x^{(n)}_{i,j}}{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}} \quad (10)$$

$$x^{(n+1)}_{i,j} = \left\{ \xi + (1 - \xi) \frac{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_j z_{i,j}}{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}} \right\} \times x^{(n)}_{i,j}. \quad (11)$$

When $\xi = 0$, this method refers to the Gauss-Seidel method. Eq.(11) indicates that this method never yields a negative value of $x^{(n+1)}_{i,j}$.

Method II $g^{(n)}_{i,j} \equiv$ the larger value between total

Table 1 Examples for the comparison of Methos I, II, III and IV

Case No.	1	2*	3*	4*	5*	
System (Components)	(1) Acetone (2) Acetonitrile (3) Water	(1) Water (2) Acetone (3) Ethanol**	(1) Acetone (2) Methanol (3) Water**	(1) Water (2) Acetone (3) Furfural**	(1) Ethylacetate (2) Ethanol (3) Butylacetate**	
Feed	No.1	j F q z_i 9 1.0 1.0 0.3 0.4 0.3	j F q z_i 11 1.0 1.0 0.0 0.0 1.0	j F q z_i 9 1.5 1.0 0.0 0.0 1.0	j F q z_i 8 0.5 0.5 0.0 0.0 1.0	j F q z_i 3 2.0 1.0 0.0 0.0 1.0
	No. 2		22 1.0 1.0 0.01 0.99 0.00	21 1.0 1.0 0.5 0.5 0.0	16 0.5 0.5 0.0 0.0 1.0	9 1.0 1.0 0.0 0.0 1.0
	No. 3				24 1.0 0.5 0.01 0.99 0.00	15 1.0 1.0 0.5 0.5 0.0
Product	No. 1	j Phase Amount 1 (D) liq. 0.3	j Phase Amount 1 (D) liq. 0.9	j Phase Amount 1 (D) liq. 0.48	j Phase Amount 1 (D) liq. 0.9	j Phase Amount 1 (D) liq. 0.8
	No. 2	17 (B) liq. 0.7	32 (B) liq. 1.1	33 (B) liq. 2.02	31 (B) liq. 1.1	26 (B) liq. 3.2
N/R	17/3.0	32/2.0	33/4.0	31/2.0	26/3.0	
Eq. for activity coefficient	Margules	Margules	Wilson	Margules	Wilson	

molar flow rate of component i leaving and that entering j -th plate at n -th iteration.

$$= \max \left\{ \left[V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_jz_{ij} \right], \left[(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j} \right] \right\} \quad (2)$$

i) When $\{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_jz_{ij}\} \leq \{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}\}$, this method is identical with method I.

ii) When $\{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_jz_{ij}\} > \{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}\}$,

$$\mu^{(n)}_{i,j} = (1-\xi) \frac{x^{(n)}_{i,j}}{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_jz_{ij}} \quad (3)$$

$$\mu^{(n)}_{i,j} = (1-\xi) \frac{x^{(n)}_{i,j}}{\max[V_{j+1}y^{(n)}_{i,j+1}, L_{j-1}x^{(n+1)}_{i,j-1}, F_jz_{ij}, (V_j + W_j)y^{(n)}_{i,j}, (L_j + U_j)x^{(n)}_{i,j}]}, \quad (6)$$

In this case, $x^{(n+1)}_{i,j}$ calculated by Eqs. (3) and (6) is not always positive. When it is negative, $x^{(n+1)}_{i,j}$ is recalculated by Eq.(11) of Method I.

Method IV $g^{(n)}_{i,j} \equiv$ the larger value between vapor and liquid flow rate of component i leaving j -th plate at n -th iteration.

$$= \max[(V_j + W_j)y^{(n)}_{i,j}, (L_j + U_j)x^{(n)}_{i,j}] \quad (7)$$

Then,

$$x^{(n+1)}_{i,j} = \left\{ (2-\xi) - (1-\xi) \right\} \times \frac{(V_j + W_j)y^{(n)}_{i,j} + (L_j + U_j)x^{(n)}_{i,j}}{V_{j+1}y^{(n)}_{i,j+1} + L_{j-1}x^{(n+1)}_{i,j-1} + F_jz_{ij}} x^{(n)}_{i,j} \quad (4)$$

Eq. (4) always gives a positive value of $x^{(n+1)}_{i,j}$.

Method III $g^{(n)}_{i,j} \equiv$ the largest value among all flow rates of component i leaving and entering j -th plate at n -th iteration.

$$= \max[V_{j+1}y^{(n)}_{i,j+1}, L_{j-1}x^{(n+1)}_{i,j-1}, F_jz_{ij}, (V_j + W_j)y^{(n)}_{i,j}, (L_j + U_j)x^{(n)}_{i,j}] \quad (5)$$

Then,

$$\mu^{(n)}_{i,j} = (1-\xi) \times \frac{x^{(n)}_{i,j}}{\max[(V_j + W_j)y^{(n)}_{i,j}, (L_j + U_j)x^{(n)}_{i,j}]} \quad (8)$$

Also in this case, $x^{(n+1)}_{i,j}$ calculated by Eqs.(3) and (8) is not always positive. When the value is negative, the same recalculation procedure as that of Method III is taken.

Five examples listed in Table 1 were considered for testing the rate and stability of converge-

Table 2 Comparison of required number of iterations obtained by using Methods I, II, III and IV for the examples shown in Table 1

Method	Method Required number of iterations*				
	Case 1	Case 2	Case 3	Case 4	Case 5
I	379	236	658	208	365
II	379	238	654	209	366
III	111	85	70	68	145
IV	97	not converged	51	43	130

* Convergence criteria

$$\frac{\left| \sum_j F_j z_{i,j} - \sum_j (U_j x_{i,j}^{(n)} + W_j y_{i,j}^{(n)}) \right|}{\sum_j F_j z_{i,j}} < 0.001, \text{ for each component}$$

$$\frac{|x_{i,j}^{(n)} - x_{i,j}^{(n-1)}|}{x_{i,j}^{(n)}} < 0.001, \text{ for each component on each plate}$$

nce of the above four methods. All examples are non-ideal. Case 1 is a conventional distillation which

Ishikawa and Hirata⁴⁾ used for the test of their method. Cases 2 to 5 are extractive distillations

Table 3 Examples for the comparison of Methods III,V,VI and VII

Case No.	6	7	8	9	10*	
System (Components)	(1) Water (2) Ethanol (3) Methanol	(1) Water (2) Ethanol (3) Methanol	(1) Acetone (2) Methanol (3) Water	(1) Methanol (2) Ethanol (3) n-Propanol (4) Water	(1) Water (2) Acetone (3) Furfural**	
Feed	No. 1	j F q z_i 6 1.0 1.0 0.5 0.2 0.3	j F q z_i 6 300 1.0 0.3 0.6	j F q z_i 5 1.0 0.0 0.7 0.2 0.1	j F q z_i 7 1.0 0.0 0.4 0.3 0.2 0.1	j F q z_i 8 0.5 1.0 0.0 0.0 1.0
	No. 2		12 400 0.5 0.3 0.5	10 1.0 1.0 0.5 0.3 0.2	15 1.0 1.0 0.25 0.25 0.25 0.25	16 0.5 1.0 0.0 0.0 1.0
	No. 3		15 300 1.0 0.3 0.4 0.3	15 1.0 0.0 0.3 0.4 0.3	23 1.0 1.0 0.1 0.2 0.3 0.4	24 1.0 1.0 0.01 0.99 0.00
	No. 4			20 1.0 1.0 0.2 0.3 0.5		
	No. 5			25 1.0 1.0 0.1 0.2 0.7		
Product	No. 1	j Phase Amount	j Phase Amount	j Phase Amount	j Phase Amount	j Phase Amount
	No. 2	1 (D) liq. 0.5	1 (D) liq. 300	1 (D) liq. 1.0	1 (D) liq. 1.0	1 (D) liq. 0.8
	No. 3	11 (B) liq. 0.5	8 vap. 200	1 (D) vap. 1.0	4 vap. 0.5	31 (B) liq. 1.2
	No. 4		13 liq. 200	8 vap. 0.5	26 liq. 0.5	
	No. 5		18 (B) liq. 300	22 liq. 0.5 30 (B) liq. 2.0	30 (B) liq. 1.0	
N/R	11/3.0	18/3.0	30/3.0	30/3.0	31/7.0	
Eq. for activity coefficient	Margules	Margules	Wilson	Wilson	Margules	

*Extractive distillation column **Solvent component

with one main feed and one or two solvent feeds. The number of iterations required for convergence are listed in Table 2. In the comparison, ξ was fixed at 0, and the initial liquid composition on all plates was set by the average mole fraction of all feeds, in all cases.

Method IV gives the fastest convergence in some cases, but fails to converge depending on cases. This results in giving too large value of the relaxation factor. The results in Table 2 and our experience with other extensive calculations indicates that method III is superior over the other methods.

In comparison, the Successive Point Relaxation Method and the Group Relaxation Method of Ishikawa and Hirata required 1091 and 671 iterations respectively for convergence in case 1.⁴⁾

5. Methods of Calculation Procedure

In the above discussion, Eq.(3) was used as the working equation, and the numerical calculations were proceeded plate to plate from the top to the bottom of the column. In the opposite direction, i.e. from the bottom to the top, the calculations may be proceeded by the similar working equation:

$$x^{(n+1)}_{i,j} = x^{(n)}_{i,j} + \mu^{(n)}_{i,j} [\{ V_{j+1} y^{(n+1)}_{i,j+1} + L_{j-1} x^{(n)}_{i,j-1} + F_{j,z_{i,j}} \} - \{ (V_j + W_j) y^{(n)}_{i,j} + (L_j + U_j) x^{(n)}_{i,j} \}] \tag{9}$$

The rate and stability of convergence also depend on the calculation procedure due to the difference in the locus of the transmission of the com-

position change in the hold-up on each plate. In this section, it is intended to determine the optimum method of the calculation procedure among the following possible methods with single or combination use of Eqs.(3) and (9).

Method III...Single use of Eq.(3)

- (1) Calculate $x^{(n+1)}_{i,j}$ for all "i" by Eq.(3) systematically from the top to the bottom.
- (2) Normalize $x^{(n+1)}_{i,j}$ for all "j".
- (3) Calculate $y^{(n+1)}_{i,j}$ for all "j" by means of the bubble point evaluation.
- (4) Repeat steps 1-3 until $x^{(n+1)}_{i,j}$ converges.

Method V...Single use of Eq. (9)

- (1) Calculate $x^{(n+1)}_{i,j}$ at the bottom for all "i" by Eq.(9).
- (2) Normalize $x^{(n+1)}_{i,j}$ and calculate $y^{(n+1)}_{i,j}$ at the bottom by means of the bubble point evaluation.
- (3) Steps 1-2 are preceded upward plate to plate systematically from the bottom to the top.
- (4) Repeat steps 1-3 until $x^{(n+1)}_{i,j}$ converges.

Method VI...Combination use of Eqs.(3) and (9)

- (1) Steps 1-3 of method III.
- (2) Steps 1-3 of method V.
- (3) Repeat steps 1-2 until $x^{(n+1)}_{i,j}$ converges.

Method VII...Combination use of Eqs.(3) and (9)

- (1) Steps 1-2 of method III.
- (2) Steps 1-3 of method V.
- (3) Repeat steps 1-2 until $x^{(n+1)}_{i,j}$ converges.

In all methods of the calculation procedure, $\mu^{(n)}_{i,j}$ of method III discussed in the preceding section is used.

Table 4 Comparison of required number of iterations obtained by using Methods III, V, VI and VII for the examples shown in Table 3

Method	Required number of iterations*				
	Case 6	Case 7	Case 8	Case 9	Case 10
III	79	33	250	65	84
V	127	32	110	75	449
VI	65	52	351	76	96
VII	66	24	139	not converged	46

*Convergence criteria are identical with those of Table 2

Additional 5 examples listed in Table 3 were considered for testing the availability of these methods. The results are summarized in Table 4. Iterations are counted by a use of Eq.(3) or Eq.(9). The results indicate that the single use of Eq.(9)

and the combination use of Eqs.(3) and (9) give the improvement in some particular cases but not in general. It may therefore be reasonable to recommend method III for use in all cases because of its simplicity.

6. Application to Multiple Distillation Columns Interlinked by Recycle Streams.

The former works^{2,6,10)} on this subject took use of Thiele-Geddes method or matrix method with a specialized calculation procedure and convergence method. On the other hand, by use of the relaxation method, this interlinked columns can be simultaneously solved almost analogously to a single column by the following working equation:

$$x_{i,j,k}^{(n+1)} = x_{i,i,k}^{(n)} + \mu_{i,j,k}^{(n)} [\{ V_{j+1,k} y_{i,j+1,k}^{(n)} + L_{j-1,k} \times x_{i,j-1,k}^{(n+1)} + F_{j,k} z_{i,j,k}^* \} - \{ (V_{j,k} + W_{j,k}) y_{i,j,k}^{(n)} + (L_{j,k} + U_{j,k}) x_{i,j,k}^{(n)} \}] \quad (20)$$

where

$$\mu_{i,j,k}^{(n)} = \Delta t_{i,j,k}^{(n)} / H_{j,k}^L \quad (21)$$

in which the suffix *k* denotes the column number and *z*^{*}_{*i,j,k*} represents the composition of feed or recycle feed stream into *k*-th column. $\mu_{i,j,k}^{(n)}$ is $\mu_{i,j}^{(n)}$ of *k*-th column.

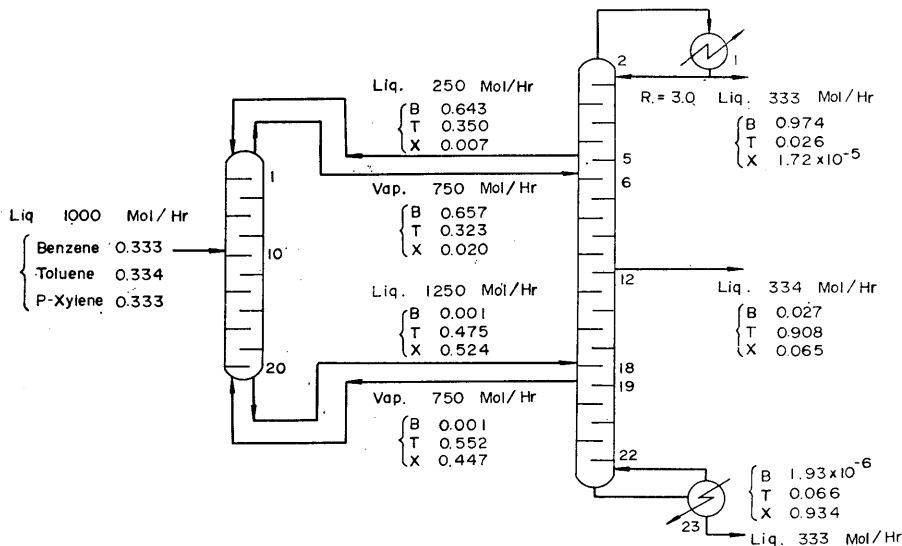


Fig. 2 A thermally coupled distillation system (an example of multiple distillation columns interlinked by recycle streams)

The thermally coupled distillation system proposed by Petlyuk et. al.^{7,8)} was considered for testing the applicability of method III. The system is shown in Fig. 2. The second column is an ordinary complex column with two feeds and three side-cut streams. The first column has neither a condenser nor a reboiler, and its reflux liquid and reboiling vapor are supplied from the second column. Separation of equimolar mixture of benzene, toluene and *p*-xylene is under consideration as an example. Specifications such as number of plates, flow rates and reflux ratio are indicated in Fig. 2 along with the calculated results of mole fractions of products and recycle streams.

Calculations were proceeded having the stream interrelation table between the columns and following the procedure replaced "j" of method III by "j" and "k". The solution was obtained by 139 iterations.

The rate of convergence may be regarded as within the tolerance.

7. Conclusion

A new method is developed for determining the value of the relaxation factor. Although this value may not be optimum from the stand point of minimizing the number of iterations, it gives assurance of stable and rapid approach toward the solution. The advantages of the proposed method are the simplicity and the generality of the logic which requires no special empirical techniques, and the applicability to wide variety of distillation problems regardless of the extent of non-ideality and number of feeds or side draw off streams.

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Nomenclature

- B =molar flow rate of bottoms
 D =molar flow rate of distillate
 F =molar flow rate of column feed
 g =molar flow rate defined by Eqs.(9), (12), (15) or (17)
 H^L_j =liquid hold-up on j -th plate
 L =molar flow of liquid
 N =total number of plates including condenser and reboiler
 q =thermal factor of column feed
 R =reflux ratio
 Δt =time interval
 U =molar flow rate of liquid product draw off
 V =molar flow rate of vapor
 W =molar flow rate of vapor product draw off
 x =mole fraction of liquid
 y =mole fraction of vapor
 z =mole fraction of column feed
 μ =relaxation factor
 ξ =parameter
 $\max[]$ =denotes the maximum value in[]

<Subscripts>

i =component number

j =plate number

k =column number

<Superscript>

(n) = n -th iteration

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