

TEARING ALGORITHMS FOR SEPARATION PROCESS SIMULATION

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Abstract—A modified sum-rates method based on insights provided by an analysis given by Sridhar and Lucia (*Ind. Engng Chem. Res.* **28**, 793-803, 1989) is presented. Newton's method is used to accelerate the inner loop of combined mass balance and phase equilibrium equations and to solve the outer loop of energy balance equations. All partial derivative information is obtained in analytical form. Several literature examples are used to show that the proposed algorithm is more reliable and more efficient than traditional sum-rates methods and provides the capability of solving problems involving intermediate and narrow boiling mixtures.

1. INTRODUCTION

Bubble point and sum-rates methods belong to a class of methods known as equation-tearing algorithms, and they are still readily available in commercial process simulators such as ASPEN Plus and Design II for simulating multistage separation processes, for those who prefer to use them. While many modifications have been suggested over the years, guidelines for their applicability have remained steadfast. Bubble point methods are traditionally recommended for narrow boiling mixtures; sum-rates methods are usually applied to wide boiling mixtures. Intermediate boiling mixtures, on the other hand, represent something of a dichotomy (see for example, Friday and Smith, 1964).

In a recent manuscript, Sridhar and Lucia (1989) provide a rigorous analysis of multistage separation processes involving homogeneous binary mixtures. The main objectives of this paper are to illustrate that insights from this analysis can be used to develop a modified sum-rates algorithm and to demonstrate that this modified method is an improvement over existing algorithms.

2. SUM-RATES ALGORITHMS

It is widely accepted that for relatively high values of Δ_{DB} , where Δ_{DB} is the difference between the bubble and dew point temperatures of the primary feed, sum-rates algorithms will solve separation pro-

cess model equations easily. The model equations that must be solved are the mass balance, phase equilibrium and energy balance equations, and the unknown variables that are calculated are the temperature T_j , and the liquid and vapor component flows l_{ij} and v_{ij} , for all stages. The liquid and vapor component flows are computed in an inner loop, in which the temperature and pressure of each stage is held fixed, by solving the mass balance and phase equilibrium equations in a stage-to-stage manner. The temperature profile, on the other hand, is adjusted in an outer loop using the energy balance relationships. The reader is referred to the papers by Sujata (1961) and Burningham and Otto (1967) for the details of traditional sum-rates methods. The remainder of this section is concerned with a presentation of the salient features of a sum-rates method in the context of the analysis given in Sridhar and Lucia (1989).

2.1. The inner loop

Our calculational procedure for the inner loop involves decoupling the stages of the process, and traversing the separator by solving the mass balance and phase equilibrium equations for each stage simultaneously. Single-stage perturbation relationships derived by Sridhar and Lucia are then used to reassemble the column model as a fixed-point iteration of the form:

$$X^{k+1} = G(X^k), \quad (1)$$

where $X^T = (v_2^T, v_3^T, \dots, v_n^T)^T$. The Jacobian matrix of this fixed-point iteration is given by:

$$G' = \begin{bmatrix} M_2(I - M_1) & M_2 & 0 & 0 \dots 0 \\ M_3(I - M_2)(I - M_1) & M_3(I - M_2) & M_3 & 0 \dots 0 \\ \vdots & \vdots & \vdots & \vdots \\ \vdots & \vdots & \vdots & M_{n-1} \\ M_n(I - M_{n-1}) \dots (I - M_1) & M_n(I - M_{n-1}) \dots (I - M_2) & \dots & M_n(I - M_{n-1}) \end{bmatrix}, \quad (2)$$

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where $M_j = (\nabla^2 G_j^V + \nabla^2 G_j^L)^{-1} \nabla^2 G_j^L$, and where $\nabla^2 G_j^L$ and $\nabla^2 G_j^V$ are the Hessian matrices of the Gibbs free energy function for the liquid and vapor phases associated with stage j .

It is more convenient to express the fixed-point iteration given by equation (1) in the form:

$$F(X) = X - G(X) = 0, \quad (3)$$

where the corresponding Jacobian matrix of equation (3) is given by:

$$F'(X) = I - G'(X). \quad (4)$$

While many choices of approximations to $F'(X)$ exist, we prefer to use Newton acceleration of the inner loop, in which the iterates are computed using the equation:

$$X^{k+1} = X^k - [F']^{-1} F(X^k), \quad (5)$$

where $G'(X)$ is calculated using equation (2).

2.2. The outer loop

The temperature profile for the separator is adjusted in an outer loop using the energy balance equations and results in the iteration:

$$T^{k+1} = T^k + \Delta T^k, \quad (6)$$

where

$$\Delta T^k = \left[\frac{\partial Q_i}{\partial T_j} \right]^{-1} (Q - \hat{Q}), \quad (7)$$

and where $Q = (Q_1, Q_2, \dots, Q_n)^T$ is a vector of stage heat duty specifications and \hat{Q} is a similar vector of heat duties that satisfies the single-stage energy balance equations. In order to use equation (7), the Jacobian matrix, $[\partial Q_i / \partial T_j]$, must be constructed. In traditional sum-rates algorithms, the Jacobian matrix is approximated by a tridiagonal matrix using *only* changes in molar enthalpy with respect to temperature. That is:

$$\frac{\partial Q_i}{\partial T_j} = L_j \frac{\partial H_j^L}{\partial T_j} + V_j \frac{\partial H_j^V}{\partial T_j}. \quad (8)$$

In cases where the sensitivity of flows to variations in temperature is high, as in mixtures that are not very wide boiling, such a simplified Jacobian matrix can be a poor approximation.

In order to obtain a more accurate Jacobian matrix approximation, it is essential to be able to compute the variation of liquid and vapor molar flows with respect to temperature. An analytical expression for the variation of the vapor profile with respect to temperature was developed by Sridhar and Lucia, and is a direct consequence of Newton acceleration of the inner loop. In particular, the variation in vapor component flows Δv with respect to temperature perturbations ΔT is given by:

$$\Delta v = (I - G')^{-1} r, \quad (9)$$

where G' is defined by equation (2), where the vector r is given by:

$$r = \begin{bmatrix} r_2 \\ r_3 \\ r_4 \\ \vdots \\ r_n \end{bmatrix} = \begin{bmatrix} t_2 - M_2 t_1 \\ t_3 - M_3 t_2 - M_3 (I - M_2) t_1 \\ t_4 - M_4 t_3 - M_4 (I - M_3) t_2 - \\ \quad M_4 (I - M_3) (I - M_2) t_1 \\ \vdots \end{bmatrix}. \quad (10)$$

and where

$$t_k = (\nabla^2 G_k^L + \nabla^2 G_k^V)^{-1} (\bar{H}_k^V - \bar{H}_k^L) \frac{\Delta T_k}{T_k}. \quad (11)$$

With this, a tridiagonal approximation of the Jacobian matrix for the outer loop can be constructed. For example, for $k = 1, \dots, n_r$, the diagonal terms of the Jacobian matrix are given by:

$$\begin{aligned} \frac{\partial Q_k}{\partial T_k} &= H_k^V \left(\frac{\partial V_k}{\partial T_k} \right) + V_k \left(\frac{\partial H_k^V}{\partial T_k} \right) \\ &+ H_k^L \left(\frac{\partial L_k}{\partial T_k} \right) + L_k \left(\frac{\partial H_k^L}{\partial T_k} \right) \\ &- H_{k+1}^V \left(\frac{\partial V_{k+1}}{\partial T_k} \right) - H_{k-1}^L \left(\frac{\partial L_{k-1}}{\partial T_k} \right). \end{aligned} \quad (12)$$

Furthermore, the nontraditional terms of the Jacobian matrix are, as noted, easily obtained in analytical form as a byproduct of accelerating the inner loop by Newton's method.

3. A MODIFIED SUM-RATES

A flowchart of the proposed algorithm is given in Fig. 1. The stages of the separator are decoupled and the mass balance and phase equilibrium relationships for each stage are solved using Newton-like methods. Asymmetric trust region methods are used to ensure that the iteration variables for the single-stage flash calculations (i.e. the component flowrates) remain feasible and to improve the reliability of Newton-like methods on these, and other, small subproblems (see Sridhar, 1990 for details). Convergence is assumed when the two-norm of the mass balance and equilibrium equations reaches a value less than 10^{-5} .

Convergence of the single-stage calculations for all stages results in a new vapor component flowrate profile. Equation (2) is then used to accelerate the inner loop calculations. The single-stage and acceleration calculations are repeated alternately until convergence of the inner loop to a tolerance of 10^{-5} is obtained. Typically, 3-5 Newton acceleration iterations are required. Equations (6) and (7) are then used to calculate a new temperature profile. The outer loop is assumed to be converged when

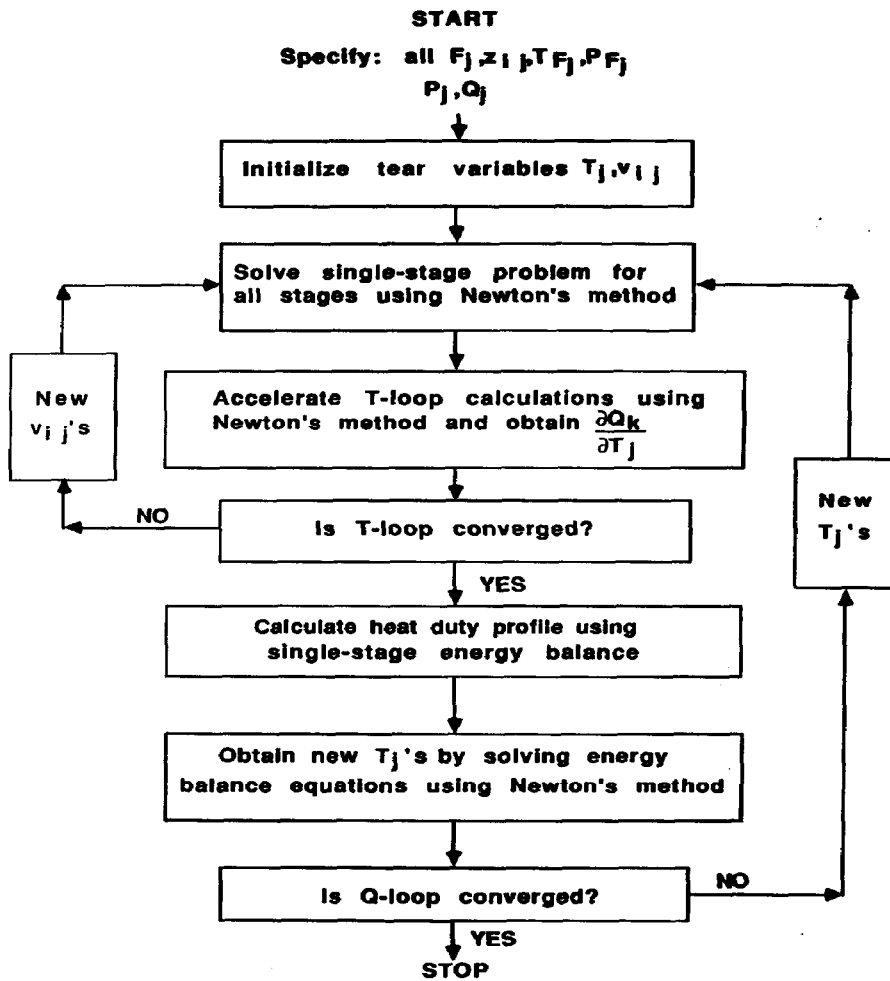


Fig. 1. Flowchart for sum-rates algorithm.

the two-norm of the energy balance equations falls below a tolerance of 10^2 . Initialization of all variables at all levels of computation is done automatically (see Sridhar, 1990) and Newton-like methods are used for all equation-solving tasks.

4. NUMERICAL RESULTS AND DISCUSSION

In this section, the numerical performance of the proposed algorithm is compared to that of traditional sum-rates methods on a set of six example problems taken from the literature. The procedures and data given in Prausnitz *et al.* (1980) were used to model the physical properties in all cases, and all calculations were done on a Gould 9080 computer in double-precision arithmetic.

4.1. Numerical results

A description of the example problems is given in Table 1, and the performance of both algorithms is

compared in Table 2. Overall, it was found that the modified algorithm took fewer iterations than the traditional sum-rates algorithm on problems involving wide boiling mixtures. On the other hand, for problems involving intermediate and narrow boiling mixtures, the traditional sum-rates algorithm failed to converge, while the modified sum-rates algorithm converged to the solution.

4.2. Discussion of a sample problem

Consider a problem, such as Example 5, involving a mixture which can be classified as intermediate boiling. As seen in Table 2, the traditional sum-rates method failed on this problem while the modified sum-rates algorithm converged in six iterations. This marked difference in numerical performance is due to significant differences in the Jacobian matrix approximations $[\partial Q_k / \partial T_j]$ for the two algorithms. Note that the initial Jacobian matrix for the traditional sum-rates algorithm, in which only the changes

Table 1. Problem descriptions

Problem No. (Reference)	No. of stages	Pressure (MPa)	Heat duty specifications (MJ h ⁻¹)	Feed specifications Flowrates (kmol h ⁻¹)
1 (Shinohara <i>et al.</i> , 1972)	6	0.5175	$Q_j = 0$	1 liquid, 1 vapor stage 1, 305.22 K; stage 6, 288.55 K 0.0 C ₁ 1644.11 0.0 nC ₄ 51.439 0.0 C ₂ 166.19 0.0 nC ₅ 23.74 0.0 C ₃ 94.96 533.0 nC ₁₀ 0
2 (Shinohara <i>et al.</i> , 1972)	6	0.5175	$Q_3 = -1$ $Q_j = 0, j \neq 3$	Same as 1
3 (Henley and Seader, 1981) pp. 465-66	6	2.76	$Q_j = 0$	1 liquid, 1 vapor stage 1, 305.22 K; stage 6, 313.55 K 0 C ₁ 160 0 nC ₄ 25 0 C ₂ 370 0.78 nC ₅ 5 0 C ₃ 240 164.17 nC ₁₀ 0
4	8	1.013	$Q_1 = -2.0113$ $Q_j = 0$ $j \neq 1, 8$ $Q_8 = 2.615$	1 liquid stage 2, 176.15 K C ₂ H ₄ 50 C ₂ H ₆ 50
5 (Shinohara <i>et al.</i> , 1972)	4	4.013	$Q_1 = -1.994$ $Q_2 = 8.371$ $Q_3 = 4.935$ $Q_4 = 4.452$	1 liquid stage 2, 343.44 K nC ₄ 40 nC ₁₀ 30
6 (Shinohara <i>et al.</i> , 1972)	8	1.013	$Q_1 = -66.69$ $Q_2 = -2.274$ $Q_3 = 0.265$ $Q_4 = 1.023$ $Q_5 = -0.34$ $Q_6 = 0.2259$ $Q_7 = 0.566$ $Q_8 = 81.67$	1 liquid stage 4, 348.45 K MeOH 40 H ₂ O 60

in enthalpy with respect to temperature are considered, is:

$$\left[\frac{\partial Q_i}{\partial T_j} \right] = \begin{bmatrix} 5316.78 & -5712.66 & 0 & 0 \\ -81.2938 & 17,313 & -1977.8 & 0 \\ 0 & -11,600.3 & 13,536.4 & -532.389 \\ 0 & 0 & -12,338.6 & 13,276.9 \end{bmatrix} \quad (13)$$

On the other hand, the initial Jacobian matrix for the proposed algorithm, in which changes in total flow with respect to temperature are also included, is:

$$\left[\frac{\partial Q_i}{\partial T_j} \right] = \begin{bmatrix} 7289.57 & -11,388.57 & 0 & 0 \\ -2770.31 & 29,888.7 & -3532.82 & 0 \\ 0 & -18,500 & 19,656.9 & -3033.64 \\ 0 & 0 & -16,124.1 & 16,961.2 \end{bmatrix} \quad (14)$$

It is easily seen that the Jacobian matrix approximation for the traditional sum-rates algorithm differs significantly from the one for the modified sum-rates method. Furthermore, our experience shows that the corresponding temperature step obtained from

equation (7) by the traditional sum-rates algorithm is often a poor one, and that a series of such steps frequently causes the temperatures of some of the stages in the separator to leave the two-phase region, and that this usually results in the failure of traditional sum-rates algorithms. In contrast, the modified sum-rates method results in good temperature steps and usually converges in relatively few iterations.

5. CONCLUSIONS

Rigorous mathematical analysis was used to develop a modified sum-rates method for simulating multistage separation processes. This modified

Table 2. Numerical results

Problem No.	Iterations	
	Modified sum-rates	Traditional sum-rates
1	5	9
2	5	9
3	6	7
4	12	F
5	6	F
6	26	F

algorithm was shown to be more reliable for solving problems involving intermediate and narrow boiling mixtures and more efficient for problems involving wide boiling mixtures than traditional sum-rates methods. Newton's method was used to solve the appropriate model equations at all levels of computation, including the initialization strategy for the single-stage flash calculations, and asymmetric trust region methods were used to guarantee feasible iterates for related bubble point, dew point and flash calculations. No reliability difficulties were experienced and, consequently, the proposed sum-rates method performed very well on the example problems tested.

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NOMENCLATURE

G^L, G^V = Total Gibbs free energy of liquid phase, of vapor phase.
 G' = Jacobian matrix of fixed-point iteration
 H^L, H^V = Molar enthalpy of liquid phase, of vapor phase
 \bar{H}^L, \bar{H}^V = Partial molar enthalpy of liquid phase, of vapor phase
 L = Total liquid flowrate
 Q = Input heat duty to any stage
 T = Temperature
 V = Total vapor flowrate

Subscripts

i = Component index
 j, k = Stage index

Superscripts

k = Iteration counter
 L, V = Liquid, vapor

Greek letter

Δ = Perturbation in any variable.

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