Interlinked, Multistaged Separators with Nonstandard Specifications Solved by the Newton-Raphson Method

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We shall assume as standard specifications, the following conditions:

Variables: x_{ij}, y_{ij}, V_j, L_j, T_j
Parameters: F_j, z_{ij}, Q_j, U_j, W_j $\quad (i=1,M;\ j=1,N)$

where M stands for the number of components and N for the sum of stages of the considered distillation columns. The choice of the above variables and parameters is not absolutely necessary for the proposed algorithm; for the sake of simplicity, we start by considering them as standard specifications.

Therefore, the linearized equations of component material balance, equilibrium and stoichiometric relationships, global material, and enthalpy balances, written for each stage, as a whole, form a system showing the well-known tridiagonal submatrices structure, if interlinks are not present. This base-matrix is shown in Figure 1 which, for the case at hand, is formed by the submatrices $A_j(K,K)$, $B_j(K,K)$, $C_j(K,K)$, and t_j (K).

If one or more of the parameters, F_j, z_{ij}, Q_j, U_j, and W_j, is considered as an unknown variable (this happens whenever nonstandard specification functions are assigned), the tridiagonal-block structure of the coefficient matrix is not always guaranteed. Naphtali and Sandholm (1971), indeed, showed a particular situation where nonstandard specification functions saved the tridiagonal-block structure. In fact, they assumed that the new variable is Q_j, and that the specification equation is a function only of the variables of stage j, for instance T_j=const. In this case, it is possible to save the tridiagonal-block structure by substituting the enthalpy balance on stage j with the specification equation. But, consider the following cases:

(1) If the duty Q_j is a variable but the specification equation that substitutes the enthalpy balance on stage j is related to variables of stage k $(k\neq j)$, two consequences follow: the whole matrix cannot have any longer a tridiagonal-block structure and the submatrix B_j contains a zero-row which prevents its inversion (Hofeling and Seader, 1978; Buzzi Ferraris, 1980).

(2) If we choose one of the remaining parameters, i.e., F_j, z_{ij}, U_j, and W_j, as an unknown variable, it is quite impossible to substitute one of the linearized equations with a nonstandard specification equation. The tridiagonal-block structure may be formally saved only in the case of specification equations related to the same stage of the new variable at the cost of enlarging the submatrices, A_j, B_j, C_j, and t_j.

SPECIFICATION EQUATIONS

If the goal is an algorithm capable of treating the widest range of situations, it is more convenient to adopt a different approach, as developed in this note. If one or more of the parameters, F_j, z_{ij}, Q_j, U_j, and W_j, are considered as unknown variables, we introduce (in the right side of the base-matrix) a submatrix $D_j(K,L)$ with as many columns as the number (L) of the new variables, while the rows number (K) is the same as A_j, B_j, C_j, and t_j. D_j contains the coefficients of the increments of the new variables, which are obtained by linearization of the basic equations written for the stage j at hand, i.e., component material balances, equilibrium relationships, etc.

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The number of specification equations that must be given is obviously equal to the number (L) of the new variables. Consider the specification equations as functions of whatever complexity of the entire set of variables (i.e., the original and the new ones); for instance, T_2=const.; V_4/L_4=const.; $(U_1x_{5,1})/(F_7z_{5,7})$=const. etc. The linearized specification equations are introduced at the bottom of the base-matrix to form the new submatrices, $P_j(L,K)$ and $G(L,L)$, as well as the vector $t_G(L)$. P_j contains the coefficients of the increments of the original variables in the linearized specification equations, while G contains those of the new variables. The vector t_G contains the negative of the residual of each specification equation. The shape of the new base-matrix is shown in Figure 2, while Table 1 reports the algorithm to solve the linear system.

Consider now an interlink from stage r to n $(r>n+1)$ and let $UX_{r,n}$ and $WY_{r,n}$ stand, respectively, for the liquid and vapor flow rates from stage r to stage n. The basic equations are properly modified by the introduction of terms in these quantities. As for the linearized equations, this results in a modification of the B_r submatrix as well in the introduction of a new one, $S_{n,r}(K,K)$, which lies in the (n,r) position above the tridiagonal-block. If $UX_{r,n}$ and/or $WY_{r,n}$ are considered as parameters, no modification occurs in the D_n submatrix. Otherwise, should $UX_{r,n}$ and/or $WY_{r,n}$ be taken as variables, D_n is enlarged to contain as many columns as the number of new variables.

If an interlink occurs from the stage m to k $(k>m+1)$, the new terms in $UX_{m,k}$ and $WY_{m,k}$ in the basic equations, both modify the submatrix B_m and introduce a new one $E_{k,m}(K,K)$ under the tridiagonal-block. In this case too, $UX_{m,k}$ and/or $WY_{m,k}$ may be considered as either parameters or variables, in agreement with what was previously stated.

The shape of the base-matrix, when interlinks are present and nonstandard specifications are considered, is shown in Figure 3, where for sake of simplicity only two S and E type submatrices are present.

Some Authors (Ketchum, 1975; Kubicek et al., 1976) have proposed an interesting, but not completely satisfactory, al-

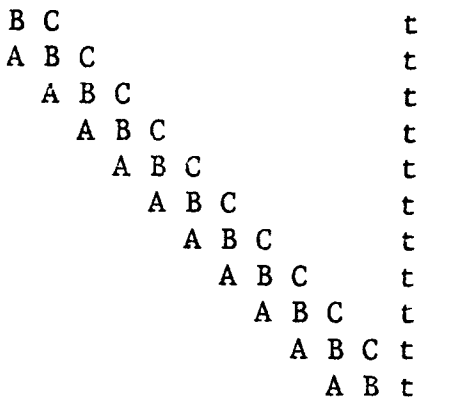

Figure 1.

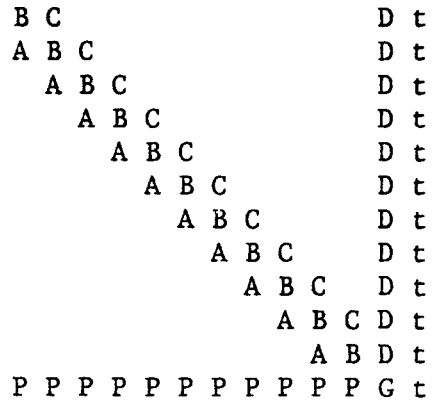

Figure 2.

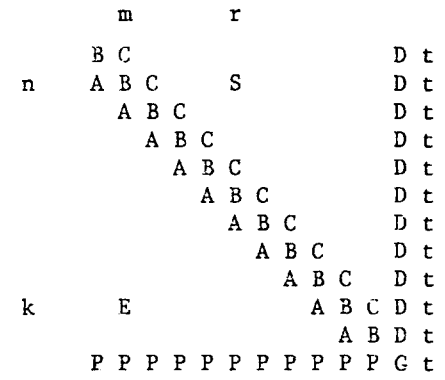

Figure 3.

gorithm to solve the base-matrix in Figure 3. In fact, they eliminate the submatrices of S and E type by means of the introduction of new variables and equations that enlarge the dimensions of the submatrices of D, P, and G type. For instance the system:

$$\begin{aligned}
b_1x_1+c_1x_2 \qquad\qquad +s_1x_4 \qquad\qquad +d_1x_6&=t_1\\
a_2x_1+b_2x_2+c_2x_3 \qquad\qquad\qquad +d_2x_6&=t_2\\
a_3x_2+b_3x_3+c_3x_4 \qquad\qquad d_3x_6&=t_3\\
a_4x_3+b_4x_4+c_4x_5+d_4x_6&=t_4\\
e_5x_1 \qquad\qquad a_5x_4+b_5x_5+d_5x_6&=t_5\\
p_1x_1+p_2x_2+p_3x_3+p_4x_4+p_5x_5+g_6x_6&=t_6
\end{aligned}$$

is converted into the following equivalent one by the introduction of the new variables x_7, x_8:

TABLE 1. SEE FIGURE 2

	$j=1,N$ (Step 1,9)		
Step 1	C_j	$\leftarrow B_j^{-1}C_j$	$j\leq N-1$
Step 2	D_j	$\leftarrow B_j^{-1}D_j$	$j\leq N$
Step 3	t_j	$\leftarrow B_j^{-1}t_j$	$j\leq N$
Step 4	B_{j+1}	$\leftarrow B_{j+1}-A_{j+1}C_j$	$j\leq N-1$
Step 5	D_{j+1}	$\leftarrow D_{j+1}-A_{j+1}D_j$	$j\leq N-1$
Step 6	t_{j+1}	$\leftarrow t_{j+1}-A_{j+1}t_j$	$j\leq N-1$
Step 7	P_{j+1}	$\leftarrow P_{j+1}-P_jC_j$	$j\leq N-1$
Step 8	G	$\leftarrow G-P_jD_j$	$j\leq N$
Step 9	t_G	$\leftarrow t_G-P_jt_j$	$j\leq N$
Step 10	δX_D	$\leftarrow G^{-1}t_G$	
Step 11	δX_N	$\leftarrow t_N-D_N\delta X_D$	
	$j=N-1,\ldots,1$ (Step 12)		
Step 12	δX_j	$\leftarrow t_j-C_j\delta X_{j+1}-D_j\delta X_D$	

$$\begin{aligned}
b_1x_1+c_1x_2 \qquad\qquad +d_1x_6+s_1x_7 \qquad\qquad &=t_1\\
a_2x_1+b_2x_2+c_2x_3 \qquad\qquad +d_2x_6 \qquad\qquad &=t_2\\
a_3x_2+b_3x_3+c_3x_4 \qquad +d_3x_6 \qquad\qquad &=t_3\\
a_4x_3+b_4x_4+c_4x_5+d_4x_6 \qquad\qquad &=t_4\\
a_5x_4+b_5x_5+d_5x_6 \qquad +e_5x_8&=t_5\\
p_1x_1+p_2x_2+p_3x_3+p_4x_4+p_5x_5+g_6x_6 \qquad\qquad &=t_6\\
x_4 \qquad\qquad -x_7 \qquad\qquad &=0\\
x_1 \qquad\qquad\qquad\qquad -x_8&=0
\end{aligned}$$

The coefficients-matrix now has a structure equal to that of Figure 2; therefore, may be solved by the algorithm shown in Table 1. This approach is very efficient and convenient when the number of the newly introduced variables is low. For the problem at hand, the number of new variables is equal to the product of the number of interlinks and the value K. (In general, K is equal to $2M+3$ or, better, to $M+3$ as it will be shown later.) As consequence:

(1) The resulting number of the columns of submatrices D and G, and the rows of P and G may be very large.

(2) Step 10 in Table 1 may be time-consuming.

(3) Steps 5 and 7 require a number of operations that may be avoided. For instance, submatrix $S_{n,r}$ is involved in calculations at Step 5 from stage $j=n$ to $j=N-1$, while the proposed algorithm below stops the calculations at $j=r-2$. A similar benefit is obtained in calculations involving E submatrices in Step 7.

(4) Presumably less accuracy may be obtained due to major round-off errors, as a consequence of the increased number of calculations.

The proposed algorithm is derived from the upper-triangularization method, and applies to the base-matrix of Figure 3 without any preliminary elimination of S and E type subma-

TABLE 2. SEE FIGURE 4

	$j=1,N$ (Step 1,4)
Step 1	$C_j\leftarrow B_j^{-1}C_j$ $(j\leq N-1)$; $S_j\leftarrow B_j^{-1}S_j$ $(n\leq j\leq r-2)$; $D_j\leftarrow B_j^{-1}D_j$ $(j\leq N)$; $t_j\leftarrow B_j^{-1}t_j$ $(j\leq N)$
Step 2	$E_{k,j+1}\leftarrow E_{k,j}C_j$ $(m\leq j\leq r-2)$; $E_{k,r}\leftarrow E_{k,r}-E_{k,j}S_j$ $(m\leq j\leq r-2$, and $n\leq j)$; $E_{k,r}\leftarrow E_{k,r}-E_{k,j}C_j$ $(j=r-1)$; $E_{k,j+1}\leftarrow -E_{k,j}C_j$ $(r\leq j<k-2)$; $A_k\leftarrow A_k-E_{k,j}C_j$ $(j=k-2)$; $D_k\leftarrow D_k-E_{k,j}D_j$ $(m\leq j\leq k-2)$; $t_k\leftarrow t_k-E_{k,j}t_j$ $(m\leq j\leq k-2)$; $E_{k,j}\leftarrow 0$
Step 3	$P_{j+1}\leftarrow P_{j+1}-P_jC_j$ $(j\leq N-1)$; $P_r\leftarrow P_r-P_jS_j$ $(n\leq j\leq r-2)$; $G\leftarrow G-P_jD_j$ $(j\leq N)$; $t_G\leftarrow t_G-P_jt_j$ $(j\leq N)$
Step 4	$B_{j+1}\leftarrow B_{j+1}-A_{j+1}C_j$ $(j\leq N-1)$; $D_{j+1}\leftarrow D_{j+1}-A_{j+1}D_j$ $(j\leq N-1)$; $t_{j+1}\leftarrow t_{j+1}-A_{j+1}t_j$ $(j\leq N-1)$; $S_{j+1}\leftarrow S_{j+1}-A_{j+1}S_j$ $(n\leq j<r-2)$; $C_{j+1}\leftarrow C_{j+1}-A_{j+1}S_j(j=r-2)$
Step 5	$\delta X_D\leftarrow G^{-1}t_G$; $\delta X_N\leftarrow t_N-D_N\delta X_D$
Step 6	$\delta X_j\leftarrow t_j-C_j\delta X_{j+1}-D_j\delta X_D \qquad j=N-1,\ldots,r-1$ $\delta X_j\leftarrow t_j-C_j\delta X_{j+1}-D_j\delta X_D-S_j\delta X_r \qquad j=r-2,\ldots,n$ $\delta X_j\leftarrow t_j-C_j\delta X_{j+1}-D_j\delta X_D \qquad j=n-1,\ldots,1$

A single subscript refers to row of matrix for the submatrices A,B,C,D,S,t, and to column of matrix for the submatrices P. In a double subscript, the first index refers to the row; the second, to the column.

trices. The steps involved in solving such a system are shown in Table 2.

CONCLUSION

1. To start the procedure, it is necessary to introduce into the scheme some zero auxiliary submatrices:

- Each submatrix $S_{n,r}$ $(r>n+2)$ requires the introduction of auxiliary submatrices $S_{n+1,r}$; $S_{n+2,r}$; . . . $S_{r-2,r}$.
- If submatrices $S_{n,r}$ $(r>n+1)$ and $E_{k,m}$ $(k>m+1)$ are simultaneously present and if $m<r$, the following four possibilities occur:

a) $k\leq n$: auxiliary submatrices are not necessary
b) $n<k<r-1$: on row k there is the auxiliary submatrix $S_{k,r}$ which is on the right side of the tridiagonal-block
c) $r-1\leq k\leq r+1$: auxiliary submatrices are not necessary
d) $k>r+1$: on row k, there is the auxiliary submatrix $E_{k,r}$ which is on the left side of the tridiagonal-block.

Therefore, the system of Figure 3 is converted into the system shown in Figure 4.

2. The value N may be, for practical problems, as high as a few hundred, because, as stated before, it stands for the total number of stages of the interlinked columns. If we have a small value of N (for instance, a topping-section), it happens that the number M of components may be wide enough (30 to 50) so that the dimension of the entire system to be solved remains large.

In this situation, it becomes necessary or at least convenient to use auxiliary core memory, and it is worthwhile to optimize the use of the central memory with Input/Output time.

Among the different and possible procedures to do this, we propose the following: Define as "signed" rows, those that contain E-type submatrices. All submatrices, present in each "signed" row, must be first built up and stored in a compact form by referring to them by their column index. If the central memory is not sufficient to store them, it is convenient, if possible, to use as many auxiliary memories as the number of "signed" rows. For the particular system of Figure 4, the submatrices of the "signed" row k are stored in a memory 1 (auxiliary or central) which has the following form:

$$\text{Memory 1:}/E_{k,m}E_{k,r}A_kB_kC_kD_kt_k/$$

The submatrices with column indices $m,r,k-1,k,k+1$, and the D_k submatrix and the t_k vector are the elements contained in it.

The coefficients of nonstandard specification functions are built up and the related submatrices P and G, and the vector t_G should be stored in Memory 2. A storage location inside the central memory is more convenient; otherwise, auxiliary ones must be used. If the latter, storage may be more efficient if it is made row by row for the entire set of submatrices instead of the alternative one, submatrix P by submatrix P. In fact, this will lead to the reading and writing of some large blocks for a few times instead of reading and writing small blocks for many times, because the number of specification functions L is, in general, considerably lower than the number of stages N. The coefficients of the elements B_1, C_1, D_1, and t_1, related to stage number 1, are built up and stored inside the central memory.

The calculations start from Step 1 of Table 2; the obtained results are stored in an auxiliary Memory 3, which is the only one that is practically indispensable.

$$\text{Memory 3: }/C_1,D_1,t_1/C_2,D_2,t_2/\ldots/C_n,S_n,D_n,t_n/\ldots/C_{r-2},S_{r-2},D_{r-2},t_{r-2}/$$
$$/C_{r-1},D_{r-1},t_{r-1}/\ldots/C_{N-1},D_{N-1},t_{N-1}/D_N,t_N/$$

Step 2 is executed for $m\leq j\leq k-2$; in the meantime, Memory 1 will be active for the collection and the storage of information. Then Step 3, which uses Memory 2, is executed.

The execution of Step 4 is preceded by the build-up and storage in central memory of submatrices, A_{j+1},B_{j+1}, C_{j+1},S_{j+1},D_{j+1}, and t_{j+1}, when $j+1\neq k$. Otherwise, $(j+1=k)$; the above submatrices are already at disposal inside Memory 1 from Step 2. This procedure strictly requires the storage of only the submatrices of two rows $j, j+1$ or j, k inside the central memory. The possibility to enlarge storage to "signed" rows and/or to submatrices pertaining to the specification equations must be examined in terms of further facilities offered by the capability of the utilized computer.

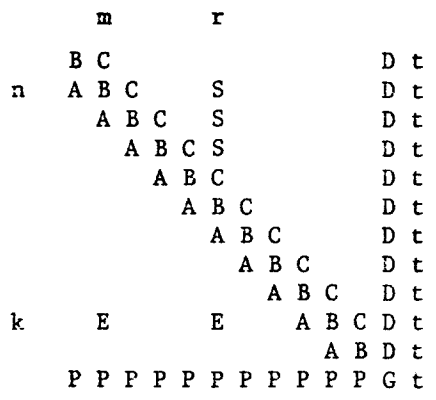

Figure 4.

As the proposed method is by nature a forward one, the E-type submatrices are eliminated by utilizing only once the information of the "not-signed" rows. The utilization of such rows would be iterative, if a backward procedure should be utilized (Hofeling and Seader, 1978; Buzzi Ferraris, 1980).

Therefore, it is convenient to save the above "not-signed" rows inside the central memory. This requirement may be sometimes troublesome. In general, the necessity of central memory is more critical with the backward method than with the forward procedure as the number of "not-signed" rows is higher than that of "signed" ones. (The order of magnitude of their difference may be roughly obtained by comparing the number of interlinks with the number of stages.) Thus, the forward method used here may result in a great saving of I/O time.

The remaining Steps in Table 2 are then applied in sequence, noting that Step 6 utilizes submatrices stored inside Memory 3.

3. The number of submatrices that are simultaneously present in each row remains constant during the computing phase. This peculiarity is related both to the introduction of auxiliary submatrices, as shown, and to the fact that whenever a submatrix is built up ($E_{k,j+1}$ in Step 2) in whichever row, another disappears ($E_{k,j}$ in Step 2) in the same row. The auxiliary submatrices that convert the matrix of Figure 3 to that one of Figure 4 are, in general, a few, so that their storage inside central memory does not create great problems since the central memory is utilized strictly for the storage of the elements of two rows each time.

4. When the residuals of equations are small, it is useful to adopt a simple Quasi-Newton method. For this purpose, it is necessary both to calculate, in the preceding iteration, the submatrices, B_j^{-1} and to store the submatrices, B_j^{-1}, $E_{k,j},A_{j+1},P_j,G^{-1}$, in an auxiliary Memory 4. In this way, it is required to execute only the steps that involve the new residuals t_j of the equations. The solution obtained is the same that would be obtained considering as constant the Jacobi matrix of the system.

5. Some authors eliminate from the system the variables y_{ij}, substituting them with $K_{ij}x_{ij}$. When the K-ratios are weakly dependent on y, it is better to use the following linearized equilibrium equation for δy_{ij}:

$$\delta y_{ij} = -\phi_{ij} + K_{ij}\delta x_{ij} + x_{ij}\frac{\delta K_{ij}}{\delta T_j}\delta T_j + \sum_{1^n}^{M} x_{ij}\frac{\delta K_{ij}}{\delta x_{nj}}\delta x_{nj}$$

where ϕ_{ij} is the residual of equilibrium equation: $y_{ij}-K_{ij}x_{ij}=0$. The substitution of this expression within the remaining linearized equations strongly reduces both the submatrices dimension and the computing time necessary to solve the linearized system.

6. If

$$h_j = \sum_{1^n}^{M} h^m_{nj}x_{nj}$$

and

$$H_j = \sum_{1^n}^{M} H^m_{nj} y_{nj}$$

then,

$$\frac{\partial h_j}{\partial x_{nj}} = h^m_{nj}$$

and

$$\frac{\partial H_j}{\partial y_{nj}} = H^m_{nj}$$

It is always convenient to use these relationships (neglecting possible heats of mixing), when computing the derivatives.

7. Numerical computation of the derivatives $\partial K_{ij}/\partial T_j$ is more conveniently performed only on few selected trays. For the remaining ones, a very good approximation is obtained from the relation:

$$\frac{\partial K_{ij}}{\partial T_j} = a \frac{K_{ij}}{T_j^2}$$

where the parameter a is given by linear interpolation between two trays, where the computation has been performed numerically.

8. Strong dependence of K_{ij} on liquid and/or vapor composition may require the introduction of terms:

$$\frac{\partial K_{ij}}{\partial x_{nj}}, \frac{\partial K_{ij}}{\partial y_{nj}}$$

The evaluation time for these derivatives is relatively long; therefore, some loops without this computation may be more conveniently performed at the beginning. Then, the derivatives are evaluated only for a few selected stages, interpolating them linearly for the others. If convergence is not reached by this procedure, it may be useful to evaluate them for each stage. Not all interlinked columns show this problem. So, it is necessary only to use this procedure for those that require it.

NOTATION

A_j = submatrix of partial derivatives of linearized equations on stage j with respect to variables on stage $j-1$
B_j = submatrix of partial derivatives of linearized equations on stage j with respect to variables on stage j
C_j = submatrix of partial derivatives of linearized equations on stage j with respect to variables on stage $j+1$
D_j = submatrix of partial derivatives of linearized equations on stage j with respect to nonstandard variables
$E_{k,m}$ = submatrix of partial derivatives of linearized equations on stage k with respect to variables on stage m ($k>m+1$)
F_j = total feed to stage j
G = submatrix of partial derivatives of linearized specification equations with respect to nonstandard variables
h_j = molar enthalpy of the stream L_j
H_j = molar enthalpy of the stream V_j
K_{ij} = equilibrium constant for component i, tray j
K = row and column number for submatrices A,B,C
L_j = liquid stream to state $j+1$
L = number of new variables and of nonstandard specification equations
M = number of components
N = total number of trays for all the columns
P_j = submatrix of partial derivatives of linearized specification equations with respect to variables on stage j
Q_j = heat from stage j
$S_{n,r}$ = submatrix of partial derivatives of linearized equations on stage n with respect to variables on stage r ($r>n+1$)
T_j = temperature on stage j
t_j = residuals of the equations associated to the stage j, with the opposite sign
t_G = residuals of the nonstandard specification equations, with the opposite sign
U_j = liquid-side stream from stage j
$UX_{r,n}$ = liquid total flow rate from stage r to stage n
V_j = vapor stream to stage $j-1$
W_j = vapor-side stream from stage j
$WY_{r,n}$ = vapor total flow rate from stage r to stage n
z_{ij} = molar fraction of component i in F_j
x_{ij} = molar fraction of component i in L_j
y_{ij} = molar fraction of component i in V_j
δX = Newton correction to variable X
ϕ_{ij} = residual of equilibrium equation (component i, tray j)

LITERATURE CITED

Buzzi Ferraris, G., "Letter to Editor," *AIChE. J.* **26**, 703 (1980).

Hofeling, B. S. and J. D. Seader, "A Modified Naphtali-Sandholm Method for General Systems of Interlinked, Multistaged Separators," *AIChE J.*, **24**, 1131 (1978).

Ketchum, R. G., "Strategy for the Computation of Interlinked Separation Columns for Non-Ideal Mixtures," *Proc. of the Symp. on Computers in the Design and Erection of Chemical Plants*, Karlovy Vary (Aug. 31-Sept. 4, 1975).

Kubicek, M., V. Hlavàcek, and F. Prochàska, "Global Modular Newton-Raphson Technique for Simulation of Interconnected Plant applied to Complex Rectification Columns," *Chem. Eng. Sci.*, **31**, 277 (1976).

Naphtali, L. M., and D. P. Sandholm, "Multicomponent Separation Calculations by Linearization," *AIChE J.*, **17**, 148 (1971).

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A Comment on the Equation of State by Hirschfelder et al.

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We find a flaw in the gas equation of state formulated by Hirschfelder et al. For densities somewhat smaller than the critical density and temperatures in the neighborhood of the critical temperature, $\partial P/\partial \rho$ at constant temperature is negative. This violates the requirement of mechanical stability. The importance of the violation depends on the use being made of the equation of state.

In previous articles, Hirschfelder, Buehler, McGee, and Sutton (1958) presented an analytic equation of state for gases and liquids which was designed to match the corresponding states

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