Linares-Solano, A., O. P. Mahajan, and P. L. Walker, Jr., "Reactivity of Heat-Treated Coals in Steam," *Fuel*, 58, 327 (1979).
Mahajan, O. P., R. Yarzab, and P. L. Walker, Jr., "Unification of Coal-Char Gasification Reaction Mechanisms," *ibid.*, 57, 643 (1978).
Mahajan, O. P., and P. L. Walker, Jr., "Effect of Inorganic Matter Removal from Coals and Chars on their Surface Areas," *ibid.*, 58, 333 (1979).
Petersen, E. E., "Reaction of Porous Solids," *AIChE J.*, 3, 443 (1957).
Simons, G. A., and M. L. Finson, "The Structure of Coal Char: Part I. Pore Branching," *Comb. Sci. Tech.*, 19, 217 (1979).
Simons, G. A., "The Structure of Coal Char: Part II. Pore Combination," *ibid.*, 19, 227 (1979).
Szekely, J., and J. W. Evans, "A Structural Model for Gas-Solid Reactions with a Moving Boundary," *Chem. Eng. Sci.*, 25, 1091 (1970).
Szekely, J., and J. W. Evans, "A Structural Model for Gas-Solid Reactions with a Moving Boundary—II," *ibid.*, 26, 1901 (1971).
Tomita, A., O. P. Mahajan, and P. L. Walker, Jr., "Reactivity of Heat-Treated Coals in Hydrogen," *Fuel*, 56, 137 (1977).

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An Optimal Arrangement of Simultaneous Linearized Equations for General Systems of Interlinked, Multistaged Separators

R. HIDALGO S., A. CORREA V., A. GOMEZ M.
Instituto Mexicano del Petroleo, Mexico, Mexico D. F., Mexico
and
J. D. SEADER
University of Utah, Salt Lake City, Utah 84112

The simultaneous solution of all the equations for a general system of interlinked, multistaged separators is considered. The interlinks may be simple streams or reciprocal streams, and a particular separator may have bypassing streams (for example, pumparounds). Algorithms are developed that automatically arrange the linearized equations so that for a Newton-Raphson type of solution procedure using, for example, the Naphtali-Sandholm technique, a minimum or nearly minimum number of nonzero blocks occur outside the tridiagonal band so that computational memory and time requirements are minimized. The nonzero and nonidentity blocks are stored in a single vector of variable dimension.

SCOPE

In recent years, techniques of designing industrial plants with digital computers, as described by Crowe et al. (1971) and Seader et al. (1977), have been widely applied. Sometimes, such plants consist of complex arrangements of interlinked unit operation modules, each of which can be calculated as an independent unit. Two general approaches can be applied to the solution of the set of equations corresponding to an interlinked system. The first consists of solving the separate equations for each unit sequentially and iteratively and is called the sequential method. In the second procedure, called the simultaneous method, the complete set of equations for the system is solved. The sequential approach is usually adopted for designing processes because the approach offers great modularity; that is, each unit of the plant can be modeled separately. However, the simultaneous approach offers advantages for problems with a high degree of feedback or many recycle streams. Recycle problems are becoming more popular for ecological reasons and because of the energy and raw material crises.

The simultaneous method can involve a large set of nonlinear equations. Therefore, numerical methods of solving these equations must take advantage of the structure of the system in order to minimize computer memory requirements. For example, consider the general separation system of Sargent and Gaminibandara (1975) which performs the separation of n components as shown in Figure 1 (for simplicity, condensers and reboilers are not shown). This structure offers great flexibility, since column sections and streams can be developed by starting with a given processing scheme (analysis of a flow sheet) or can begin by looking for a good structure (synthesis). Any complex separation scheme obtained from the general one is represented by a set of nonlinear equations that can be solved using a Newton-Raphson technique. Such a set of equations exhibits a sparse structure with a number of nonzero block elements outside a tridiagonal band as presented in the case of interlinked distillation column systems by Stupin and Lockhart (1972), Browne, Ishii and Otto (1977), Tedder and Rudd (1978),

A. Correa V. is also at ESFM of Instituto Politécnico Nacional, Mexico City, Mexico.

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Kubiĉek et al. (1976) and Hutchison and Shewchuk (1974).

Hofeling and Seader (1978) have shown that the Naphtali-Sandholm method (1971) for single separators can be extended to retain the linearization and simultaneous solution technique in the case of interlinked separators. However, their method does not offer a systematic approach to the arrangement of the different stages in order to minimize the number of nonzero elements outside of the tridiagonal band, and the modification of the Thomas algorithm still retains all zero elements from the upper triangle of the matrix. Most importantly, the Hofeling-Seader technique is not readily extended to the case where elements outside the main band overlap each other after forward substitution.

This paper considers two types of systems. In the first (case 1), all interlinks between separators are reciprocal streams in that a stream leaving a stage of one separator for a stage of another is matched by a stream of the other phase flowing in the opposite direction between the same two stages. In the second type of system (case 2), interlinks of simple (one-directional) streams as well as reciprocal streams exist. Furthermore, the simple streams can connect two separators or, as with bypasses and pumparounds, two noncontiguous stages in a particular separator. An example of the first type of system is thermally coupled distillation as described by Stupin and Lockhart (1972); the second type of system is typified by a crude unit with side-cut strippers and pumparounds.

CONCLUSIONS AND SIGNIFICANCE

In this paper, techniques to automatically partition the separators into units and arrange these units in order to minimize the number of nonzero elements after forward substitution are presented. The resulting equations are solved using a modified Gauss type of technique in which only the nonzero elements are stored in a linear vector V which is used in place of the whole matrix. Application of the techniques can drastically reduce the search space for the optimal arrangement of units in the Jacobian matrix.

THE GENERAL PROBLEM

Let $\bar{F}\{\bar{x}\}$ be a vector of functions (for example, energy balances, material balances and phase equilibrium relations) and $\bar{x}$ a vector of variables (for example, temperature and component flow rates) such that

$$\bar{F} = [\bar{F}_1, \bar{F}_2, \bar{F}_3, \ldots, \bar{F}_N]^T \tag{1}$$

and

$$\bar{x} = [\bar{x}_1, \bar{x}_2, \bar{x}_3, \ldots, \bar{x}_N]^T \tag{2}$$

The vector $\bar{x}$ that satisfies the system of equations

$$\bar{F}\{\bar{x}\} = 0 \tag{3}$$

can be solved by the Newton-Raphson method using

$$\left(\frac{\overline{\overline{dF}}}{dx}\right)^{(m)} \overline{\Delta x}^{(m+1)} = -\bar{F}\{\bar{x}^{(m)}\} \tag{4}$$

where $\overline{\Delta x}$ is the correction vector, and $(\overline{\overline{dF}}/dx)$ is the Jacobian matrix.

For an ordinary distillation column, where each stage interacts directly only with its immediate neighboring stages, the Jacobian matrix is a blocked, banded matrix (block tridiagonal) and for this case $\overline{\Delta x}^{(m+1)}$ can be computed using the Thomas algorithm. However, if, as with interlinked separators, a stream exists that connects stage k to stage i, where k and i are not neighbors, then

$$\bar{F}_i = \bar{F}_i[\bar{x}_{i-1}, \bar{x}_i, \bar{x}_{i+1}, \bar{x}_k] \tag{5}$$

and the resulting Jacobian matrix will have a nonzero (disperse) block element at a position (i, k) outside of the tridiagonal band. If $i > k$, stage k is located before stage i, and the disperse block element will be below the tridiagonal band. If $i < k$, the opposite situation will exist. A general model for such an equilibrium stage, i, is presented in Figure 2.

In order to compute $\overline{\Delta x}^{(m+1)}$ for the case of disperse block elements, a Gauss elimination technique is used as described later. Each disperse element above the tridiagonal band generates a certain number of nonzero block elements, all of which are located between the disperse block element and the tridiagonal band as shown in Figure 3.

In this paper, we first analyze a multiseparator, interlinked system having only reciprocal streams. We search for the optimal arrangement of the functions in order to obtain a Jacobian with a minimum number of nonzero elements after Gauss elimination. We then treat the more general case where both reciprocal and simple streams occur. A variable-dimension vector V that contains only the nonzero blocks, excluding the identity submatrices from the Jacobian, is used.

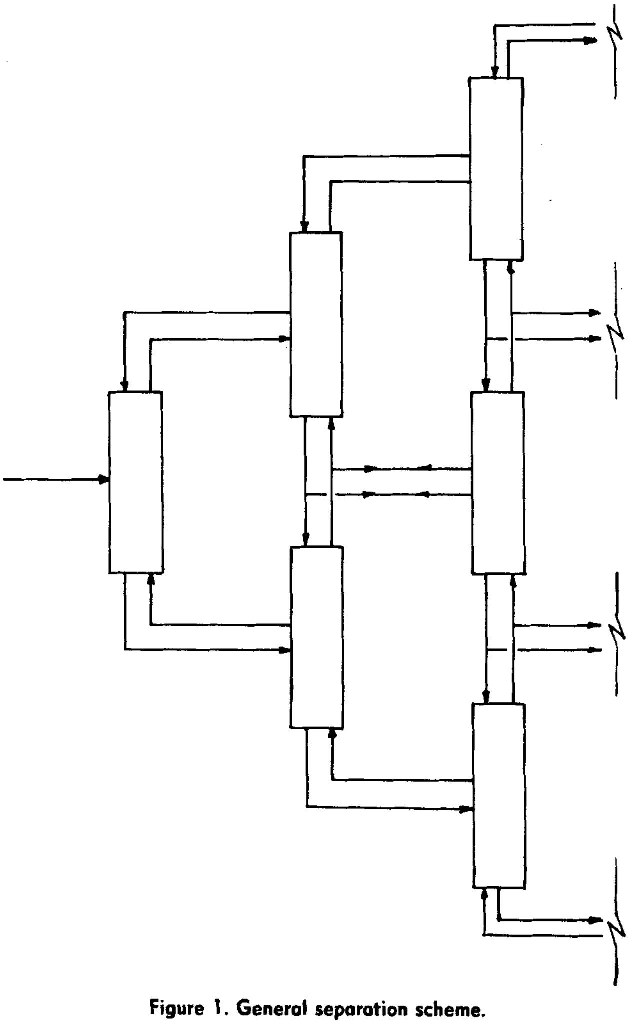

Figure 1. General separation scheme.

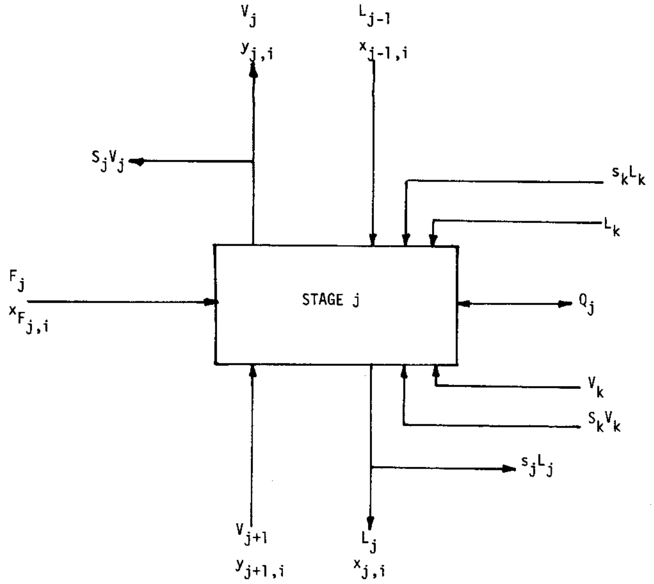

Figure 2. General model for an equilibrium stage.

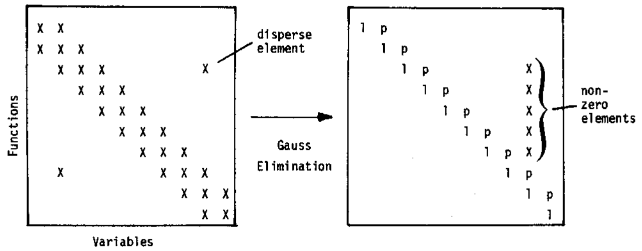

Figure 3. Nonzero disperse elements before and after Gauss elimination.

ARRANGEMENT OF FUNCTIONS FOR CASE 1

To explain the technique, when only reciprocal streams exist, the interlinked system shown in Figure 4*a* is first considered, where each of the three separators is multistaged, and, for simplicity, feed, products, side streams and energy exchangers are not shown. This system can be partitioned as follows. Each separator having interconnecting streams at an intermediate stage location is torn into units at the vicinity of the stage at which the interconnection occurs in such a way that when following the interconnecting liquid flow, the connections will be from the bottom of a unit to the top of a unit, rather than from top-to-top or bottom-to-bottom. Thus, as shown in Figure 4, separator I, which has no intermediate interconnections, is not torn but is renamed unit A. Separator II is torn between stages M-1 and M into two units B and C, so that liquid connecting separators I and II flows from stage Q at the bottom of unit A to stage M at the top of unit C. Similarly, liquid from stage M-1 at the bottom of unit B flows to stage M at the top of unit C. Separator III is torn into units D and E to achieve the same type of bottom-to-top liquid connection. For simplicity, interconnecting liquid and vapor streams that are shown as separate streams in Figure 4*a* are drawn as single, two-directional streams in Figure 4*b*.

The streams of the partitioned system can be characterized by a bottom-top (B-T) vector of dimension S×2, where S is the number of interconnecting reciprocal streams for the partitioned system. For example, this method of tearing in Figure 4 produces the following bottom-top (B-T) vector:

$$B-T = \begin{bmatrix} A, & C \\ B, & C \\ D, & B \\ D, & E \end{bmatrix} = \begin{bmatrix} \text{stream } 1 \\ \text{stream } 2 \\ \text{stream } 3 \\ \text{stream } 4 \end{bmatrix} \quad (6)$$

where each pair (*X*, *Y*) of B-T represents a reciprocal stream connection between the bottom of unit X and the top of unit Y.

Once the system has been partitioned and a B-T vector is obtained, the equations to represent it can be written unit-by-unit from top stage to bottom stage by following any sequence of units to form the functions-variables (F-V) matrix. Consider, for example, the matrix for sequence DBECA of Figure 4*b* as shown in Figure 5. For this order, the disperse block elements are caused by streams 1, 2 and 4. After the Gauss elimination [or the modified Thomas algorithm proposed by Hofeling and Seader (1978)] is applied to the matrix of Figure 5, the matrix of Figure 6 results. In it, the total number of nonzero block elements in the triangle above the tridiagonal band is B + E +(A+C−2), where the letters stand for the number of stages in the respective units. A schematic representation of the proposed arrangement for the example of Figure 4 is shown in Figure 7. In this diagram, two types of interconnections are evident: those that connect the bottom of a unit to the top of the unit directly below, and the others, called interlinks, that make bridges over stages of one or more units. The number of nonzero disperse block elements above the tridiagonal band in the F-V matrix (Figure 5) is equal to the number of interlinks. The number of nonzero disperse block elements in the F-V matrix after Gauss elimination (Figure 6) is equal to the number of

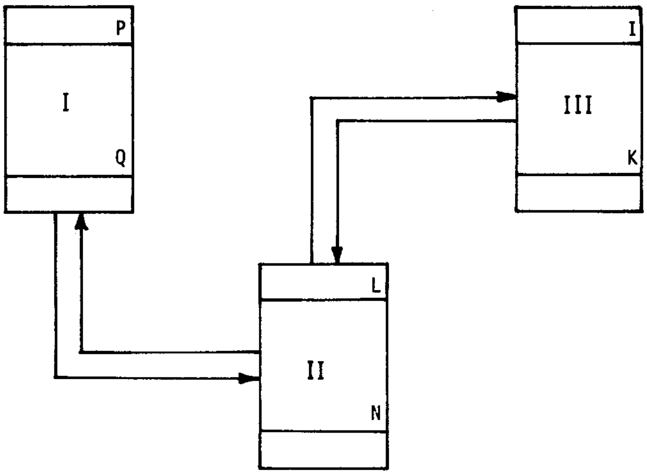

Figure 4*a*. Example of interlinked separators.

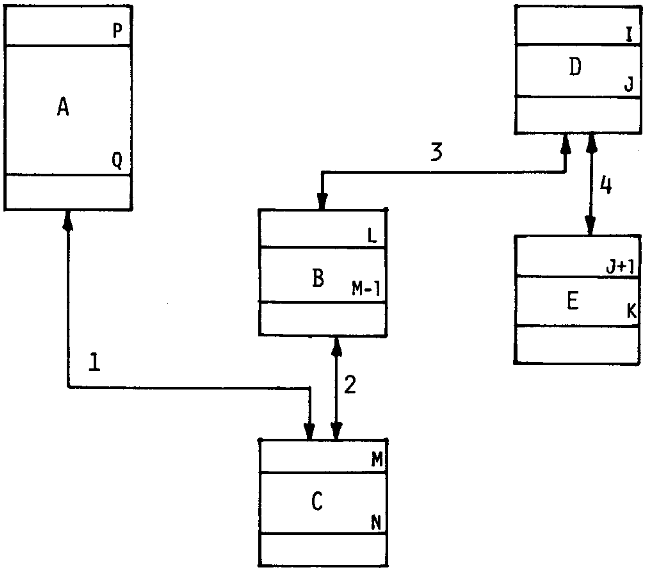

Figure 4*b*. Partitioning of separators into units.

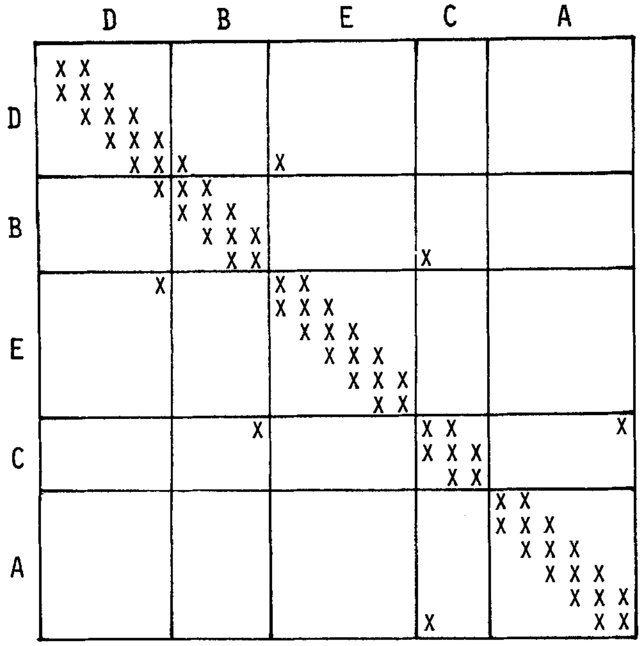

Figure 5. F-V matrix for example of Figure 4*b* with order DBECA.

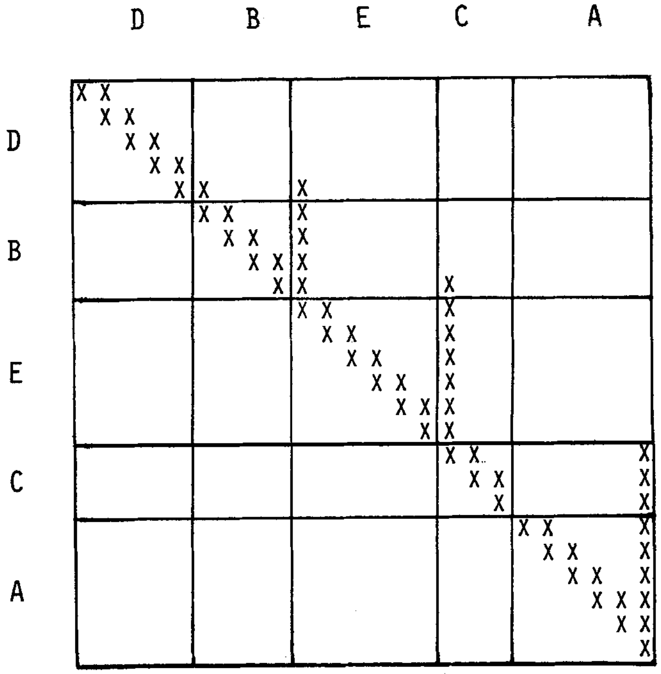

Figure 6. F-V matrix of Figure 5 after Gauss elimination.

stages bridged by the interlinks. In Figure 7, streams 4, 2 and 1 bridge, respectively, B, E and (A+C-2) stages.

Different arrangements of units can produce different numbers of nonzero block elements. For example, consider Figure 8 for the arrangement ADBEC, whose F-V matrix is shown in Figure 9. The B-T vector showing the bottom-top connections is given by Equation (6). For this arrangement, as shown in Figure 9, the nonzero disperse block elements generated by interlinks 1 and 2 are overlapped. That is, they occupy the same column in the F-V matrix. This is always the case of streams that connect one specific stage of a unit (unit C in this case) with two or more units that precede it (units A and B in this case).

All of the interconnecting streams are not necessary in the B-T vector when overlapping occurs. For example, in Figure 8 interconnecting stream 2 is dropped and 1 is retained, because, upon Gauss elimination, the latter will generate all the nonzero disperse block elements generated by the former. The B-T vector in Equation (6) is consequently modified to give:

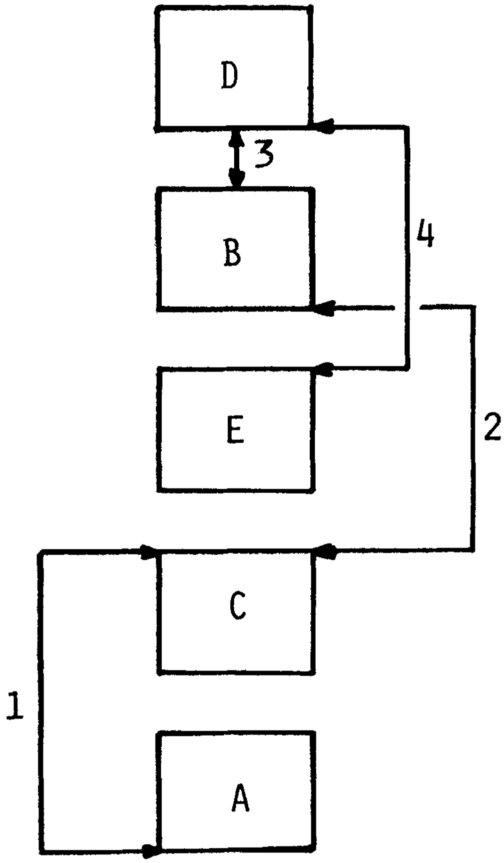

Figure 7. Arrangement DBECA for example of Figure 4*b*.

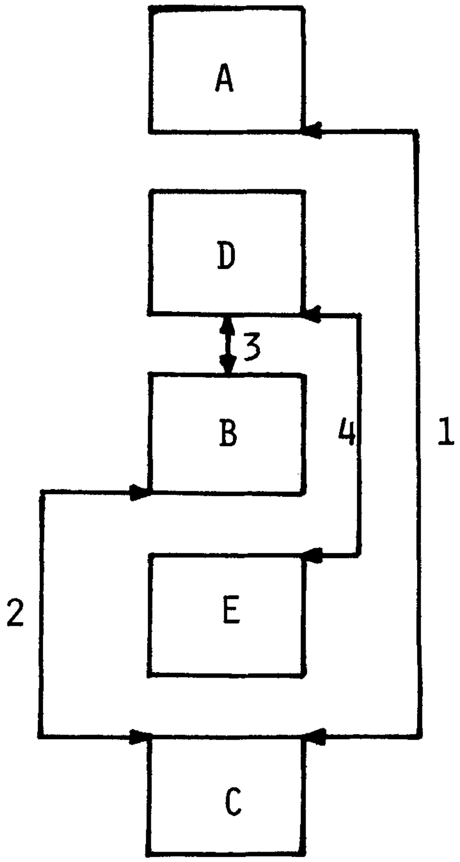

Figure 8. Arrangement ADBEC for example of Figure 4*b*.

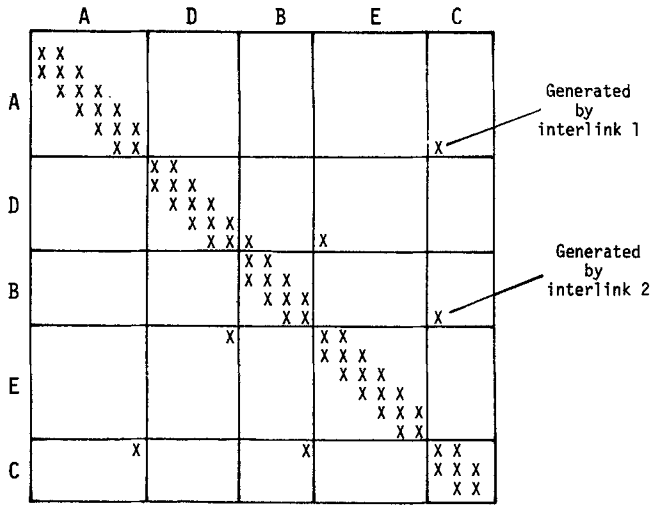

Figure 9. F-V matrix for example of Figure 4b with order ADBEC.

$$B-T = \begin{bmatrix} A, C \\ D, B \\ D, E \end{bmatrix} = \begin{bmatrix} \text{stream 1} \\ \text{stream 3} \\ \text{stream 4} \end{bmatrix}$$

Once the B-T vector has been modified, if we consider only streams that do not overlap disperse block elements after Gauss elimination, the number of nonzero disperse block elements generated can be computed by counting the number of stages bypassed by each stream. This number is directly computed using the B-T vector and remembering that the number of nonzero disperse block elements is equal to the number of bypassed stages. In the general case, the following procedure applies:

Counting rule: If X, Y represents a pair in the modified B-T vector, where unit X is before unit Y in the sequence being considered, and the connection is from the bottom of X to the top of Y, then the number of stages bypassed by the stream connecting X and Y is equal to the number of stages between unit X and unit Y in the sequence. If unit Y precedes unit X, then the number of bypassed stages is equal to the number of stages between unit X and unit Y plus the stages forming units X and Y minus 2.

In applying the counting rule to Figure 8, it is easy to see that pair A, C (where unit A is before unit C) generates D + B + E nonzero disperse block elements. Pair D, B generates none, while pair D, E generates B nonzero disperse block elements. The total number of nonzero disperse block elements generated by sequence ADBEC is then D + 2B + E. This compares to the earlier sequence DBECA, which generated A + B + C + E−2 nonzeroes. Now, if

$$(A + B + C + E-2) < (D + 2B + E)$$

or

$$(A + C-2) < (D + B)$$

then sequence DBECA is preferred over arrangement ADBEC.

In general, for a system torn into N units, N! different sequences can be formed. The optimal sequence is taken to be the one that results in a minimum number of nonzero disperse block elements after Gauss elimination.

Because the search space is large (N!), a computer program was written to perform the search. But even with a very efficient computer program, it is not practical to perform an exhaustive search. It is possible to limit the size of the search space by using the following property:

Property 1: Let X, Y be an element of vector B-T. Then an optimal sequence is found among the sequences in which unit X is before unit Y. The proof is given in the appendix. Optimal arrangements can be generated directly from the B-T vector using the following algorithm and applying property 1.

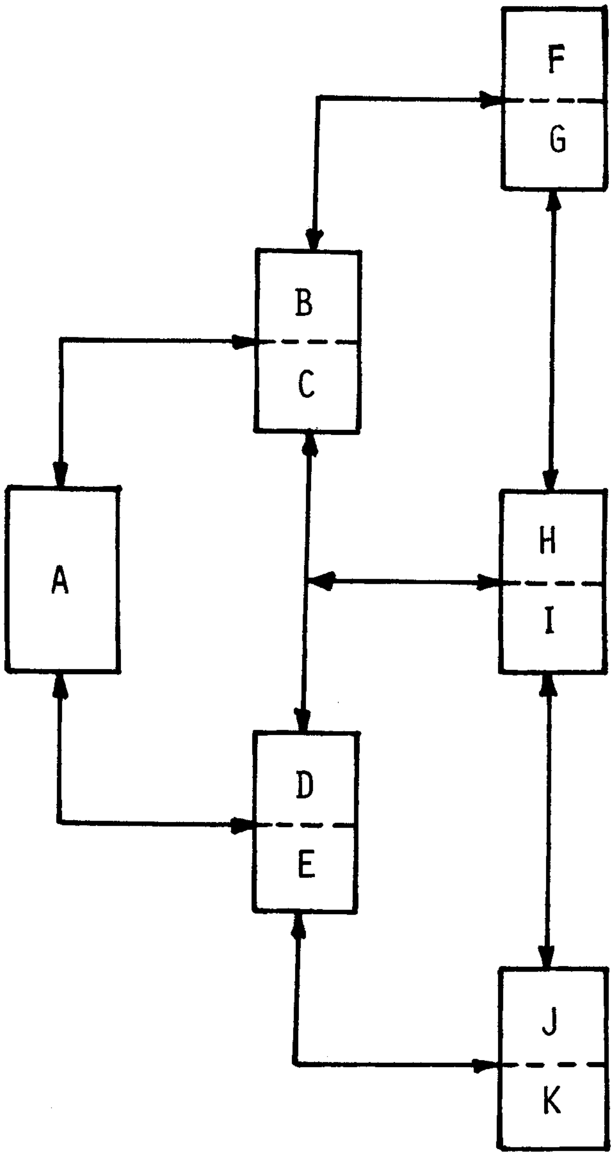

Figure 10. Six-separator example.

Algorithm:

1. Choose all units that appear in the first column of the B-T vector but do not appear in the second column, and name them heads. For example, for Figure 4b, whose B-T vector is given by Equation (6), units A and D are heads.

2. Choose one of the heads to be in the first unit of a family of sequences and eliminate all terms, X, Y from B-T where that unit appears in the B-T vector. If in dropping these terms one contains a unit that does not appear elsewhere in the B-T vector, then that unit should follow the first unit in the sequence. For the example of Figure 4b, if unit A is selected as the head of a sequence, the reduced B-T vector becomes

$$B-T = \begin{bmatrix} \cancel{A, C} \\ B, C \\ D, B \\ D, E \end{bmatrix} = \begin{bmatrix} B, C \\ D, B \\ D, E \end{bmatrix}$$

In dropping the term (A, C), unit C still appears in another term. If unit D is chosen as the head unit, the reduced vector becomes

$$B-T = \begin{bmatrix} A, C \\ B, C \\ \cancel{D, B} \\ \cancel{D, E} \end{bmatrix} = \begin{bmatrix} A, C \\ B, C \end{bmatrix}$$

Because unit E does not appear elsewhere, it is marked as a follower of unit D and must go anywhere in the sequence after unit D. This is indicated by the notation ^{D}E.

3. If the reduced B-T vector is not empty, return to step 1; otherwise exit.

Applying this algorithm to the example of Figure 4*b*, three families are obtained

$AD^{D}EB^{B}C \qquad D^{D}EABC \qquad D^{D}EBAC$

from which eleven sequences are generated:

ADEBC	DEABC	DEBAC
ADBEC	DAEBC	DBEAC
ADBCE	DABEC	DBAEC
	DABCE	DBACE

Thus, the algorithm has selected eleven of 120 (5!) sequences.

Once the favored sequences are obtained, interlinks that generate overlapped nonzero disperse block elements are eliminated in order to compute the correct number of nonzero disperse block elements. This can be done by associating the series of numbers (1), (2), (3), (4), . . .(N) to the sequence being considered. For example, for sequence DABEC

D	A	B	E	C
(1)	(2)	(3)	(4)	(5)

Each number represents the position of the unit in the sequence. The B-T vector can now be written as

$$B-T = \begin{bmatrix} A, & C \\ B, & C \\ D, & B \\ D, & E \end{bmatrix} = \begin{bmatrix} 2, & 5 \\ 3, & 5 \\ 1, & 3 \\ 1, & 4 \end{bmatrix}$$

The following rule is used to eliminate interlinking streams that generate overlapped nonzero disperse block elements:

Elimination rule: search the second column of the B—T matrix for identical elements and mark the rows where they occur. For these marked rows, keep only the one where the element of the first column is the lowest digit. Repeat as often as possible.

For the B−T matrix above, pairs (2,5) and (3,5) are marked. Pair (2,5) is kept because $2 < 3$. The reduced B−T matrix becomes

$$B-T = \begin{bmatrix} 2, & 5 \\ 1, & 3 \\ 1, & 4 \end{bmatrix}$$

No further reduction is possible.

From the final reduced B−T matrix, the minimum number of nonzero disperse block elements is determined by applying the following rule:

Minimum number rule: if (i, j) is a pair in B−T, the number of nonzero disperse block elements generated is obtained by summing the terms in the series from (i+1) to (j−1). For example

interlink (2, 5) generates	(3) + (4)	elements
interlink (1, 3) generates	(2)	elements
interlink (1, 4) generates	(2) + (3)	elements
Total	2(2) +2(3) + (4)	
or	2(A) +2(B) + E	

This analysis is repeated for each favored sequence. The optimal sequence is the one that generates a minimum number of nonzero dispersed block elements.

A computer program was written to implement the rules and algorithm presented above. Several examples were solved and optimal arrangements were found. As an example, the six-separator, eleven-unit scheme, as presented in Figure 10, for which 39 916 800 (11!) possible arrangements exist, was treated. It was found that there exist nine optimal, equivalent sequences out of 291 favored sequences. These optimal sequences are presented in Table 1. For this example, it was interesting to observe that the optimal sequences are independent of the size of the units, and all of them generate G + H + B + 2A + 2C + 2D + I + E + J nonzero disperse block elements outside of the tridiagonal band.

TABLE 1. OPTIMAL SEQUENCES FOR SIX-SEPARATOR EXAMPLE OF FIGURE 10

F	G	H	B	A	C	D	I	J	K	E
F	G	H	B	A	C	D	I	E	J	K
F	G	H	B	A	C	D	E	I	J	K
F	G	H	B	A	C	D	I	J	E	K
F	G	H	B	C	A	D	I	E	J	K
F	G	H	B	C	A	D	E	I	J	K
F	G	H	B	C	D	A	I	J	E	K
F	G	H	B	C	D	A	I	E	J	K
F	G	H	B	C	D	A	E	I	J	K

ARRANGEMENT OF FUNCTIONS FOR CASE 2

In case the system of separators contains reciprocal and simple streams, it is possible to develop a criterion that permits the determination of possible optimal sequences. The strategy is as follows. All sequences in which the simple streams do not generate nonzero disperse block elements are constructed, and those sequences in which the reciprocal streams generate a minimum of nonzero disperse block elements are chosen. This can be done by remembering that a simple stream does not generate such elements when the simple stream goes from a given unit to another unit that follows it in the sequence.

The method can be illustrated with the system of columns from Figure 4 where, however, streams 1 and 3 now become simple streams that only go from A to C and B to D, respectively. The steps are:

1. The system is partitioned as if the simple streams were reciprocal. For the example, the system (as revised) in Figure 4*a* is partitioned to give the system (as revised) in Figure 4*b*.

2. The B-T vector is constructed as if all the streams were reciprocal, but the simple streams are written with a positive sign if they go from bottom to top and with a negative sign otherwise. The reciprocal streams are written without sign. Thus, the B-T vector for the system (as revised) of Figure 4 is modified from Equation (6) to give

$$B-T = \begin{Bmatrix} +A, & C \\ B, & C \\ -D, & B \\ D, & E \end{Bmatrix}$$

3. Those entries having a negative sign are permuted, and the negative sign is replaced by a positive sign. This produces a B−T′ vector. For the example

$$B-T' = \begin{Bmatrix} +A, & C \\ B, & C \\ +B, & D \\ D, & E \end{Bmatrix}$$

TABLE 2. SEQUENCES GENERATED FOR FIGURE 4 (AS REVISED FOR CASE 2)

Sequence					Number of nonzero disperse block elements generated
A	B	C	D	E	0
A	B	D	C	E	C + D
A	B	D	E	C	D + E
B	A	C	D	E	A
B	A	D	C	E	A + C + D
B	A	D	E	C	A + D + E
B	D	E	A	C	A + D + E
B	D	A	E	C	2A + D + E
B	D	A	C	E	2A + D + C

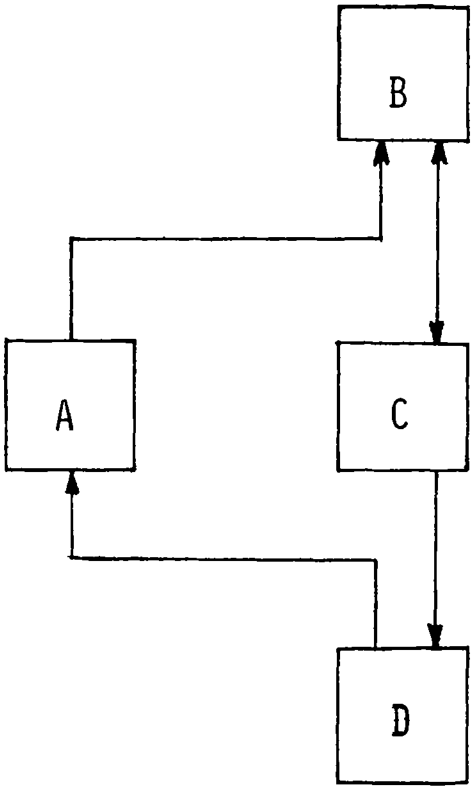

Figure 11. Example of a cycle with simple streams.

4. All the consistent sequences with the B−T′ vector are generated by applying the algorithm for case 1, taking care that only the reciprocal streams (those without sign) generate nonzero disperse block elements. The consistent sequences for the example of Figure 4 (as revised) are given in Table 2, and on the right the number of nonzero blocks generated are written. For this example, an optimal sequence, ABCDE, is obtained.

Unfortunately, this method fails when, during the construction of sequences, no possible head can be found in order to continue the sequence. This happens only when, in the partitioned diagram of the system, the simple upward streams form a branch that closes a cycle. In that event, it is possible to continue the procedure and to develop at least near-optimal sequences by tearing the cycle through any upward stream that closes the cycle.

B−T vector	Sequence
$B-T_1'$	BCDA
$B-T_2'$	ABCD

An example of such a cycle is shown in Figure 11. This cycle can be part of a larger system. The B−T and B−T′ vectors for the cycle of Figure 11 are

$$B-T = \begin{bmatrix} -\ B,\ A \\ B,\ C \\ -\ A,\ D \\ +\ C,\ D \end{bmatrix} \qquad B-T' = \begin{bmatrix} +\ A,\ B \\ B,\ C \\ +\ D,\ A \\ +\ C,\ D \end{bmatrix}$$

It is not possible to generate a family of sequences for the B−T′ vector because the application of the algorithm for case 1 indicates that no head exists. To overcome this difficulty, we tear one of the simple upward directed streams that closes the cycle.

In the example of Figure 11 there are two simple streams that can be torn; the corresponding B−T′ vectors named $B-T_1'$ and $B-T_2'$ are

$$B-T_1' = \begin{bmatrix} -\ B,\ A \\ B,\ C \\ *\ D,\ A \\ +\ C,\ D \end{bmatrix} \qquad B-T_2' = \begin{bmatrix} *\ A,\ B \\ B,\ C \\ -\ A,\ D \\ +\ C,\ D \end{bmatrix}$$

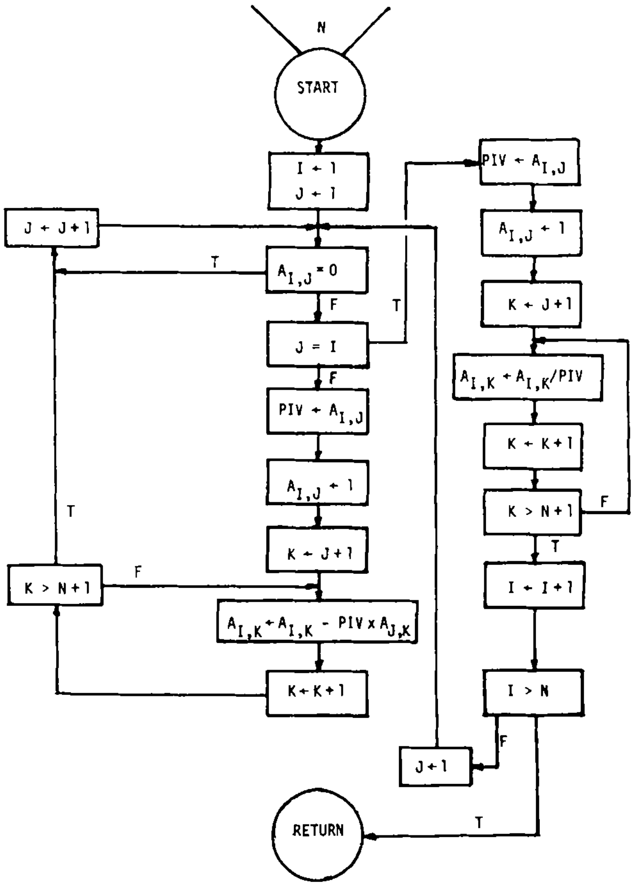

Figure 12. Algorithm for Gauss elimination technique.

where the tearing is indicated by *D, A in $B-T_1'$ and by *A, B in $B-T_1'$.

In the consistent sequences developed from these vectors, construction of the F−V matrices shows that the negative streams will always generate nonzero disperse block elements as if the negative streams were reciprocal streams. In that case, to each vector will correspond a sequence. The results for the example, using this modification of the counting rule for case 1, are

Number of nonzero disperse block elements generated

C + D

B + C

Because the torn stream does not generate nonzero disperse block elements, it is desirable to tear the cycle through that stream that involves units having the largest number of stages. However, when the cycle forms part of a larger system, it is preferable to tear the cycle in such a way that the head of the cycle is the unit involved in one of the eliminated streams of the B−T′ vector.

V–VECTOR AND GAUSSIAN ELIMINATION

When an optimal or near-optimal arrangement of units has been found, a simultaneous solution of $\bar{F}\{\bar{x}\} = 0$ is performed in a manner similar to Naphtali and Sandholm (1971) using a modified Gauss type of method. A flow diagram of the technique is presented in Figure 12. The linear system to be solved is Equation (4), given here as $J^{(m)}[\overline{\Delta x}^{(m+1)}] = -\bar{F}[\bar{x}^{(m)}]$. As shown in Figure 13, a matrix A′ is constructed using matrix J, and including as its last column vector F, so that A′ is of order $M \times (M+1)$. Using a Gauss elimination technique, matrix A′ can be transformed into a matrix A as shown in Figure 14 for the case of no nonzero disperse block elements and in Figure 15 for the case of nonzero disperse block elements.

To perform the elimination, it is not necessary to work with the full matrix; rather the transformation can be carried out by

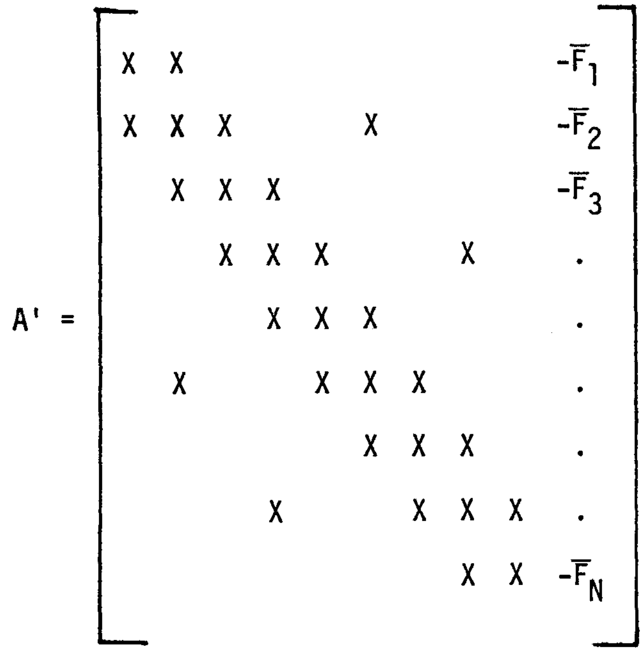

Figure 13. Matrix A′.

working with two vectors of dimension (M+1) that are used as scratch vectors, and a vector V where all important information is stored. The order of storage is arbitrary, but the order used in this work is shown in Figure 15.

If p represents the number of disperse elements that generate nonzeroes (not overlapped), then

$$\text{dimension of vector V} = 2M - (p+1) + \sum_{k=1}^{p} (y_k - X_k) \quad (7)$$

where y_k and X_k represent the coordinates of the disperse element, which can be obtained from the B−T vector.

Notice in Figure 15 that pointers Z_1 and Z_2 are important because they indicate where the α and β quantities begin. It is

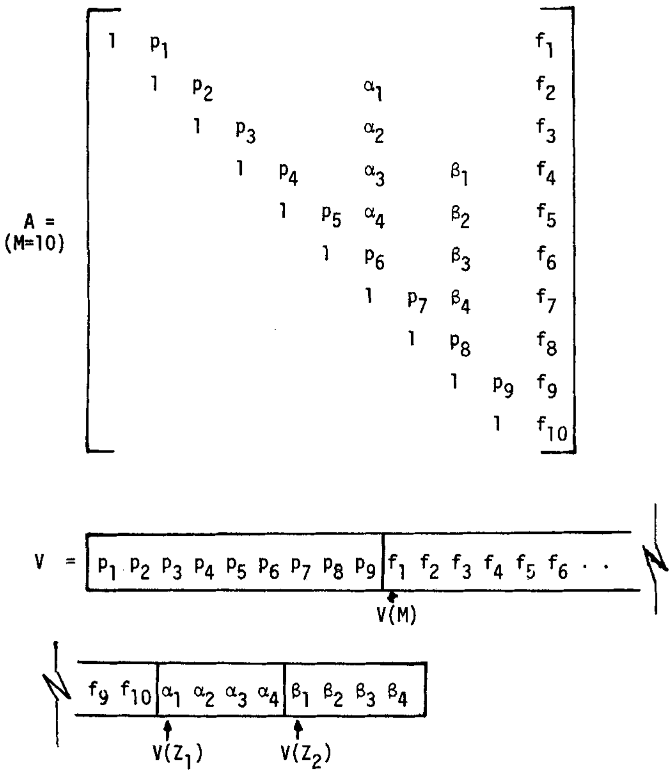

Figure 15. Example of V vector.

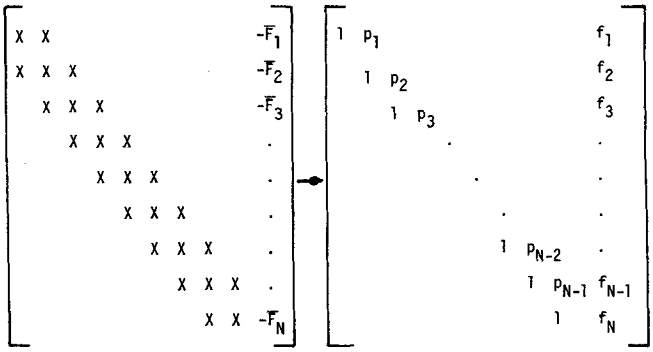

Figure 14. Transformation of matrix A′ to matrix A using Gauss elimination.

possible to create a vector Z which contains all the pointers as follows:

$$Z_1 = 2M$$
$$Z_2 = Z_1 + y_1 - X_1 - 1$$
$$Z_3 = Z_2 + y_2 - X_2 - 1$$
$$\cdot$$
$$\cdot$$
$$\cdot$$
$$Z_p \; Z_{p-1} + y_p - X_p - 1$$

After vector V is constructed, an element A_{ij} can be easily obtained using a procedure called the equivalence algorithm, whose flow diagram is shown in Figure 16. This is equivalent to

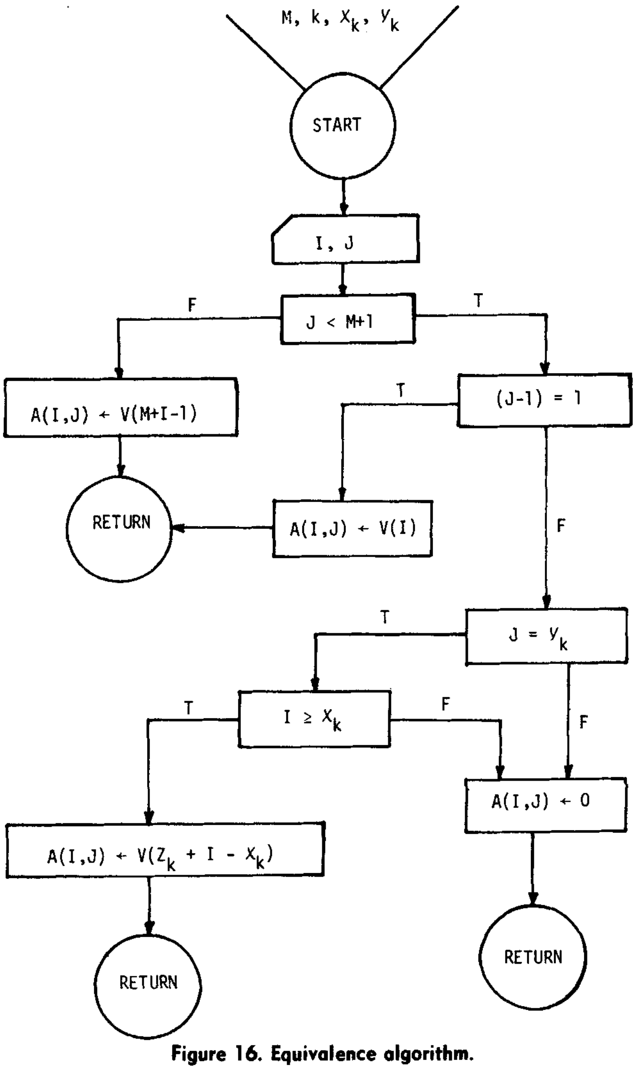

Figure 16. Equivalence algorithm.

using matrix A with two differences: a zero element is never stored, and the number of nonzero elements is a minimum.

This procedure has been successfully applied to the solution of a number of cases. For example with a 50 × 50 matrix containing twenty disperse elements, CPU time is 1.321 s and main memory required is 3 501 words on a Univac 1108. This compares to 1.327 s and 7 060 words of memory for the same example by the method of Kubićek et al. (1976).

APPENDIX: PROOF OF PROPERTY 1

Let X, Y be an element of vector B-T. Consider two arrangements of the vector, where the only difference is a permutation of unit X and unit Y. If the arrangement in which X precedes Y generates n nonzero disperse block elements, then from the counting rule the other arrangement in which Y precedes X must generate $(n + X + Y - 2)$ nonzero disperse block elements, where X and Y also stand for the respective number of stages in units X and Y, respectively. Therefore, the first arrangement will contain less nonzero disperse block elements provided that $(X + Y) > 2$, which will always be the case.

NOTATION

A′ = augmented Jacobian matrix
B–T = bottom-top vector
$\bar{F}$ = vector of functions
f = transformed function
F–V = functions-variables matrix
J = Jacobian matrix
M = dimension of matrix J
P = off-diagonal, nonzero elements
V = stored vector
Δx = correction vector
$\bar{x}$ = vector of variables
X = coordinate of disperse element
X,Y = reciprocal stream connection between bottom of unit X and top of unit Y
y = coordinate of disperse element
Z = pointer
α,β = nonzero disperse elements

Superscripts

m = iteration index
T = transpose

Subscript

i = stage number
p = number of disperse nonzero elements

LITERATURE CITED

Browne, D. W., Y. Ishii, and F. D. Otto, "Solving Multicolumn Equilibrium Stage Operations by Total Linearization," *Can. J. Chem. Eng.*, **55**, 307 (1977).
Crowe, C. M., A. E. Hamielec, T. W. Hoffman, A. I. Johnson, D. R. Woods, and P. T. Shannon, *Chemical Plant Simulation*, Prentice-Hall, Englewood Cliffs, N.J. (1971).
Hofeling, B. S., and J. D. Seader, "A Modified Naphtali-Sandholm Method for General Systems of Interlinked, Multistaged Separators," *AIChE J.*, **24**, 1131 (1978).
Hutchison, H. P., and C. F. Shewchuk, "A Computational Method for Multiple Distillation Towers," *Trans. Inst. Chem. Engrs.*, **52**, 325 (1974).
Kubîcek, M., V. Hlavaĉek, and F. Prochasko, "Global Modular Newton-Raphson Technique for Simulation of an 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).
Sargent, R. W. H., and K. Gaminibandara, "Optimum Design of Plate Distillation Columns," paper presented at IMA Conference on Optimization in Action, University of Bristol (Jan., 1975).
Seader, J. D., W. D. Seider, and A. C. Pauls, *Flowtran Simulation—An Introduction*, 2nd ed., CACHE, Cambridge, Mass. (1977).
Stupin, W. J., and F. J. Lockhart, "Thermally Coupled Distillation—A Case History," *Chem. Eng. Progr.*, **68**, No. 10, 71 (1972).
Tedder, D. W., and D. F. Rudd, "Parametric Studies in Industrial Distillation," *AIChE J.*, **24**, 303 (1978).

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Sulfur Dioxide Transport Through Aqueous Solutions:

Part I. Theory

Expressions are derived for the steady state flux of sulfur dioxide through films of water and neutral and alkaline salt solutions. A local equilibrium approximation yields an analytical expression for the flux through water and neutral salt solutions. To account for deviations from local equilibrium, the conversion from sulfur dioxide (aq) to sulfur containing ions (and vice versa) is assumed frozen near the boundaries of the film ($x = 0, x = 1$). The thickness of the boundary layer near $x = 1$ is pH dependent, indicating an increasing deviation from equilibrium flux in increasingly alkaline solutions. Strong nonequilibrium behavior can be realized under some conditions for a film of concentrated sodium hydroxide operating as a liquid membrane and in conventional scrubbers.

DARYL L. ROBERTS

and

SHELDON K. FRIEDLANDER

Department of Chemical Engineering
California Institute of Technology
Pasadena, California 91106

SCOPE

Transport of sulfur dioxide in aqueous solutions is receiving attention in connection with environmental problems. In the absorption of sulfur dioxide in flue gas desulfurization (FGD) scrubbers and by bodies of water, rain, vegetation and lung mucus, the liquid phase can play an important role (Corbett et al., 1977; Aul et al., 1977; Husar, 1978).

Hetherington (1968), Onda et al. (1971) and Hikita et al. (1977) have derived expressions for the absorption of sulfur dioxide into solutions of sodium hydroxide, sodium bisulfite and sodium sulfite. These expressions are based on penetration theory with the assumption that sulfur dioxide reacts instantaneously and irreversibly with OH^- or $SO_3^=$. Besides assuming

Daryl L. Roberts is with the Shell Development Company, P.O. Box 1380, Houston, Texas 77001. Sheldon K. Friedlander is at the School of Engineering and Applied Science, University of California, Los Angeles, California 90024.

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