Linear Process Calculations as Convergence Accelerators in Flowsheet-Sequenced Programs

Linear process calculations were developed for use in conjunction with flowsheet-sequenced calculations, as methods of convergence acceleration. Two such methods were compared with the successive substitution and Wegstein methods on relatively simple processes. "Linear Process Simulation," which requires user-derived equations, produced the best results.

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SCOPE

Flowsheet simulation and design programs require large amounts of computer time for convergence of process loop calculations. The inability of such sequential unit calculations to readily utilize overall material balances or simultaneous material balances contributes to the need for many iterative loop calculations to determine accurate recycle flows. This study uses linear material balance calculations based on split fractions to estimate flows for the following iterative calculations and thus speed up convergence. The split fractions used are estimated from the previous calculations, whether linear or nonlinear, and thus allow linear methods to estimate any desired flow for succeeding calculations. These calculations are based on the methods of Rosen (1962), Reklaitis et al. (1979), and Henley and Williams (1973) and permit user-derived programs or other simplified methods to accelerate convergence. The objectives of this paper are to compare and evaluate such methods with other general convergence methods.

CONCLUSIONS AND SIGNIFICANCE

Linear methods based on unit split fractions have been used to estimate component flows in process flowsheets. These methods have been shown to be useful in accelerating convergence in flowsheet-based simulation and design programs. One method, termed linear process simulation or LPS, involves user-derived linear process equations to calculate all recycle mixer outputs, and then flowsheet-sequenced unit calculations follow the mixer outputs. The unit calculations are used to obtain split fractions for each unit for use in the user-derived equations in the following iterative calculations. This method compares favorably with other general methods of convergence acceleration. Other methods, which are based on the recycle fraction of the total feed or on the ratio of recycle to fresh feed, and which do not require user-derived equations are also compared. If user-derived equations are not used, the method utilizing the recycle fraction of the total feed with application to newly determined fresh feed rates appears to be the most desirable method for the processes studied. This recycle fraction method appears to be better than the general Wegstein method. The LPS method or an equivalent method using user-derived linear equations with split fractions appears to be the most suitable convergence method for flowsheet-sequenced process simulation or design programs involving recycle loops.

The recycle fraction method as well as other methods compared should be easily programmable into conventional flowsheet-based programs as one method of convergence acceleration. The LPS or an equivalent method can be programmed into flowsheet-sequenced programs to provide better convergence results than the recycle fraction method, but for each system studied the appropriate linear simulation equations must be derived prior to use.

INTRODUCTION

The solution of material balance problems is a very important part of plant design and simulation calculations and is required either prior to or concurrently with equipment, energy requirement, and other calculations. Sequential unit calculations are the basis of most flowsheet-based general purpose programs and such methods are relatively easy to program and initialize

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for a wide variety of processes. Normally a flowsheet sequence does not use overall material balances or simultaneous multiple unit balances, as might be desired to calculate or estimate recycle streams. For this reason, recycle streams must be calculated by repetitive iterations around the recycle loops until the differences between initial and calculated recycle component flows converge significantly close to zero. The computer time required for convergence of such calculations may become very large and the estimation of recycle streams based on the process structure could be of significant value in reducing the number of iterative trials to reach convergence.

If multiunit processes involve only linear equations, these equations can be solved by a variety of methods to calculate any desired stream in the process. Matrix methods and other methods by Nagiev (1957), Rosen (1962), Hutchison (1974), and Reklaitis et al. (1979) can be programmed for computer solution based on the structure of the process flow diagram and the use of estimated or calculated component split fractions, i.e., the fraction of a component in the feed to a unit that leaves the unit in a given output stream. Recycle fractions, which can be calculated from split fractions, were proposed by Rosen as an approach to steady state material balances. Ravicz and Norman (1964) used sequential calculations for nonrecycle parts of a process along with simultaneous solutions for those parts of a process containing recycle streams.

This paper is concerned with possible methods of using split fractions in linear material balance calculations in conjunction with sequential unit calculations, either linear or nonlinear. The split fractions can be calculated from each unit calculation and can then be used to estimate component flows for the next iteration on the process loop.

LINEAR PROCESS SIMULATION (LPS)

This is a sequential process simulation method developed by Milani (1983) utilizing linearization by split fractions (Nagiev, 1957; Rosen, 1962), the linear method of solution of mixer output streams used by Reklaitis et al. (1979), and linear feedback techniques for multiple loop systems (Henley and Williams, 1973). Conventional unit programs, linear or nonlinear, are used to calculate flows and also split fractions as the program proceeds from a mixer output through the units in a loop. Using the split fractions for each unit, user-derived equations are included in the program to calculate the component flows for each mixer output involving a recycle stream, assuming split fractions remain constant. This provides the initial estimated data for the next series of process calculations, where units and split fractions are recalculated. This procedure continues until the desired measure of error is sufficiently small.

The user-derived equations are linear equations, one for each mixer output as a function of split fractions and process feed streams. They may be derived by basic algebraic derivation for simple systems. For more complex systems, the method of Reklaitis may be used. This involves deriving a product of unit split fractions in the reverse of the flow direction along each mixer input until a fresh feed or a mixer output is reached. For example, for component A in Figure 1, the following equations are derived:

$$A_3 = A_1 + F_{A8}F_{A6}F_{A5}A_4 \tag{1}$$

where

$$F_{A8} = A_8/A_6, \qquad F_{A6} = A_6/A_5 \quad \text{and} \quad FA_5 = A_5/A_4$$
$$A_4 = A_2 + A_3 \tag{2}$$

These split fractions, F_{in}, should be less than 1. The total number of equations for each component is equal to the number of mixers. These can be solved simultaneously for all the mixer outputs.

Another method useful for multiloop processes is based on the loop reduction techniques commonly used for feedback loops. For the example in Figure 2, the following equation can be derived for the loop of streams 2–3–4–6:

$$\text{Since } A_6 = F_{A6}\,[A_7 + F_{A3}\,(A_1 + A_6)], \qquad A_6 = \frac{F_{A6}A_7 + F_{A6}F_{A3}A_1}{1 - F_{A6}F_{A3}} \tag{3}$$

In general for a single loop, in terms of feeds from outside the loop, to solve for A_n within or beyond the loop,

$$A_n = \frac{\sum \text{(Product of Split Fractions of } A \text{ from Feed } K \text{ to Stream } n)A_K \text{ for Feed Streams } K = 1 \text{ to } n_F}{1 - \text{Product of Split Fractions of } A \text{ around loop}} \tag{4}$$

If no recycles enter between streams k and n, then A_n = (Product of all split fractions of A between k and n) A_k, or the summation of such terms if more than one such streams are mixed. Since the split fractions for mixers are 1, in Eq. 3 F_{A6} is the only fraction between stream 7 and 6, and F_{A6} and F_{A3} are the only fractions between 1 and 6, and also the only fractions around the loop. Note that A_7 is a feed stream for the loop considered, 2–3–4–6.

For mixer outputs in Figure 2, Eq. 4 provides the following two equations:

$$A_4 = \frac{A_3}{1 - F_{A5}F_{A7}} = \frac{F_{A3}A_2}{1 - F_{A5}F_{A7}} \tag{5}$$

$$A_2 = \frac{A_1 + F_{A6}A_7}{1 - F_{A3}F_{A6}} = \frac{A_1 + F_{A6}F_{A7}F_{A5}A_4}{1 - F_{A3}F_{A6}} \tag{6}$$

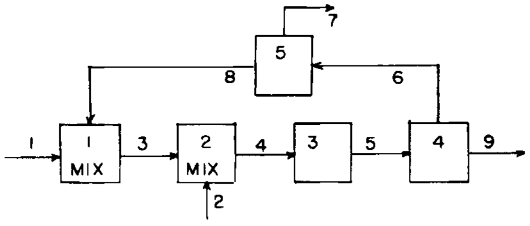

Figure 1. Example process.

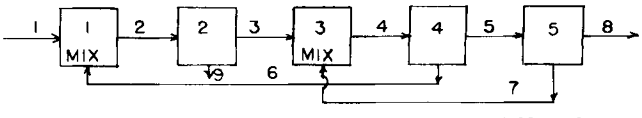

Figure 2. Example process: Process II if units 2, 4, and 5 are flash units; $N_7/N_8 = R$, $N_9/N_3 = 1/(1 + R)$, $N_9 = N_8 = 0.5\,N_1$.

These equations can be solved simultaneously to give

$$A_4 = \frac{F_{A3}A_1}{1 - F_{A5}F_{A7} - F_{A3}F_{A6}} \tag{7}$$

and

$$A_2 = \frac{A_1(1 - F_{A5}F_{A7})}{1 - F_{A5}F_{A7} - F_{A3}F_{A6}} \tag{8}$$

Thus both mixer outputs are easily calculated from the fresh feed, stream 1, and the split fractions.

For complex multiloop processes, techniques presented by Henley and Williams (1973) can be used to arrive at linear equations for any desired stream. Based on these methods, a general algorithm for deriving mixer output component flows has been presented by Milani (1983) for use with LPS on complex systems in a straightforward manner.

A computer diagram for LPS is shown in Figure 3. For the first process calculation, the recycle streams can be estimated or set equal to 0. In the remaining iterative process calculations, the mixer outputs are calculated using the split fractions obtained in the previous calculations, the user-derived equations based on process structure, and fresh feeds. The convergence technique is not a general convergence technique, but is based on the specific process structure. As split fractions approach constant values the method should approach convergence with verification on the second or third process iteration. For a design problem with a choice of independent variables, the choice of split fractions as independent (or design) variables would provide additional reduction in computer time for convergence.

An equivalent method, one that might be a more convenient technique to add to an existing flow diagram sequenced program, would be to calculate recycle flows with user-derived equations developed in a manner similar to LPS.

In LPS or other techniques using user-derived equations in terms of split fractions, it is necessary to give special consideration to reactors because the products and nonlimiting reactants are not directly related to the entering amount of the same component. The split fraction for a limiting reactant is expressable as 1 minus the fraction converted, but the products formed are a function of the limiting reactant and conversion, and act as if they were a mixer feed of product components independent of the product components entering the reactor. Similarly, the reduction of nonlimiting reactants is dependent on the conversion of limiting reactant and can be considered as a negative feed at the reactor. If the limiting reactant is calculated first, stoichiometric equations can provide the reduction of nonlimiting reactants and the production of products, which can then be used as if they were feeds (negative or positive) at the reactor. For example, if unit 3 in Figure 1 is a reactor with $aA + bB \rightarrow cC$, and X is the fraction of A reacted, the split fraction of A, $F_{A5} = 1 - X$, while the equivalent of a feed of $X(c/a)A_4$ mols of C and $-X(b/a)A_4$ mols of B should be added at unit 3 (or at unit 2 since the split fractions between the mixer feed and unit 3 are 1).

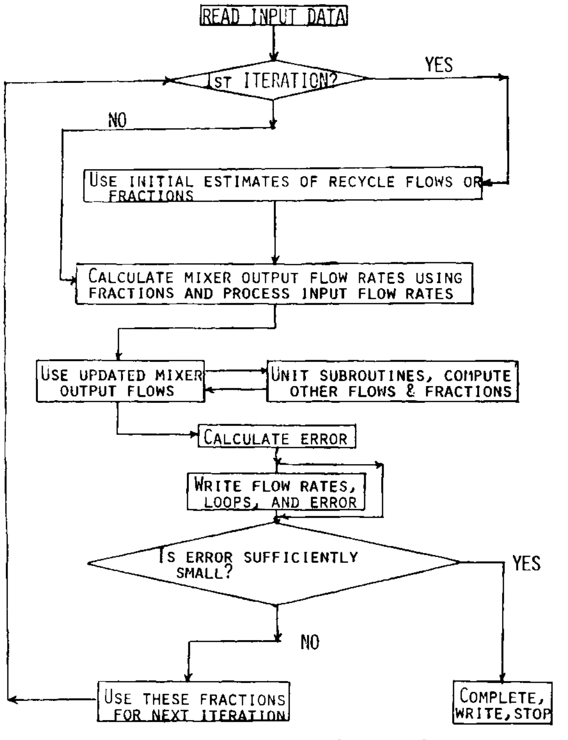

Figure 3. Linear process simulation program structure.

If appropriate algebraic methods are used with linearizing assumptions other than constant split fractions of components, then various alternative types of user-derived equations may be possible.

CONVERGENCE BASED ON RECYCLE FRACTIONS

If all the split fractions in a process were constant, the recycle stream component flows would be a constant fraction of that component flow in the feed. For example for Figure 2, the following equations can be derived for recycle streams 7 and 6.

$$A_6 = \frac{(F_{A6}F_{A3})A_1}{1 - F_{A6}F_{A3} - F_{A7}F_{A5}} \tag{9}$$

$$A_7 = \frac{F_{A3}F_{A5}F_{A7}A_1}{1 - F_{A6}F_{A3} - F_{A7}F_{A5}} \tag{10}$$

The fraction of A_2 that reaches stream 6, for the whole process including the second loop, from Eqs. 9 and 8 is:

$$\frac{A_6}{A_2} = \frac{F_{A6}F_{A3}}{1 - F_{A5}F_{A7}} \tag{11}$$

which could be considered the recycle fraction of A in loop I of the total feed of A in loop I (recycle + fresh feed) considering the whole process, K_{PAI}. Thus

$$K_{PAI}(R_{AI} + F_{DAI}) = R_{AI}$$

and

$$A_6 = R_{AI} = \frac{K_{PAI}}{1 - K_{PAI}} F_{DAI} \tag{12}$$

where

$$F_{DAI} = A_1.$$

Similarly for loop II the fraction of A in the total feed that is recycled is $A_7/A_4 = F_{A5}F_{A7} = K_{PAII}$ and $A_7 = R_{AII} = K_{PAII}F_{DAII}/(1 - K_{PAII})$ where in this case F_{DAII} is A_3.

After a conventional flowsheet sequential calculation iteration for Figure 2, the component flows for the recycle stream and the mixer output, A_6 and A_2, would be available for calculating the recycle fraction $K_{PAI} = A_6/A_2$. Similar data would be available for each component. This might not be quite as accurate an estimate of the recycle flow as could be obtained from user-derived split fraction equations because the value of A_6 might result from unconverged interior or connecting loops, whereas user-derived equations would be the equivalent of all loops being converged with the existing split fractions. However, as all loops tend to converge, the recycle fractions are almost as accurate as could be obtained from split fractions and user-derived equations. The recycle fraction would be easier to obtain than all unit split fractions, and no user-derived equations would be required for this method.

On completion of a complete process iteration, recycle fractions could be calculated for each loop. On the next iteration, whenever a loop was entered at a mixer the recycle stream component flows could be calculated as $F_{DAj} K_{PAj}/(1 - K_{PAj}) = R_{Aj}$ for component A in loop j.

The values for K_{Pij} for each component i in each loop j can be calculated at the end of each process iteration. It is then possible to estimate new values for each recycle stream based on the previous feed to each loop. For constant fresh feed flows this is adequate, but feeds to internal loops will change from one iteration to the next and it is more desirable to apply the recycle fractions for each loop after the feed stream to the loop has been recalculated. This occurs when the feed-recycle mixer is calculated, and these mixer subroutines (in some cases it might be other units with more than one input) can be flagged to be preceded by estimation of recycle streams. Since the main flowsheet programs are complex and vary appreciably, the programming method would vary, but the recycle fraction method should be programmable relatively easily into flowsheet-based programs. As in LPS, it is more desirable and more accurate to calculate the reaction products as if they were feed streams with nonlimiting reactants treated as negative feed streams. An approximate computer diagram is shown in Figure 4.

This method is useful primarily as a convergence method with conventional flowsheet-based design and simulation programs, and in this paper is compared with other convergence methods. Probably the two most common methods of convergence are successive substitution and Wegstein's method (Perry and Chilton 1973). The successive substitution method should work very well if only small changes occur in the recycle when the total feed changes, or $K_{Pij} \rightarrow 0$, or when the recycle is small in comparison to the fresh feed. It should be relatively good under such conditions even when feed rates to internal loops are changing. The Wegstein method is based on the assumption of a linear relationship for each component between the initially assumed recycle and the recycle calculated from the resulting process iteration. With two iterations this linear relationship can be determined for each component and solved for the estimated recycle of each component that will equal the calculated recyle for that component. The resulting recycle is used as an estimate for the next process iteration. In the case of a linear relationship between assumed and calculated recycle, the Wegstein method will provide immediate convergence after the first two iterations. The proposed method based on recycle fraction is based on a more specific linear relationship—i.e., Calculated recycle = K_{Pij} × (Estimated recycle of i + Feed of i)—and if this relationship is exact, both the slope constant of the Wegstein method and the recycle fraction are the same, and both methods will converge very rapidly. In cases of internal loops or reactors with changing feed this might not be the case. With constant feeds the recycle fraction method would be similar but not necessarily the same in rate of convergence as the Wegstein method. However, the recycle fraction method requires only one process iteration to establish K_{Pij} values, whereas the Wegstein method requires two iterations to establish the linear relationship. With a changing feed to an internal loop or reactor, the recycle fraction method provides an allowance immediately if applied to the new feed to a loop, while the Wegstein method would adjust to such changes more slowly.

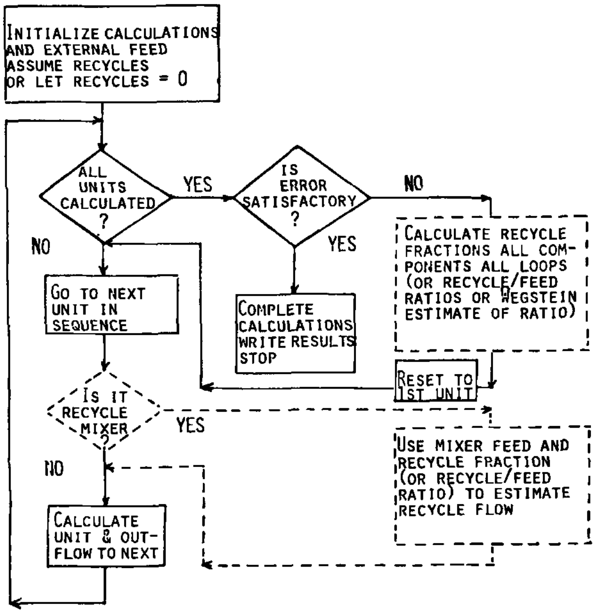

Figure 4. Program for recycle fraction convergence and similar methods. —— Conventional programs, ----- Additional convergence procedures.

The successive substitution method and the Wegstein method as usually described have no response to feed changes to internal loops or to changes in reaction products or nonlimiting reactants. However, such methods can easily be adjusted to compensate for such changes by using the ratio of recycle component to feed component as the variable for either successive substitution or Wegstein acceleration.

The procedure is essentially the same except that the ratio is estimated and multiplied by the feed component to estimate recycle, and the calculated recycle component is divided by the fresh feed component following a process iteration. This calculated ratio is either successively substituted or used in the Wegstein procedure. It should be better to estimate recycle for an internal loop after the new value for internal feed (or hypothetical reactor feed) is calculated. Basing the successive substitution method on the ratio of recycle to feed is equivalent to estimating $K_{Pij}/1 - K_{Pij}$ (for component i and loop j) at the end of a process calculation and applying this quantity to the next iteration. The use of these procedures is indicated in Figure 4. Note that if the recycle fraction, K_{Pij}, is constant, the recycle to fresh feed ratio should also be constant, but changes are not proportional. With reactors treated as previously indicated and only one feed per loop, K_{Pij} should always be less than one.

It should be noted that except for reactors, all of the methods discussed in this paper make no direct allowance for the effect of components other than the component being estimated. These effects can cause the convergence calculations to be so much in error that divergence may occur. The estimation of recycle based on such effects would be more complex, but might be worth further study for serious convergence problems. General methods of considering the effects of components other than the estimated component are the Newton methods used by Cavett (1963).

PROCEDURES

The LPS method was tested by Milani using a FORTRAN Namelist program run on an AMDAHL V8/470 computer. The test runs were based on various systems previously studied and presented in the literature. The program used and the results of these studies have been presented in his dissertation (Milani, 1983).

To study the recycle fraction method, a simple program for two components was prepared in BASIC with subroutines for a mixer, a flash separation for two components, and a two-component reactor. This permitted study of a variety of processes, conditions and procedures without excessive use of computer time. A Commodore Super Pet and an IBM PC were used for these studies. These could be computed in any desired order. The equivalent of ten different convergence routines were included as follows:

A. Based on flows from the previous iteration
   1. Successive substitution
   2. Recycle fraction method
   3. Wegstein method
   4. Wegstein method applied to recycle fraction
   5. Recycle fraction estimated by assuming a linear relationship between recycle fraction calculated and initial recycle component flow

B. Based on feed flows calculated in the new iteration
   6. Successive substitution of recycle/feed ratios
   7. Recycle fraction method
   8. Wegstein method applied to recycle/feed ratios
   9. Same as 4 applied to new feed
   10. Same as 5 applied to new feed

Five different processes were studied and four are shown in Figures 5 to 8. Process II was the same diagram as Figure 2, with units 2, 4, and 5 being flash separators, and is equivalent to three-stage distillation with feed to the reboiler. The reaction used was $2A \rightarrow B$ with the conversion of A as a specified variable, except for process V, where $A \rightarrow 2B$. The flash calculations were based on relative volatility and, in two options, either the liquid to vapor ratio or the vapor composition could be specified. In processes I, II, III, and IV, a relative volatility of 3 was used for K_A/K_B, and in V, K_A/K_B equaled 0.3333. When two or three flash calculations were combined as a distillation system, the liquid and vapor flows were based on a given reflux ratio at the upper flash, partial condensation, equal molar heats of vaporization, and liquid feed at the bubble point. The feed to processes I, III, IV, and V was 0.9 mol fraction of A and 0.1 for B. For process II the feed composition is given in Table 1, as Z_{A1} with $Z_{B1} = 1 - Z_{A1}$.

In most cases calculations were carried out on all units once per iteration and followed by the convergence procedure. In a few cases the calculations were made through all units and then repeated on the final loop for each iteration prior to the convergence procedure.

The iterative calculations continued until all recycle component flows (two times the number of recycle streams) were calculated to be within 0.1% of the initially assumed component flows, or until the number of iterations reached 21, or 31 for some cases with process V. The number of iterations required are presented in Table 1 for comparison.

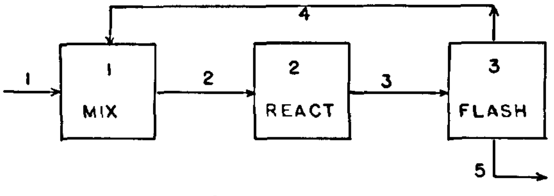

Figure 5. Process I: reactor + flash; reaction $2A \rightarrow B$; $\alpha = K_A/K_B = 3$, $Z_{A1} = 0.9$, $Z_{B1} = 0.1$.

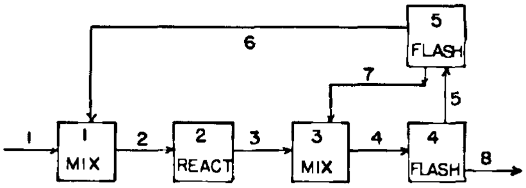

Figure 6. Process III: reactor + two-stage distillation (as two flash units) reaction $2A \rightarrow B$: $N_7/N_6 = R$, $N_8/N_5 = 1/(1 + R)$, $N_8 = N_6 = 0.5\ N_3$, $Z_{A1} = 0.9$, $Z_{B1} = 0.1$.

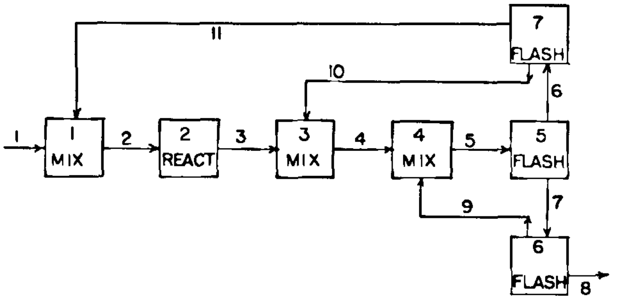

Figure 7. Process IV: reactor + three-stage distillation, reaction $2A \rightarrow B$; $K_A/K_B = 3$, $N_{10}/N_{11} = R$, $N_8/N_9 = 1/(1 + R)$, $N_7/N_6 = (2 + R)/(1 + R)$, $N_8 = N_{11} = 0.5\ N_3$, $Z_{A1} = 0.9$, $Z_{B1} = 0.1$.

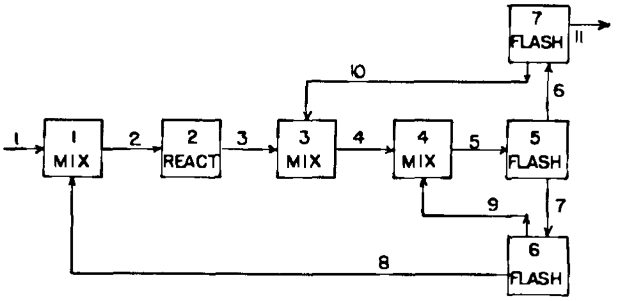

Figure 8. Process V: reactor + three-stage distillation, reaction $A \rightarrow 2B$; $K_A/K_B = 0.3333$, $N_{10}/N_{11} = R$, $N_8/N_9 = 1/(1 + R)$, $N_7/N_6 = (2 + R)/(1 + R)$, $N_8 = N_{11} = 0.5\ N_3$, $Z_{A1} = 0.9$, $Z_{B1} = 0.1$.

In processes IV and V, equations were derived from unit split fractions to estimate all recycle flows for the next iteration. This method is equivalent to the LPS method of Milani. The number of iterations for this procedure is also shown in Table 1.

LPS RESULTS

Four sample problems were solved by the LPS method of Milani. These involved relatively simple recycle problems including mixers, flash and distillation separations, reactors, and splitters. The number of components involved was from three to 16, and the flash and distillation separation calculations were highly nonlinear requiring multiple trial calculations within each loop calculation. The vapor-liquid equilibrium constants were assumed to be functions of temperature and pressure only in order to eliminate physical property subroutine requirements.

TABLE 1. COMPARISON OF CONVERGENCE PROCEDURES

	Convergence Method A. Based on Previous Iteration Feed Streams				
Process	1 Successive Substitution	2 Recycle Fraction	3 Wegstein	4 Wegstein on Recycle Fraction	5 Basis Recycle Fract. = f (recycle)
	Number of iterations to error = 0.001 fraction for all components, all recycles				
I (A) $x = 0.5$, $Z_{A4} = 0.8$	7	7	3	7	4
(B) $x = 0.5$, $Z_{A4} = 0.7$	4	9	3	9	12
(C) $x = 0.4$, $Z_{A4} = 0.8$	2	7	2	7	6
II (A) $Z_{A1} = 0.5$, $R = 3$	>21	>21	>21	>21	>21
(B) $Z_{A1} = 0.3$, $R = 3$	>21	>21	16	>21	>21
(C) $Z_{A1} = 0.3$, $R = 2$	19	>21	12	>21	>21
III (A) $x = 0.6$, $R = 2$	>21	13	19	13	>21
(B) $x = 0.5$, $R = 2$	20	14	17	14	>21
(C) $x = 0.6$, $R = 3$	>21	19	19	19	>21
(D) $x = 0.5$, $R = 3$	>21	20	13	>21	>21
IV $x = 0.5$, $R = 2$	>21	>21	5	>21	>21
V $x = 0.5$, $R = 2$	>31	>31	>31	>21	>21

	B. Based on New Iteration Feed Streams				
Process	6 Succ. Subst. Recycle/FD	7 Recycle Fraction	8 Wegstein Recycle/FD	9 Wegstein Recycle Fract.	10 Basis Recycle Fract. = f (Recycle)
I (A)	7	7	5	7	4
(B)	9	>21	9	>21	14
(C)	7	12	13	12	8
II (A)	18	7	11	7	13
(B)	17	7	16	7	11
(C)	14	7	9	7	15
III (A)	14	7	9	7	14
(B)	14	7	6	7	11
(C)	18	7	11	7	18
(D)	16	7	11	14	12
IV	15	6	>21	6	16
V	18	9	>21	9	>21

	C. Final Loop Repeated Prior to Convergence Procedure Method (see above)				
Process	1	2	3	6	7	8
II (A)	20	>21	13	17	11	6
(B)	18	>21	15	15	11	10
(C)	14	>21	12	11	10	7
III(B)	14	>21	12	9	13	6
V	>31	>31	>31	16	13	20

	D. Equivalent to LPS Method	
Process	Once-Through	Final Loop Repeated
IV	2	4
V	7	7

The first example problem was a flash separation of six components with 50% of the bottoms recycled. This problem has been solved using FLOWTRAN with bounded Wegstein acceleration in each of five iterations (Rosen and Pauls, 1977). Using LPS with constant physical properties, five iterations were required with no other convergence techniques. The LPS results might have required more iterations if K_i values had varied appreciably with composition.

The second example was the Cavett problem shown in Figure 9. This system has 16 components, four flash separations, and three recycle streams. It has been studied thoroughly by Cavett (1963) using various convergence techniques with K_i values independent of concentration. The results using LPS are compared with the various general methods used by Cavett in Figure 10.

Similar results were obtained by using approximate noniterative flash calculations in the first three or four process iterations of LPS (Milani, 1983). This resulted in an approximately 25% computer time saving with approximately the same error after 19 iterations. However, it was also noted that a high degree of accuracy in flash convergence requirements in later process iterations considerably reduced the number of process iterations required for the complete convergence.

This problem has also been solved with CHESS, version 3 (Motard and Lee, 1971), using mode 4 of the DIST subroutine for isothermal phase separation. The same constant K values were used as in the LPS calculations and the Cavett solution, and the converged solution was essentially identical to the LPS solution after 89 iterations by successive substitution.

The third example problem was a modified Cavett problem

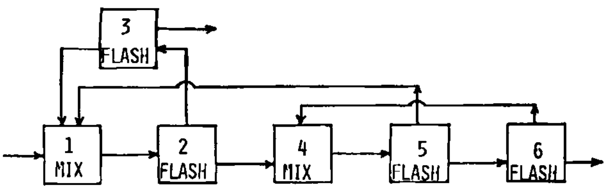

Figure 9. Cavett problem process.

using constant K values and only six components. With LPS 13 iterations were required to obtain the same convergence (0.01 mol/h for each component) as obtained by CHESS with successive substitution of recycle flows in 24 iterations.

A fourth example has the flow diagram of Figure 1, where unit 3 is a reactor, unit 4 is distillation, and unit 5 is a splitter. The following reaction occurs: $C_2H_4 + Cl_2 \rightarrow C_2H_4Cl_2$.

Napthali (1964) solved this problem in approximately 50 iterations using a nonlinear set of equations and the Newton-Raphson method with a Fibonacci search routine (a convergence acceleration method). Using LPS, 40 iterations were required to achieve the same results.

The four example problems using LPS indicate that the use of split fractions from previous nonlinear unit calculations is useful in linear calculations to estimate mixer outputs based on the complete process with all recycles considered. Exact numbers for iterations in moderately complex problems vary widely and are not necessarily good indicators for convergence in other problems. However, both logic and the above results indicate that in general the LPS approach converges more rapidly than successive substitution and about the same as the other convergence methods compared above.

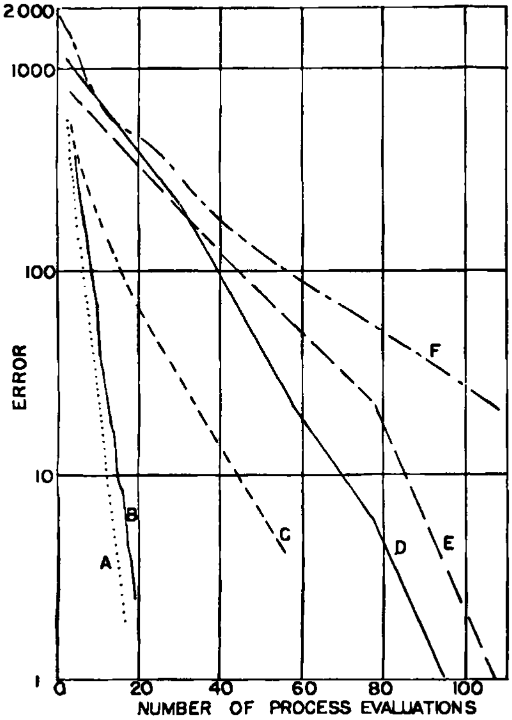

Figure 10. Cavett problem convergence.

A. Newton's method based on one partial derivative for each component (Cavett, 1963)
B. Linear process simulation (Milani, 1983)
C. Successive substitution (Cavett, 1963)
D Newton's Method based on partial derivatives of each component with respect to all nondilute components; two loops (Cavett, 1963)
E. Same as D, three loops (Cavett, 1963)
F. Relaxation method (Cavett, 1963)
Error = $[\Sigma (R_{ij}$ estimated $- R_{ij}$ calculated$)^2]^{1/2}$; i = component, j = loops, feed = 27,340 mols.

The reason that this method converges more rapidly than successive substitution seems to be logically based on the probability that the split fractions of a component in most units are more likely to be constant than are the component flows, when the mixed feed changes. It seems likely that in some special cases where recycle flows are essentially constant, due to pumps or controls, the LPS method might be less effective than methods using successive substitution convergence. Under such conditions it might be feasible to derive a different set of user-derived equations for LPS.

In comparison with other convergence acceleration methods, the LPS method is based upon the process structure and the assumption of constant split fractions. In processes where this assumption is reasonable, the LPS method should be superior to general convergence methods based on constant partial derivatives of overall effects. However, each case is different and there is always the possibility of oscillation and divergence, and it is probably desirable to have available several convergence techniques.

RECYCLE FRACTION RESULTS

Using a simple two-component flow sequence program involving mixers, reactors, and flash systems, various convergence methods were compared by determining the number of iterations required to converge all recycle component flows to an error fraction of 0.001. These results are given in Table 1.

One factor which appears significant is that for all systems except I, it is preferable to base the convergence on feed rates and hypothetical feed rates calculated in the new process iteration rather than the previous iteration (i.e., methods 6 through 10). This did not seem to be true for process I, a simple one-loop reactor-flash system for any of the convergence methods. It had been suspected that there would be an advantage in applying ratios to new values of the hypothetical feed of component B in the reactor. At lower conversions and lower values of recycle concentration, the recycle fraction method in particular oscillated appreciably around the correct value when applied to new feed rates. Successive substitution and the Wegstein method solutions did not tend to oscillate.

The lowest average number of iterations for all the methods tried (not including the LPS equivalent) was the recycle fraction method applied to the newly calculated feed rates. It was also the low number for seven out of the 12 runs calculated. The Wegstein method applied to the recycle fraction (methods 4 and 9) was almost the same as the recycle fraction method itself, because in cases of very low differences the program switched to the recycle fraction method, and this occurred in most runs. In no run did there appear to be any improvement by this method. In methods 5 and 10, there was only one case of an improvement over the recycle fraction method. In five cases out of 12, the Wegstein method gave a lower number of iterations than the recycle fraction method, either applied to the previous feed or to the new feed.

It is of interest to note that method 6, the successive substitution method applied to the ratio of recycle to feed was the only method that always converged in less than 21 iterations, probably due to very little or no tendency to oscillate. This perhaps should be a back-up method for other methods likely to oscillate.

The effect of repeating the final loop unit calculations prior to completing one iteration and prior to applying convergence techniques is also shown in Table 1. The repetition appears to improve successive substitution and the Wegstein procedure, but the results from the recycle fraction method are worse when the final loop calculations are repeated.

For processes IV and V, containing three loops, which were generally the most difficult processses to converge, an equivalent of the LPS method was applied. This involved the use of split fractions for all units to derive linear equations specifically for these processes. Following a complete iteration, all component flows in all recycles were estimated using the derived equations and split fractions from the previous iteration. These flows were utilized in the next iteration. For these two processes this LPS equivalent method was superior to all other methods in convergence. Repetition of the final loop calculations appears to have no advantage in this case, but the results are still superior to the other methods tried for process V.

The LPS method or an equivalent requiring user-derived equations appears to give the best convergence for multiple loop processes. If it is not desired to derive such equations for specific processes, then the recycle fraction method will probably provide close to the minimum number of iterations, although exceptions such as process I are possible. There may be different relationships of the various methods tested when applied to more complex and multicomponent systems, but there appears to be no inherent reason why the advantages of the recycle fraction method should not apply in more complex calculations. In fact, the more intermediate feed streams there are to interior loops, the more advantage should be expected for methods using newly calculated feeds to estimate recycles. The successive substitution and Wegstein methods, applied to the recycle to feed ratio and using newly calculated feed streams, are also improvements over the application of these methods to recycle streams only.

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NOTATION

A_n = mass flow of component A in stream n, mass/unit time
F_{in} = split fraction of component i entering stream n, i.e., the amount of component i in stream n divided by the amount of coponent i in the total inflow to the unit producing stream n
F_{Dij} = fresh feed flow of component i into recycle loop j, mass/unit time
K_i = vapor-liquid equilibrium constant for component i
K_{Pij} = fraction of component i in the total feed to loop j (fresh feed and recycle) that eventually flows back into the recycle to loop j (including the effects of other loops)
n_F = number of feed streams to a single loop
N_i = total molar flow in stream i, mol/unit time
R = reflux ratio in distillation
R_{ij} = recycle mass flow of component i in loop j, mass/unit time
X = conversion of limiting reactant (A) per pass
Z_{in} = mol fraction of component i in stream n
α = relative volatility, or K_A/K_B

LITERATURE CITED

Cavett, R. H., "Application of Numerical Methods to the Convergence of Simulated Processes Involving Recycle Loops," *A.P.I. Proc.*, Div. of Ref., **43**, iii, 57 (1963).

Henley, E. J., and R. A. Williams, *Graph Theory in Modern Engineering*, Academic Press, New York (1973).

Hutchison, H. P., "Plant Simulation by Linear Methods," *Trans. Inst. Chem. Engrs.*, **52**, 287 (1974).

Milani, S. M., "A Linear Process Simulation Method," Ph.D. Diss., Univ. of Missouri-Rolla (1983).

Motard, R. L., and H. M. Lee, *CHESS User's Guide, 3rd. ed.*, Univ. of Houston, Texas (1971).

Nagiev, M. F., "Material Balance in Complex and Multi-Stage Recycle Chemical Processes," *CEP*, **53**(6), 297 (1957).

Napthali, L. M. "Process Heat and Material Balances," *CEP* **60**(9), 70 (1964).

Perry, R. H., and C. H. Chilton, eds., *Chemical Engineers Handbook*, 5th ed., McGraw-Hill, New York, 2–99 (1973).

Ravicz, A. E., and R. L. Norman, "Heat and Mass Balancing on a Digital Computer," *CEP*, **60**(5), 71 (1964).

Reklaitis, G. V., M. K. Sood, and J. M. Woods, "Solution of Material Balances for Flow Sheets Modeled with Elementary Modules," *AIChE J.*, **25**(2), 209 (1979).

Rosen, M. E., "A Machine Computation Method for Performing Material Balances," *CEP* **58**(10), 69 (1962).

Rosen, M. E., and C. A. Pauls, "The FLOWTRAN System," *Computers and Chemical Engineering*, **1**(1), (1977).

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