OPTIMIZATION OF MULTI-STAGE ADSORPTION SYSTEMS

by

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ABSTRACT

Multi-stage adsorption configurations with equality and inequality constraints are optimized and comparisons are made between their relative effectiveness. For a given number of stages countercurrent flow is always superior, as expected. Although crossflow is generally second best, it becomes inferior to certain alternative networks for certain ranges of the magnitude of the adsorption isotherm exponent. Generally, the order of the effectiveness of various networks is believed to be according to how much the networks resemble countercurrent flow.

An algorithm is derived and used to solve the N-stage crossflow network. All other configurations had to be treated as problems in constrained optimization. For this purpose, a theorem of Courant and a method of Carroll for equality and inequality constraints respectively are used to formulate the problem. Three different optimization methods were considered with the deflected gradient method of Fletcher and Powell proving to be the superior for this particular class of optimization problems.

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INTRODUCTION

An earlier study of Lerch (7) investigated the relative effectiveness of various two-stage contacting operations involving adsorption systems. The criterion for effectiveness is based upon the minimum allocation of adsorbent for a fixed quantity of solute-free solvent that is required to reduce the outgoing solute concentration to a given fraction of the initial concentration. Lerch (7) found that irrespective of the adsorption isotherm considered, countercurrent operation was always superior to crossflow operation and that between the two types of crossflow operation the one in which the adsorbent is split into two portions was always superior to the one in which the solution is split into two portions.

The isotherms considered by Lerch (7), the Freundlich and the Koble-Corrigan, have also been used in the present study. The equations of these isotherms are given in APPENDIX VIII. These particular isotherms were employed because they both have relatively simple formulae and they closely approximate isotherms that are observed in practical systems.

The present study extends the work of Lerch (7) to configurations having a higher number of stages. Although Lerch (7) demonstrated the superiority of countercurrent flow to crossflow, it was hoped that the following could be determined:

i. The number of crossflow stages necessary to compete in effectiveness with two-stage countercurrent.

ii. The type of three and four-stage networks that are superior to three and four-stage crossflow respectively, if any.

Since countercurrent flow is always superior to any alternative, the use of other systems must be justified. Treybal (9) justifies the use of networks other than countercurrent only in small scale processing, where there may be an appreciable time lag between stages. The crossflow network may be more practical if the adsorbent deteriorates during inter-stage storage, since fresh adsorbent is employed in every crossflow stage.

In addition to providing an insight into the effectiveness of various adsorption networks, the configurations considered provided a means by which several different methods that optimize systems subject to constraints may be studied. In the course of optimizing networks subject to constraints, it was found that the pattern search technique of Hooke and Jeeves (6) as modified by Weisman, Wood, and Rivlin (10), which had been used earlier with success by Lerch (7) on his more simple configurations, was not adequate for the present class of problems. Therefore, other optimization methods were studied, in particular, the "deflected gradient" and "conjugate direction" techniques of Fletcher and Powell (5) and Powell (8) respectively, and compared with the modified pattern search technique. Since these methods were originally proposed for use on problems not involving constraints it was necessary to modify the objective function to be optimized by the addition of the constraint relations. The equality constraints were treated by the application of a theorem of Courant (3), while the inequality constraints, arising from the necessity of having certain limits on the range of the variables, were treated by a procedure suggested by Carroll (2).

THEORY

A. ADSORPTION NETWORKS.

The adsorption networks to be solved are formulated in the notation of Treybal (9), except that the isotherms are dimensionless. The Freundlich isotherm is used almost entirely since it is very simple and covers all ranges of ease of separability. The Koble-Corrigan isotherm is used only in the general two-stage network given by fig. 1. Typical shapes of these isotherms are shown in fig. 2.

To understand the notation used in the present study, reference should be made to a typical network such as the general N-stage crossflow network in fig. 3. The streams L_s and G_s refer to the quantities of adsorbent and solvent respectively, both streams being on a solute-free basis. The quantities X and Y are dimensionless concentrations of solute in the adsorbent and solvent streams, respectively. The subscripts on these concentrations are derived from the stage number from which their respective streams are leaving. The sole exception to this rule are the initial concentrations, which have two subscripts, these being zero and stream number. It is assumed implicitly throughout the entire study that the solid adsorbent is completely insoluble with the solvent which could, in principle, be either a liquid or a gas.

The general N-stage crossflow network, with split adsorbent stream, is shown in fig. 3. Since this network has been shown by Lerch (7) to be always superior to the crossflow network in which the solution stream is split, the

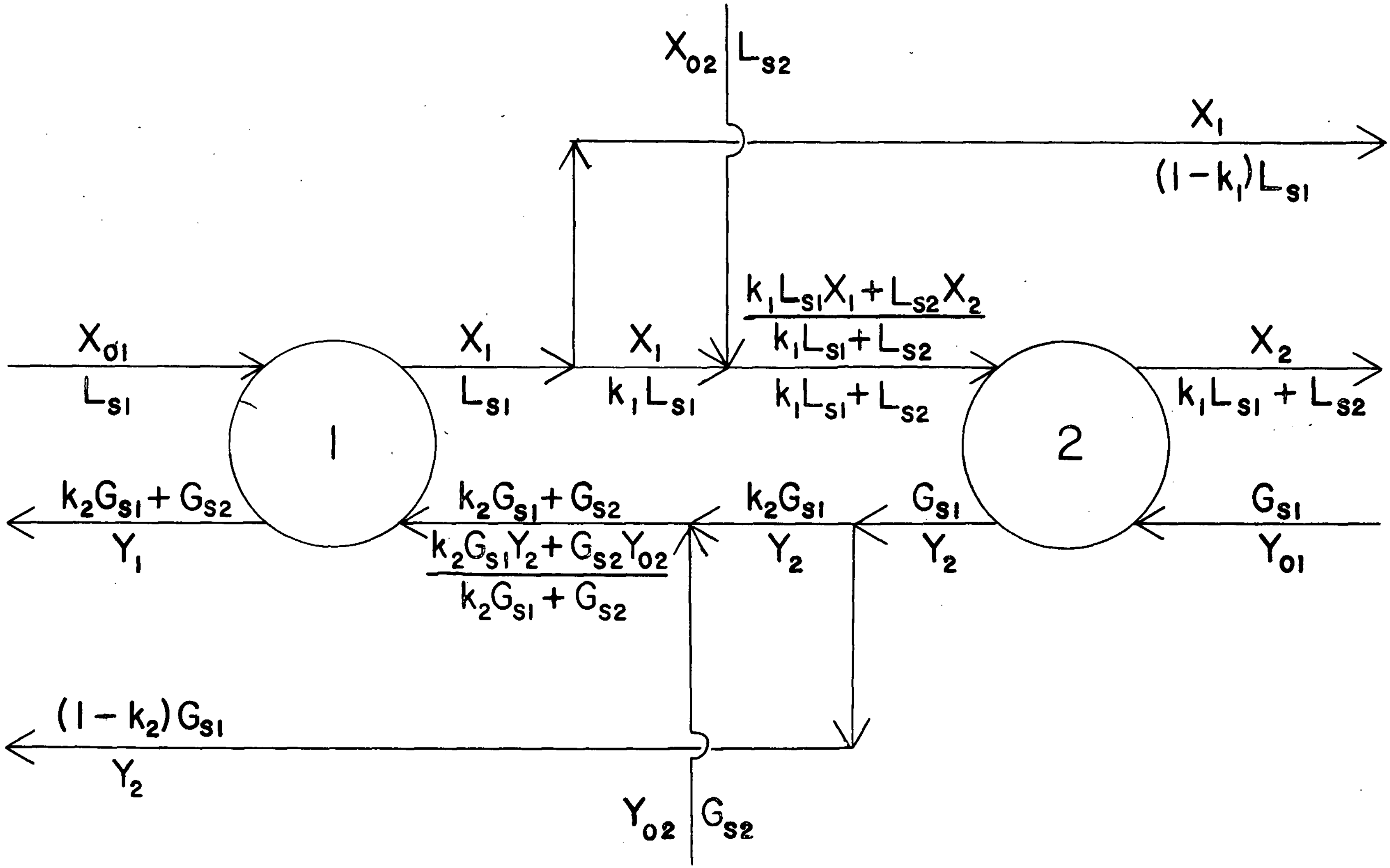

FIGURE 1. GENERAL TWO STAGE SYSTEM

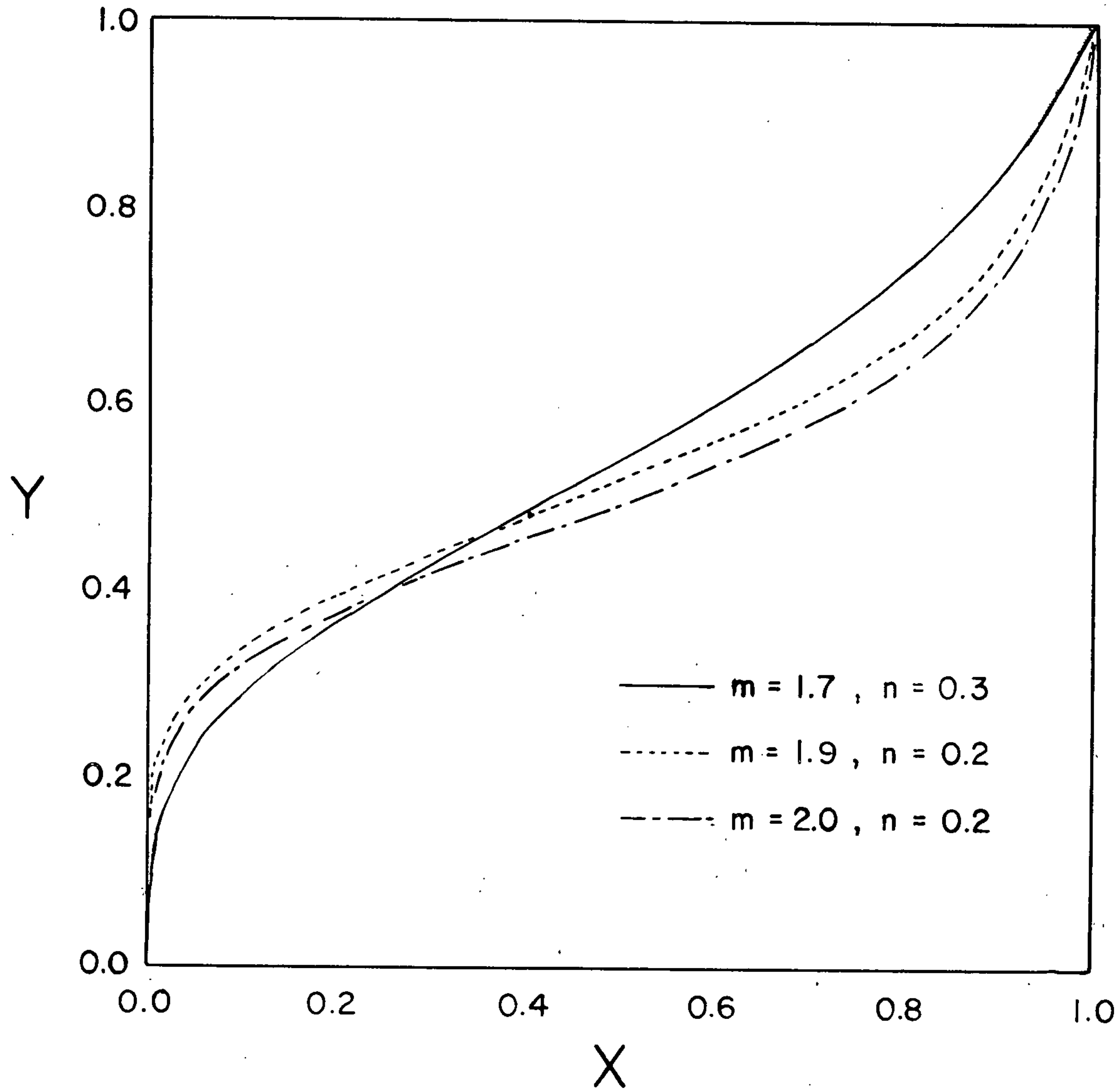

FIGURE 2. KOBLE - CORRIGAN ISOTHERMS USED IN TWO STAGE SYSTEM

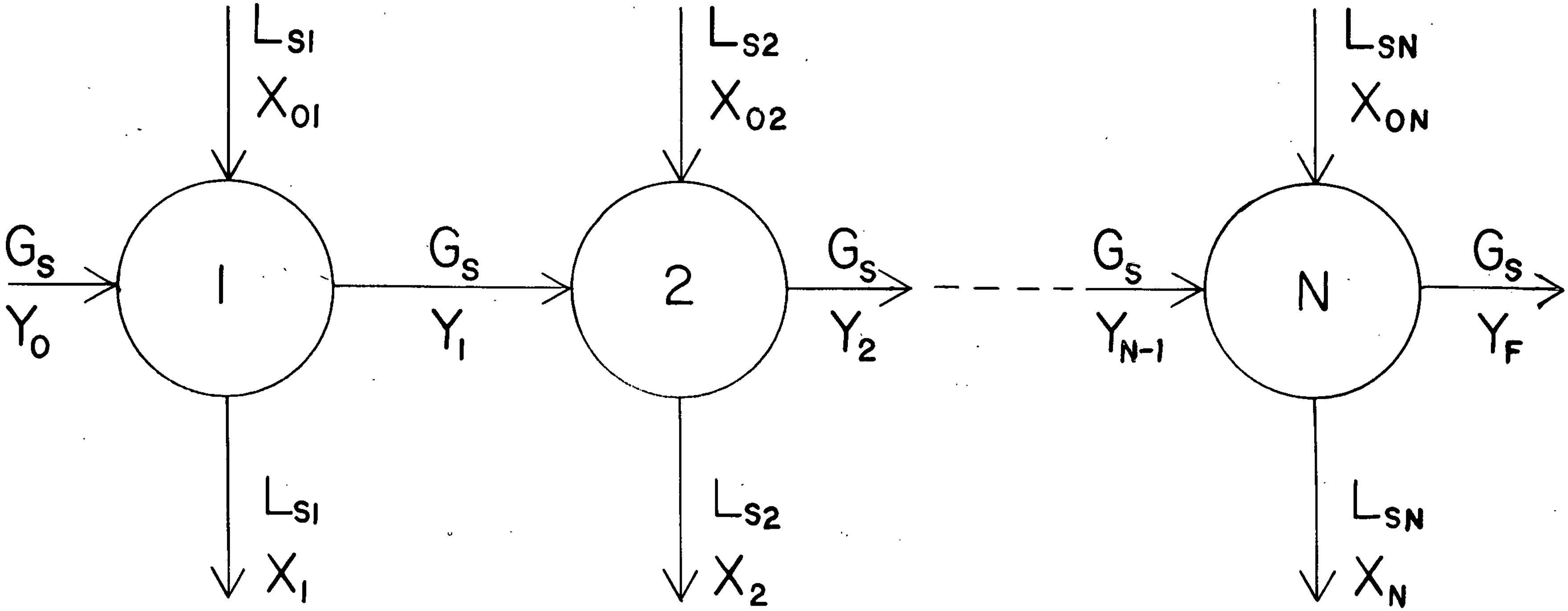

FIGURE 3. N STAGE CROSSFLOW WITH SPLIT ADSORBENT

latter configuration has not been considered. The network shown in fig. 3 is not subject to any constraints; hence an algorithm for its solution can be determined (see APPENDIX VII).

All the other adsorption problems, i.e. the general two stage configuration shown in fig. 1, the N-stage countercurrent network shown in fig. 4 and the various three and four-stage networks shown in figs. 5-9 and 10-14 respectively had to be treated as problems in constrained optimization. Some of the configurations, such as three and four-stage networks of figures 5-14 are subject to the constraint of having to blend two solvent streams to yield the required final concentration. The four-stage networks of figs. 10-14 possess an additional constraint due to the fact that only three of the four individual adsorbent-solvent ratios $(L_{si}/G_{sj}$, $i = 1,2;\ j = 1,2)$ can be fixed independently. The N-stage countercurrent system of fig. 4 is subject to a total of N-1 constraints, these deriving from the fact that the adsorbent-solvent ratio must be the same for each of the N stages. The general two-stage system of fig. 1 requires two constraints, these being due to the blending of the two solvent streams and the necessity that L/G be the same for each stage. These constraints are all of the equality type and can be treated using the method of Courant (3) discussed in section B.

B. FORMULATION OF CONSTRAINT PROCEDURES

1. Method of Courant

Courant (3) has proved a theorem for convergence of a sequence of functions to the optimum subject to an equality

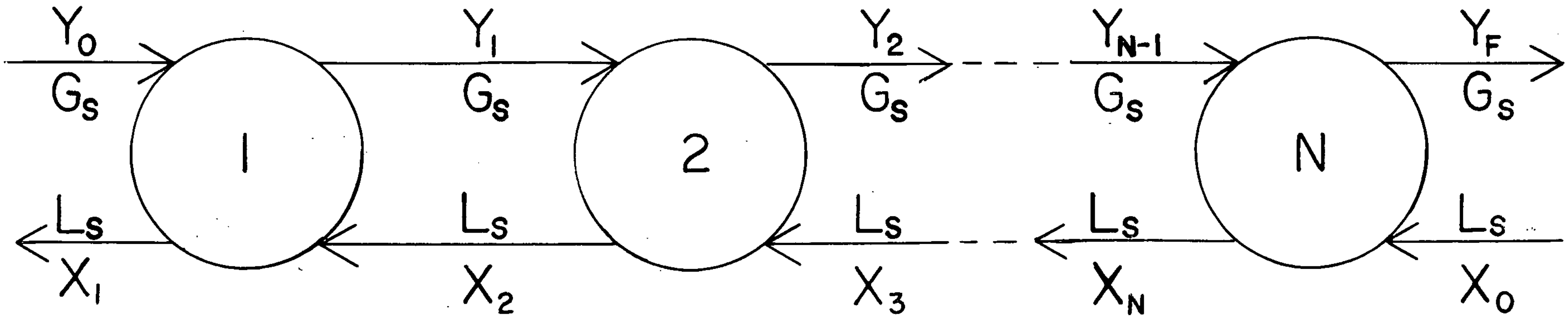

FIGURE 4. N STAGE COUNTERCURRENT

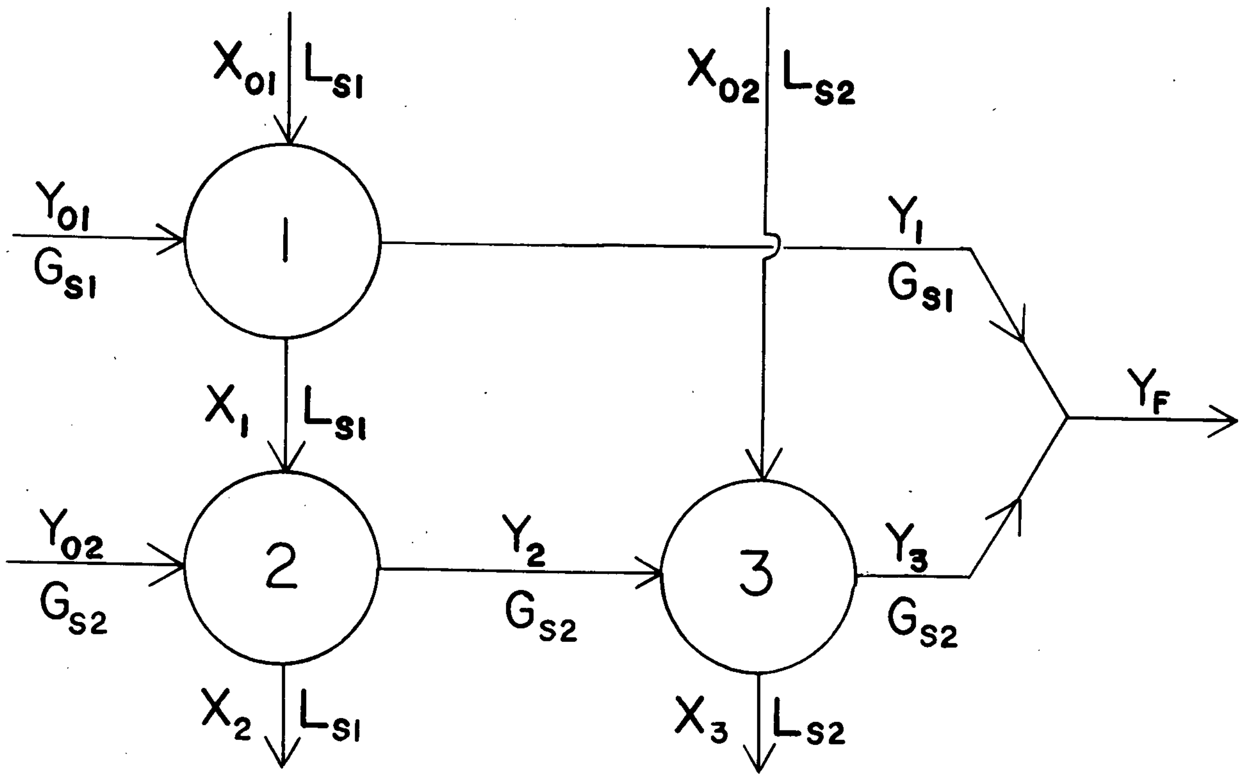

FIGURE 5. THREE STAGE SYSTEM A

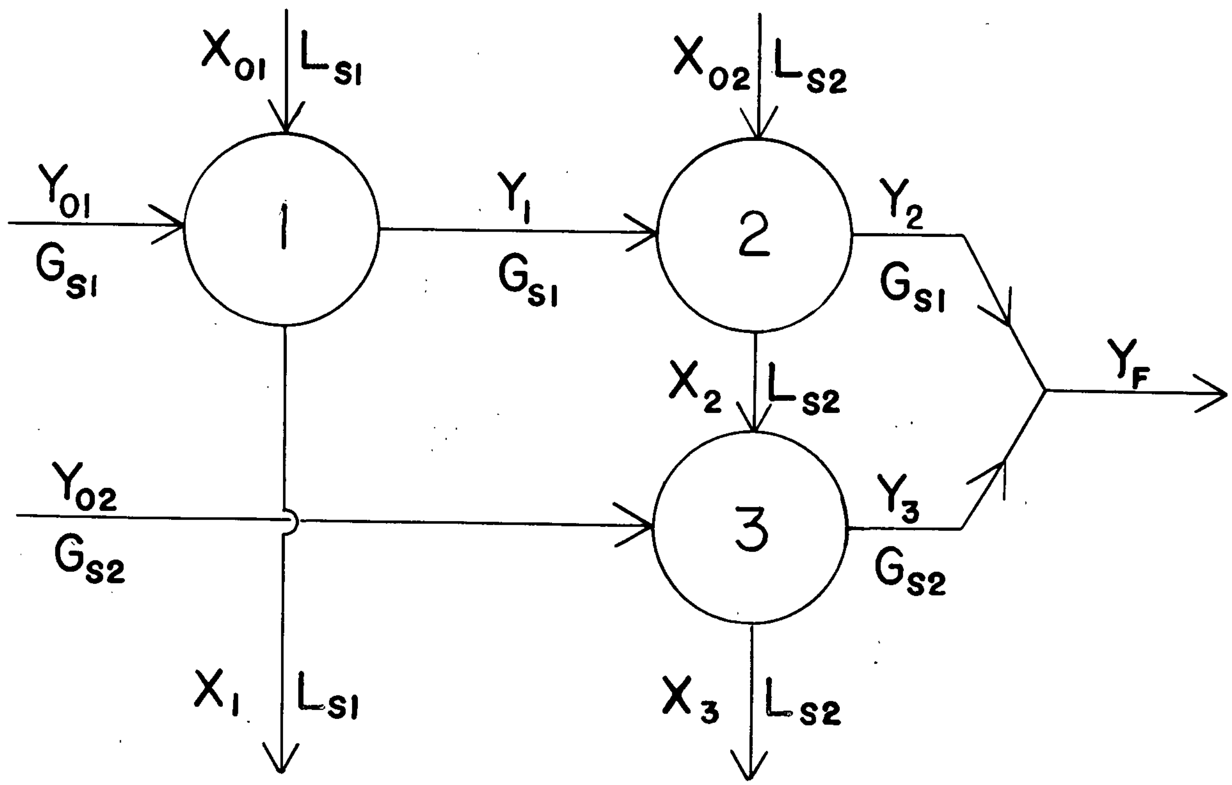

FIGURE 6. THREE STAGE SYSTEM B

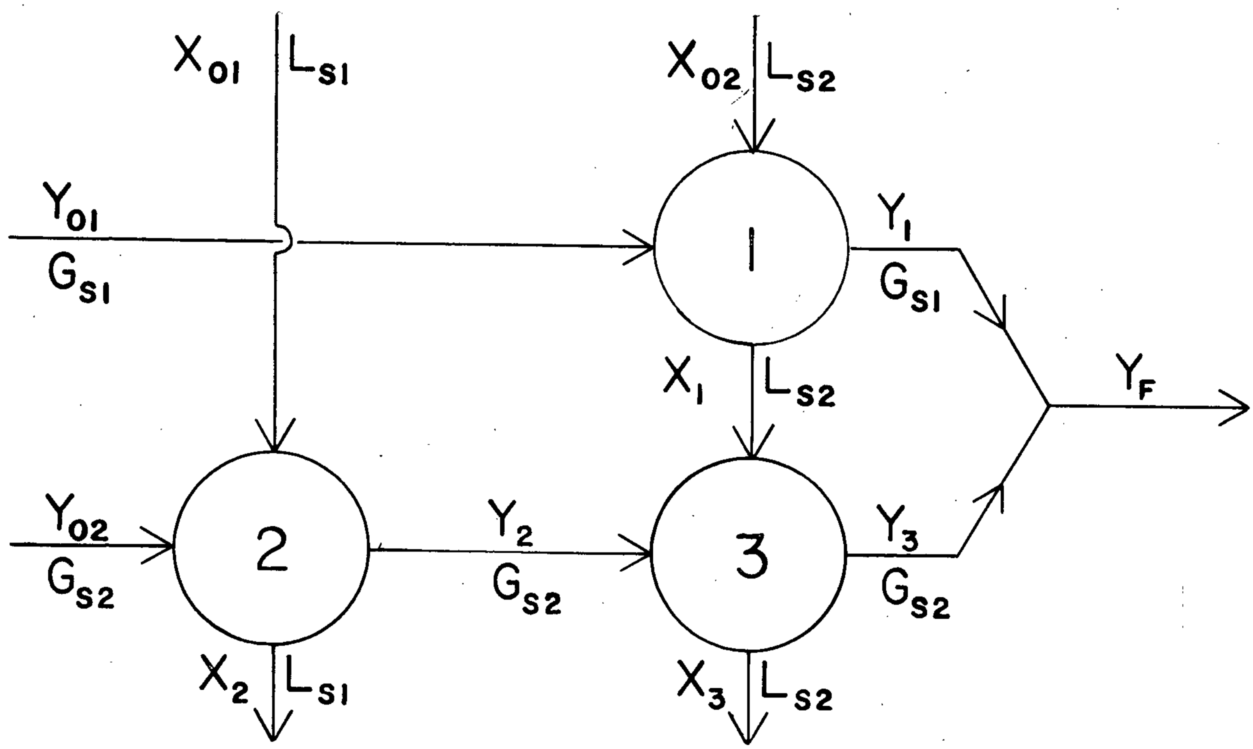

FIGURE 7. THREE STAGE SYSTEM C

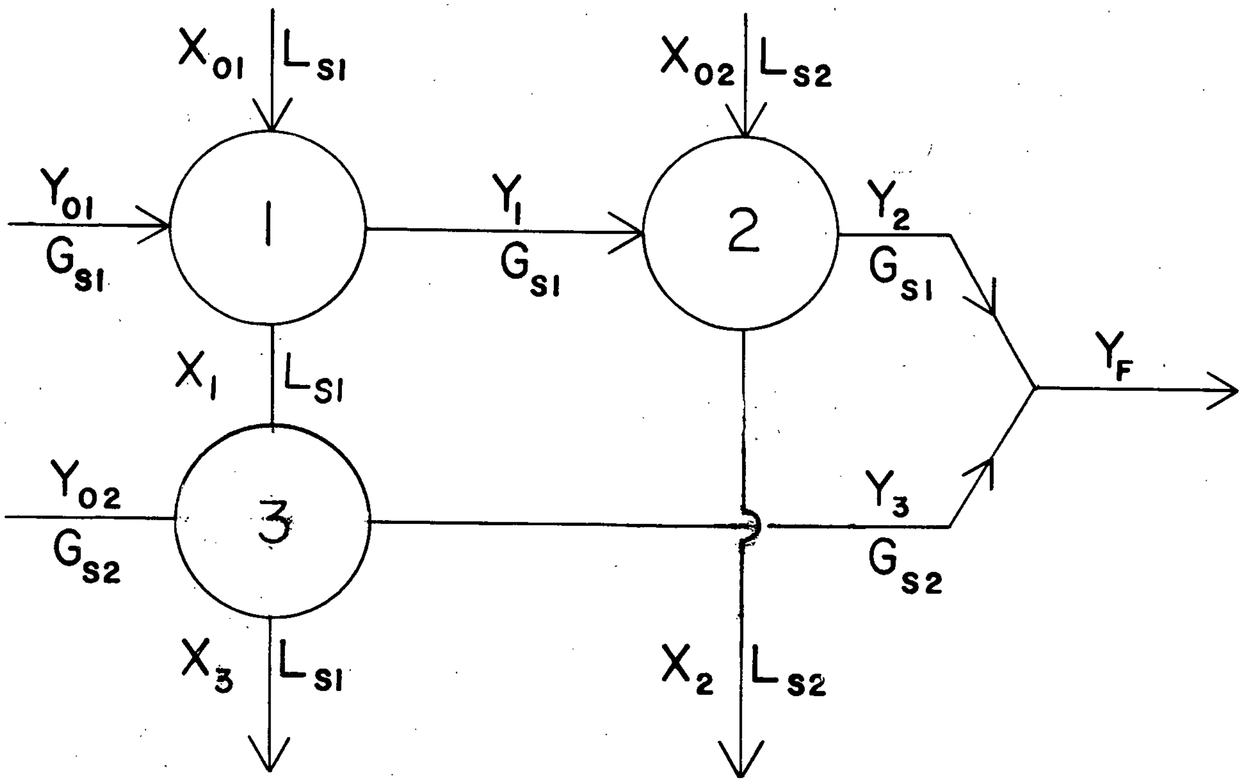

FIGURE 8. THREE STAGE SYSTEM D

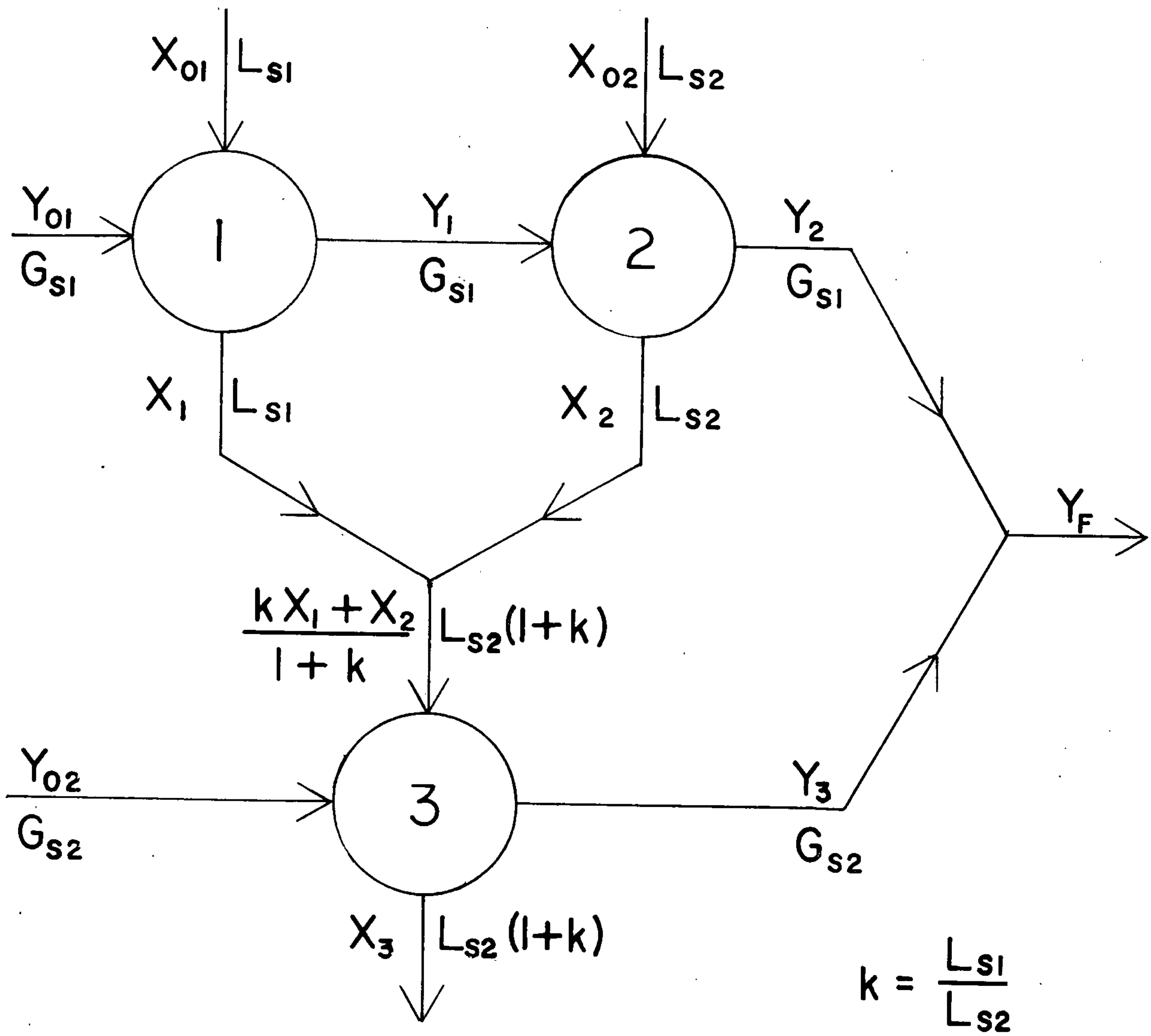

FIGURE 9. COMBINATION OF THREE STAGE SYSTEMS B AND D.

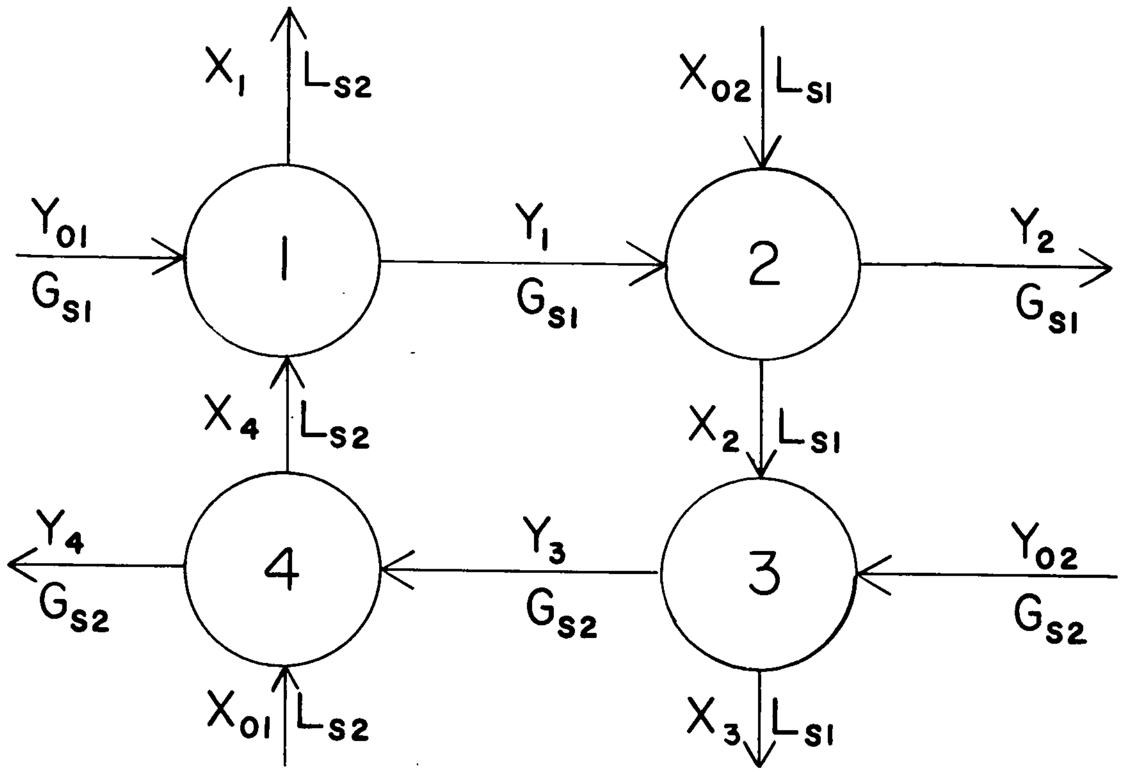

FIGURE 10. FOUR STAGE SYSTEM A

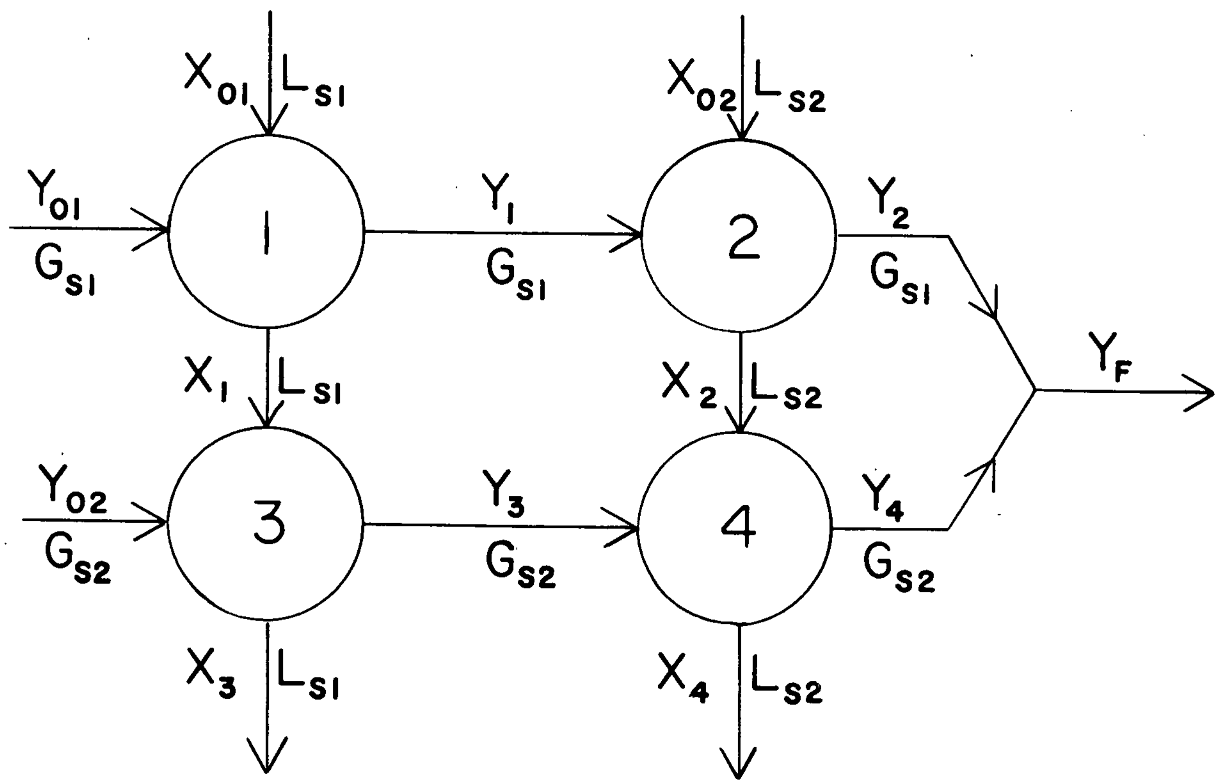

FIGURE 11. FOUR STAGE SYSTEM B

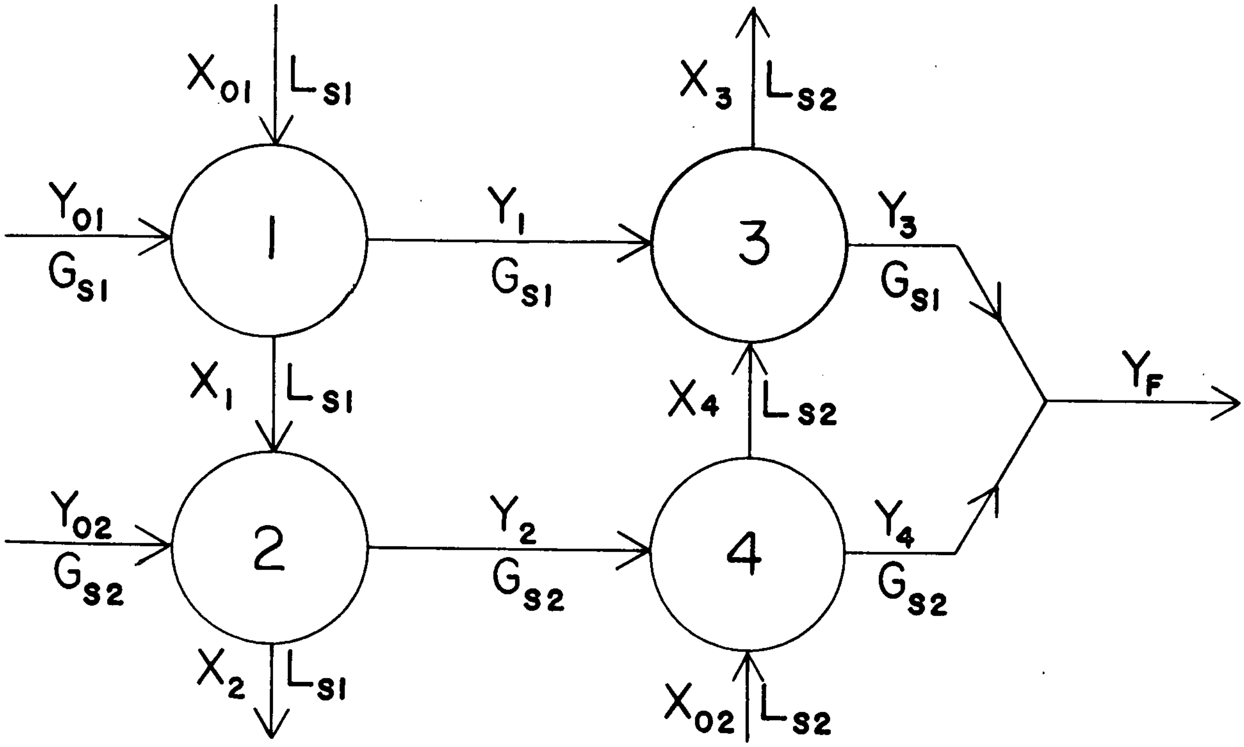

FIGURE 12. FOUR STAGE SYSTEM C.

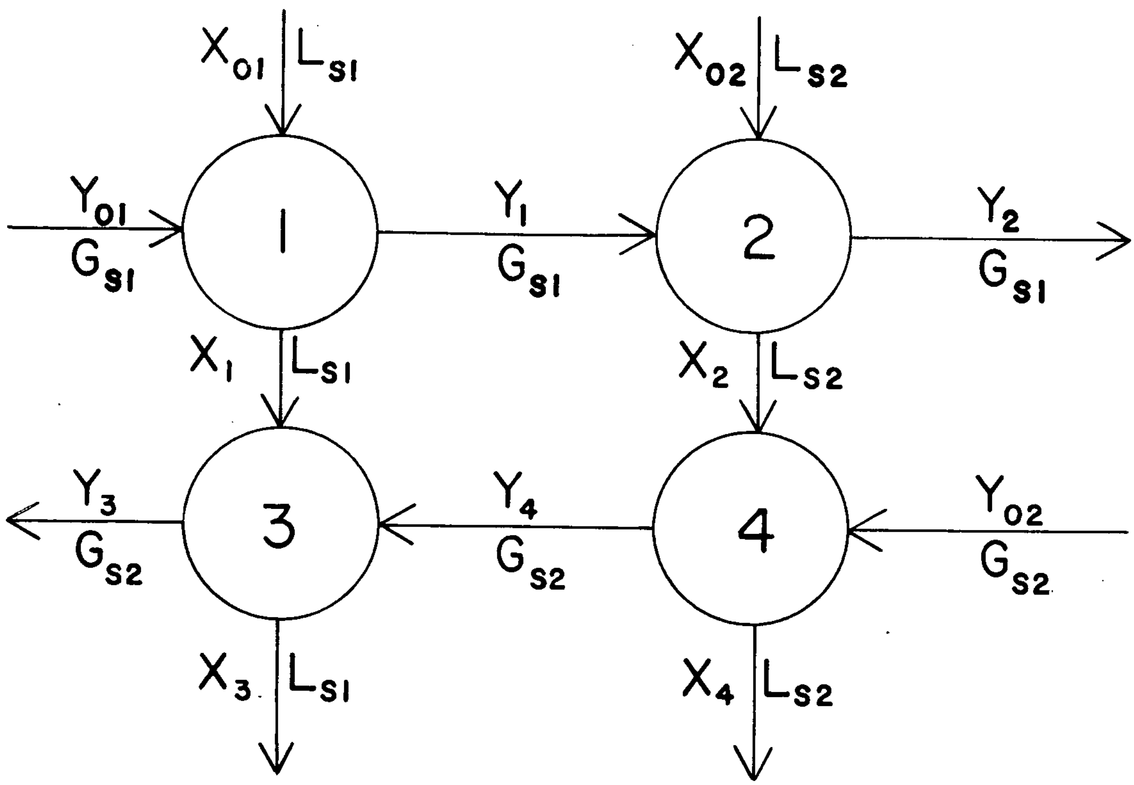

FIGURE 13. FOUR STAGE SYSTEM D

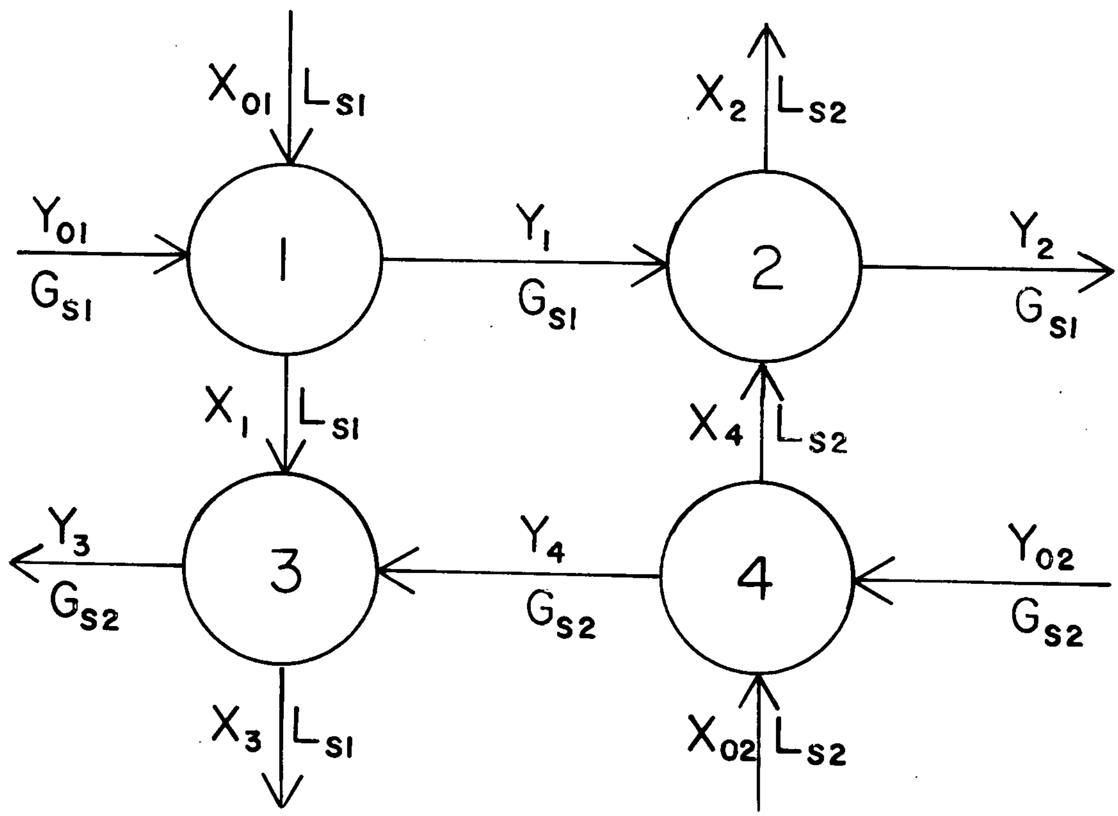

FIGURE 14. FOUR STAGE SYSTEM E

constraint:

<u>Theorem:</u> In a space S of real functions where convergence is defined, if:

i. $\psi(X_1,\ldots,X_n)$ and $\phi(X_1,\ldots,X_n)$ are lower semi-continuous functions.

ii. $\psi(X_1,\ldots,X_n) \geq 0$ for all X_i in S and there exists some X_i in S such that $\psi(X_1,\ldots,X_n) = 0$.

iii. $F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) + t_k\psi(X_{1k},\ldots,X_{nk})$, $k = 1,2,\ldots$ where t_k is a positive real number and F is a minimum for any k .

If:

$$\lim_{k\to\infty} t_k = \infty \tag{1.1}$$

then:

$$\lim_{k\to\infty} \psi(X_{1k},\ldots,X_{nk}) = 0 \tag{1.2}$$

$$\lim_{k\to\infty} F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) \tag{1.3}$$

Since $F(X_{1k},\ldots,X_{nk})$ is a sequence of minimum functions on S , $\phi(X_{1k},\ldots,X_{nk})$ is the minimum subject to the constraint $\psi(X_{1k},\ldots,X_{nk}) = 0$. The proof may be found in (3).

Weisman, Wood, and Rivlin (10) have generalized the theorem to be:

<u>Corollary:</u> If M functions $\psi_m(X_1,\ldots,X_n) \geq 0$, $m = 1,\ldots,M$ for all X_i in S and there exist X_i such that the

$\psi_m(X_1,\ldots,X_n) = 0$ simultaneously, and $F(X_{1k},\ldots,X_{nk}) =$

$\phi(X_{1k},\ldots,X_{nk}) + \sum_{m=1}^{M} t_{mk}\psi_m(X_{1k},\ldots,X_{nk})$, $k = 1,2,\ldots,$ where the t_{mk} are positive real numbers and F is a minimum for any k , then equations (1.1) and (1.3) of the theorem still hold and (1.2) becomes:

$$\lim_{k\to\infty} \psi_m(X_{1k},\ldots,X_{nk}) = 0 \qquad m = 1,\ldots,M \qquad (1.4)$$

2. Method of Carroll

Carroll (2) has proposed the following technique for solution of problems with **M' inequality constraints:**

i. Write all the inequality constraints so that they are positive:

$$\psi_m(X_{1k},\ldots,X_{nk}) \geq 0 \qquad m = M+1,\ldots,M+M' \quad (2.1)$$

ii. Choose a set of positive constants t_{m0} $m = M+1,\ldots,M+M'$.

iii. Choose the starting independent variables $(X_{10},\ldots,X_{n0})$ so that all the constraints are satisfied.

The general system may be written as follows:

$$F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) + \sum_{m=M+1}^{M+M'} \frac{t_{mk}}{\psi_m(X_{1k},\ldots,X_{nk})} \qquad (2.2)$$

The function $F(X_{1k},\ldots,X_{nk})$ is minimized successively for increasing k . The t_{mk} are decreasing with k so that:

$$\lim_{k\to\infty} t_{mk} = 0 \qquad m = M+1,\ldots,M+M' \tag{2.3}$$

$$\lim_{k\to\infty} F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) \tag{1.3}$$

Since the $F(X_{1k},\ldots,X_{nk})$ are successive minima the limiting solution is that $\phi(X_{1k},\ldots,X_{nk})$ is a minimum subject to the constraints $\psi_m(X_{1k},\ldots,X_{nk}) \geq 0$, $m = M+1,\ldots,M+M'$.

3. General Formulation

Using both the methods of (2) and (4) the general system becomes:

$$F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) + \sum_{m=1}^{M} t_{mk}\psi_m(X_{1k},\ldots,X_{nk}) + \sum_{m=M+1}^{M+M'} \frac{t_{mk}}{\psi_m(X_{1k},\ldots,X_{nk})} \tag{3.1}$$

where the $\psi_m, m = 1,\ldots,M$ are the equality constraints squared to satisfy condition ii. of Courant's Theorem. If:

$$\lim_{k\to\infty} t_{mk} = \begin{cases} \infty & m = 1,\ldots,M \\ 0 & m = M+1,\ldots,M+M' \end{cases} \tag{3.2}$$

then:

$$\lim_{k\to\infty} \psi_m(X_{1k},\ldots,X_{nk}) \begin{cases} = 0 & m = 1,\ldots,M \\ \geq 0 & m = M+1,\ldots,M+M' \end{cases} \tag{3.3}$$

$$\lim_{k\to\infty} F(X_{1k},\ldots,X_{nk}) = \phi(X_{1k},\ldots,X_{nk}) \tag{1.3}$$

In this manner $F(X_{1k},\ldots,X_{nk})$, the constrained optimum, may be determined.

C. OPTIMIZATION PROCEDURES

The adsorption networks involving constraints, discussed in section A, were examined using several different optimization methods. The first method to be tested in the present study was a modification of the pattern search method of Hooke and Jeeves (6). Weisman, Wood, and Rivlin (10) combined the pattern search procedure with the theorem of Courant (3) to solve problems involving equality constraints. However, when this procedure was applied to the four-stage network shown in fig. 10 it was found to give solutions highly dependent upon the starting parameters. In particular, the values of the acceleration and deceleration factors suggested by Weisman, Wood, and Rivlin (10) did not always result in the same solution (see table 1). However, by utilizing different values of these factors, the method could be made to yield the correct results. The network shown in fig. 10 proved to be very suitable for the evaluation of the various optimization methods since under the conditions of $Y_{01} = Y_{02} = 1.0$ and $X_{01} = X_{02} = 0$, it can be seen that this network becomes identical to the two-stage countercurrent flow configuration previously solved by Lerch (7). His results are shown in the last column of table 1 for comparison.

Since the pattern search method proved to be unreliable, other methods of optimization were investigated. It was shown by Box (1), who made a study of a wide variety of optimization methods, that for problems with sharp valleys, the ideas of Davidon (4), as formulated by Fletcher and

Powell (5), were the most successful although knowledge of the first derivatives is required. He also showed that the method of Powell (8) was a successful procedure when first derivatives are not known. Therefore, the methods of Fletcher and Powell (deflected gradient) and Powell (conjugate directions) were studied as alternatives to the pattern search technique. Both of the above procedures were modified to deal with equality constraints by employing the same theorem of Courant (3) as was used by Weisman, Wood, and Rivlin, (10). In addition, the inequality procedure of Carroll (2) was used when it was necessary to have limits on the range of variables. For example, the dimensionless concentrations X and Y had to be restricted to the range between 0 and 1 and the adsorbent-solvent ratios for each stage were required to be non-negative.

The general formulation for minimization with M equality and M' inequality constraints is given by equation (3.1). For the problems in this study the objective function, $\Phi(X_{1k},\ldots,X_{nk})$, is always equal to the ratio of total allocation of adsorbent to solvent; $L/G = \dfrac{\Sigma L_{si}}{\Sigma G_{si}}$. It is this function which must be minimized subject to the constraints on the system. $F(X_{1k},\ldots,X_{nk})$ in equation (3.1) is the constrained objective function. The coefficients t_{mk} are often referred to as penalty factors. The theorem of Courant (3) implies that if the penalty factors are increased with each iteration (index k), the equality constraints are satisfied more and more closely. However, the inequality constraints have their penalty factors decreased with each iteration. Initial high values of the penalty factors on the inequality constraints prevent them from being violated while restricting the variables to a narrow range. As the penalty factors are decreased, in subsequent iterations, the variables become freer to assume a wide range of values. It then follows from the theorem of Courant (3) and the method of Carroll (2) that equation (3.1) should converge to the constrained optimum, within certain prescribed tolerances.

The use of equation (3.1) remained the same irregardless of which of the three methods; pattern search, deflected gradient, or conjugate directions were employed. The methods themselves are discussed in more detail in the following section.

D. OPTIMIZATION METHODS

1. Pattern Search

Hooke and Jeeves (6) pattern search method is a type of direct search procedure, i.e. a sequential examination of a finite set of trial values of the function under study, incorporating a simple strategy for finding the various trial points. The pattern search program of Weisman, Wood, and Rivlin (10), modified to FORTRAN IV, is given in APPENDIX II.

2. Deflected Gradient

Fletcher and Powell (5) modified the method originally derived by Davidon (4), making it more efficient, and proving quadratic convergence. A more readily understandable description is given by Wilde and Beightler (11). The flow diagram and FORTRAN IV listing are in APPENDIX III. The method itself is such that the search moves in the direction of locally improving values of the objective function, but seldom exactly along the gradient, thereby explaining the name "deflected gradient". It is necessary with each step to find the minimum of the objective function along the deflected gradient. Fletcher and Powell (5), contending that the method of obtaining the minimum was not central to the theory, used a cubic interpolation technique given by Davidon (4), and found it to be satisfactory although it did not locate the minimum along the "deflected" gradient very accurately. However, for the class of problems considered in the present study, it was found that it is necessary to have a more precise estimate of the true one-dimensional minimum in order for the method to converge rapidly. Other one-dimensional minimization techniques considered were:

i. The parabolic approximation of Powell (8).

ii. The one-dimensional pattern search technique of Hooke and Jeeves (6). The flow diagram and FORTRAN IV list are found in APPENDIX VI.

iii. The classical Fibonacci search (see Wilde and Beightler (11)). The flow diagram and FORTRAN IV list are found in APPENDIX V.

3. Conjugate Directions.

The method of conjugate directions, formulated by Powell (8), has quadratic convergence like the deflected gradient method but does not require calculation of derivatives.

The method is a simple variation of the classical method of minimizing a function of several variables by changing one variable at a time. Since the objective function is the most important variable under consideration an additional convergence criterion was added to stop the program when little change occurs. The flow diagram and FORTRAN IV listing are in APPENDIX IV.

In the present study, the conjugate directions method was improved by incorporating the one-dimensional pattern search technique in preference to the parabolic interpolation technique originally used by Powell (8).

PROCEDURE

A. OPTIMIZATION METHODS

When $X_{01} = X_{02}$ and $Y_{01} = Y_{02}$ the system shown in fig. 10 becomes equivalent to a two-stage countercurrent system for which a known solution exists (7). Hence, the optimization procedures were tested on this simplified four-stage system using an exponent of 1 for the Freundlich isotherm to determine the best method.

1. Pattern Search

The starting values of the independent variables, the initial step size and the acceleration and decelerations ratios are varied. The solutions that result are tabulated and compared.

2. Conjugate Directions

All four methods for determining a one-dimensional minimum: cubic and parabolic interpolation, Fibonacci and one-dimensional pattern searches, were used and compared.

3. Deflected Gradient

All four methods for determing a one-dimensional minimum were used and compared.

B. FORMULATION OF ADSORPTION PROBLEMS

As previously mentioned in the THEORY, the adsorption systems all have certain similar characteristics in their formulation. Specific details with respect to various methods are discussed here. All the independent variables must be between 0 and 1 and all the adsorbent ratios for each stage must be greater than or equal to 0 .

1. General Two Stage System

The general two-stage system as shown in fig. 1 is a combination of the following networks:

i. Two-stage countercurrent.

ii. Two-stage crossflow with split adsorbent stream.

iii. Two-stage crossflow with split solvent stream.

The network produces the normal pair of independent variables (X_i) expressing the concentration in the adsorbent streams leaving the respective stages and the following:

k_1 - Fraction of the adsorbent stream leaving stage one and going to stage two; $0 \leq k_1 \leq 1$.

k_2 - Fraction of solvent stream leaving stage two and going to stage one; $0 \leq k_2 \leq 1$.

β_1 - Ratio of solvent streams (G_{s2}/G_{s1}); $\beta_1 \geq 0$.

β_2 - Ratio of adsorbent streams (L_{s2}/L_{s1}); $\beta_2 \geq 0$.

It is easily seen that when $k_1 = k_2 = 1$ and $\beta_1 = \beta_2 = 0$ the system becomes two-stage countercurrent; when $k_1 = 0$, $k_2 = 1$, $\beta_1 = 0$, and $\beta_2 \geq 0$ the system is two-stage crossflow with split adsorbent stream; and when $k_1 = 1$, $k_2 = 0$, $\beta_1 \geq 0$, and $\beta_2 = 0$ the system is two-stage crossflow with split solvent stream. The Koble-Corrigan isotherms shown in fig. 2 and Freundlich isotherms were used in this system.

2. Crossflow Systems

Since Lerch (7) showed that two-stage crossflow with split adsorbent stream is superior to two-stage crossflow with split solution stream, only systems of the first type were solved (see fig. 3).

3. Countercurrent Systems

As was previously noted in the THEORY, an N-stage countercurrent network (see fig. 4) formulates into a problem of N-1 variables and N-1 equality constraints.

4. Three-stage Systems

Four three-stage configurations (see figs. 5-8) with no joining or separating streams are optimized. They all have three independent variables and one equality constraint (the final solution concentration is constant). A combination of systems B and D (figs. 6 and 8) gives the system in figure 9, in which the adsorbent streams from stages one and two are joined before entering stage three. The joining of the two streams creates a new independent variable k ($k=L_{s1}/L_{s2}$), which is the ratio of the adsorbent streams.

5. Four-stage Systems

Five four-stage systems are created following the example of Treybal (9), (see figs. 10-14). They have four independent variables and two equality constraints. The first constraint is the fixed value of the final solution concentration, and the second involves the adsorbent-solution ratios. A thorough discussion of the constraints is found in APPENDIX VII where the system of fig. 10 has sample calculation of derivatives for the Fletcher-Powell method.

C. VARIATION OF PENALTY FACTORS

1. Equality Constraints

Initially, the penalty factors are set values of the order of 10. When only one constraint occurs in the problem the penalty factor is increased by a factor of 10 after each minimization until it reaches a value of around 10^8. The program is then terminated. When more than one equality constraint occurs the penalty factor corresponding to the constraint function of largest absolute value is increased by a factor of 10. Each of the other penalty factors is multiplied by 10 times the ratio of its corresponding constraint function to the largest constraint function. The program is then terminated when one penalty factor becomes approximately 10^8. This procedure tends to keep the values of the constraint functions approximately equal.

2. Inequality Constraints

The penalty factors on the inequality constraints are initially set to be of the order of 10. After each minimization the factors are multiplied by pre-arranged constant factors varying from 0.1 to 0.5 depending on the importance given to their respective constraints. Constraints that tend to be violated have to have their penalty factors decreased less rapidly.

RESULTS

The initial concentrations in the adsorbent and solvent streams are 0.0 and 1.0 respectively, and the final solution concentration is 0.1 in all cases. The Freundlich isotherm is used for all systems with the exponent varying from 0.3 to 3.0 . The Koble-Corrigan isotherm is used only in the general two-stage system.

A. OPTIMIZATION METHODS

Four-stage system A (see fig. 10) with a Freundlich exponent of 1 is used as a comparison of each method's effectiveness.

1. Basic Methods

a. Pattern Search

The most serious limitation of the Pattern Search procedure is its inability to give identical results for different initial and operating conditions. Table 1 shows that although the objective function is the same except for one case, to 3 significant figures, the independent variables are not, and deviate quite markedly from the expected solution of two stage countercurrent, which is shown in the last column of table 1.

b. Conjugate Directions

As shown in table 2, the solution determined by Conjugate Direction method approaches the true optimum for moderate values of the penalty factors, but then veers away as the factors are increased. The diverging property is probably due to two things:

i. The creation of a great many conjugate directions could cause the loss of an independent variable. Zangwill (12), in a very recent paper, has shown that this can occur, and when it does the method breaks down.

ii. The large penalty factors create very sharp bends in the valley that the method is following. This could cause the method to appear as if it has found an optimum and stop.

c. Deflected Gradient

In contrast to the previous procedures, the optimum (see table 16), determined by the Deflected Gradient method, is very close to the expected with very little variation due to different starting values.

2. One-dimensional Minimization

When the deflected gradient and conjugate directions methods were tried using parabolic and cubic interpolation, no solution was obtained near the expected value. The Fibonacci and One Dimensional Pattern searches give almost equivalent results that are quite acceptable when used with the deflected gradient method.

3. Computation Time

a. Pattern Search

The computation time (see table 1) varies considerably from run to run. It does seem to be somewhat dependent on how accurate the solution is; the longer the time, the closer the answer to the expected solution.

b. Conjugate Directions

The computation time is dependent on the accuracy of the final solution. For low accuracy the method gives results faster than pattern search. However, for high accuracy, the pattern search time is less.

c. Deflected Gradient

The computation time is about 20 seconds for four-stage system A, a good deal faster than the best time of the other methods.

d. One-dimensional Minimization

Although approximately 1.5 times faster than one-dimensional pattern search, Fibonacci does not give quite as good results, implying that to get the best results a thorough search for the optimum must be undertaken.

B. ADSORPTION SYSTEMS

The results discussed below were all determined using the modified deflected gradient method of Fletcher and Powell (5), using either Fibonacci or one-dimensional pattern searches to determine the one-dimensional minima.

1. Two-stage Systems

The optimum generalized two-stage system becomes equivalent to the two-stage countercurrent system for isotherms of both the Freundlich and Koble-Corrigan variety (see tables 3 and 4). These results are in good agreement with Lerch (7) for two-stage countercurrent (see table 5).

2. Countercurrent and Crossflow Systems

A comparison, in fig. 15, is made between two, three, and four-stage countercurrent systems and two, five, and forty-stage crossflow, with split adsorbent stream, systems. The results are found in tables 5, 8, and 15 for countercurrent and tables 6, 7, 14, and 20 for crossflow. It is apparent that for easy separability as determined by a high exponent

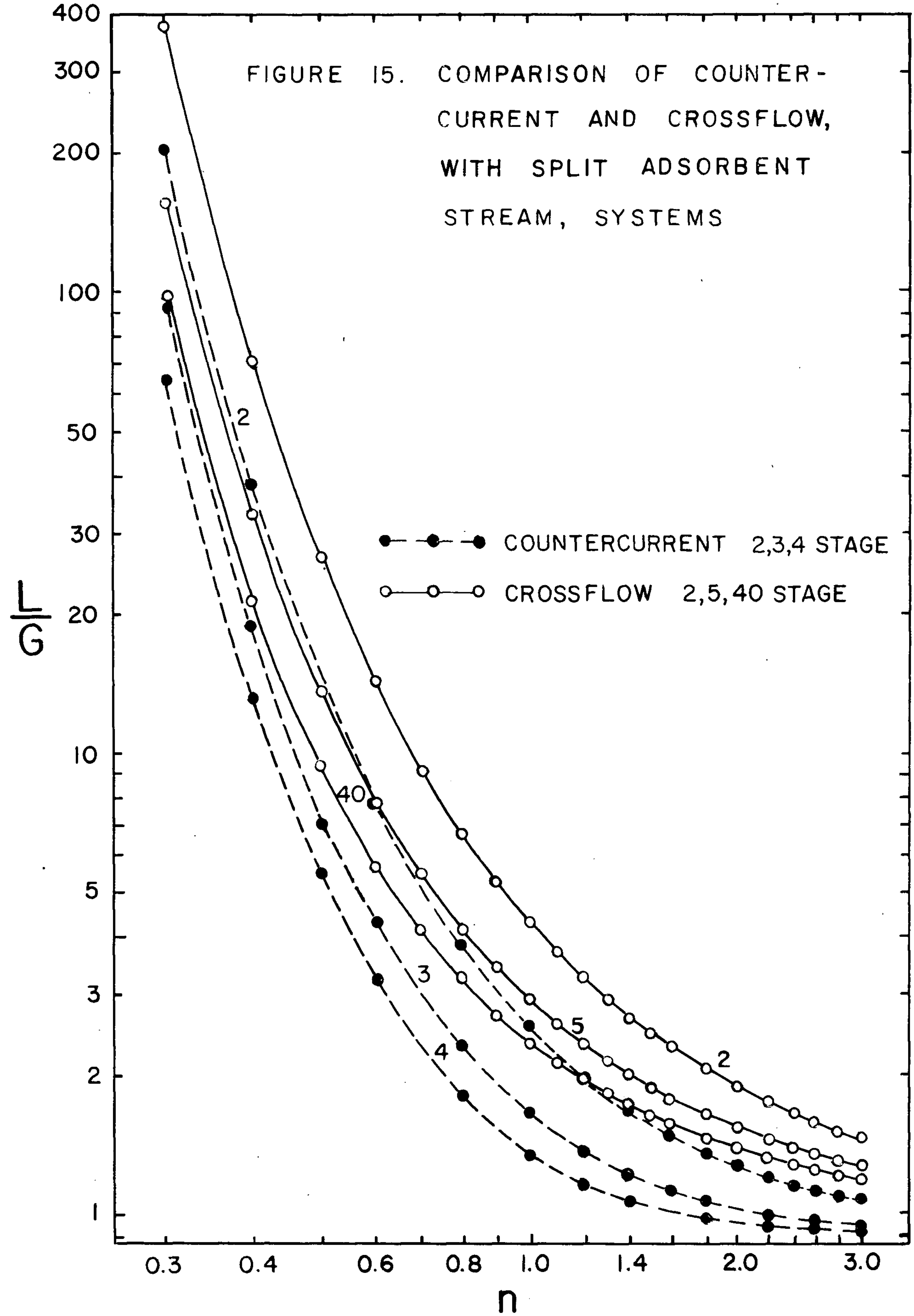
FIGURE 15. COMPARISON OF COUNTER-CURRENT AND CROSSFLOW, WITH SPLIT ADSORBENT STREAM, SYSTEMS
COUNTERCURRENT 2,3,4 STAGE
CROSSFLOW 2,5,40 STAGE
L/G
n
400
300
200
100
50
30
20
10
5
3
2
1
0.3
0.4
0.6
0.8
1.0
1.4
2.0
3.0
2
40
3
4
5
2

value on the Freundlich isotherm, countercurrent procedures are far superior. However, as the exponent value decreases, the effectiveness of the countercurrent systems decreases faster than that of the crossflow systems. Over the range of Freundlich isotherms tested, the three-stage countercurrent is superior in all cases to forty-stage crossflow. It is doubted whether this superiority would remain for lower Freundlich exponents.

3. Three-stage Systems

Three-stage countercurrent is the most effective of all three-stage systems over the range of Freundlich isotherms tested (see fig. 16). Systems A and B (see figs. 5 and 6) have almost identical objective functions, slightly better than the identical pair C and D (see figs. 7 and 8). Systems A and B are markedly inferior to three-stage crossflow for low Freundlich exponents but become superior at an exponent value of 2.4. The results for three-stage systems are in tables 7 to 13.

The systems shown in fig. 9 is found to give solutions equivalent to systems B (see fig. 6), implying that an optimum combination of the two systems does not exist for the isotherms tested. The solution is in table 13.

4. Four-stage Systems

Fig. 17 shows that countercurrent is superior to all other four-stage systems over the range of Freundlich isotherms tested. For Freundlich exponents below 0.4 crossflow is superior to all of systems A - D (see figs. 10 - 13). However, systems A and B become superior to crossflow at exponent values of 0.4 and 1.2 respectively. It should be noted that the

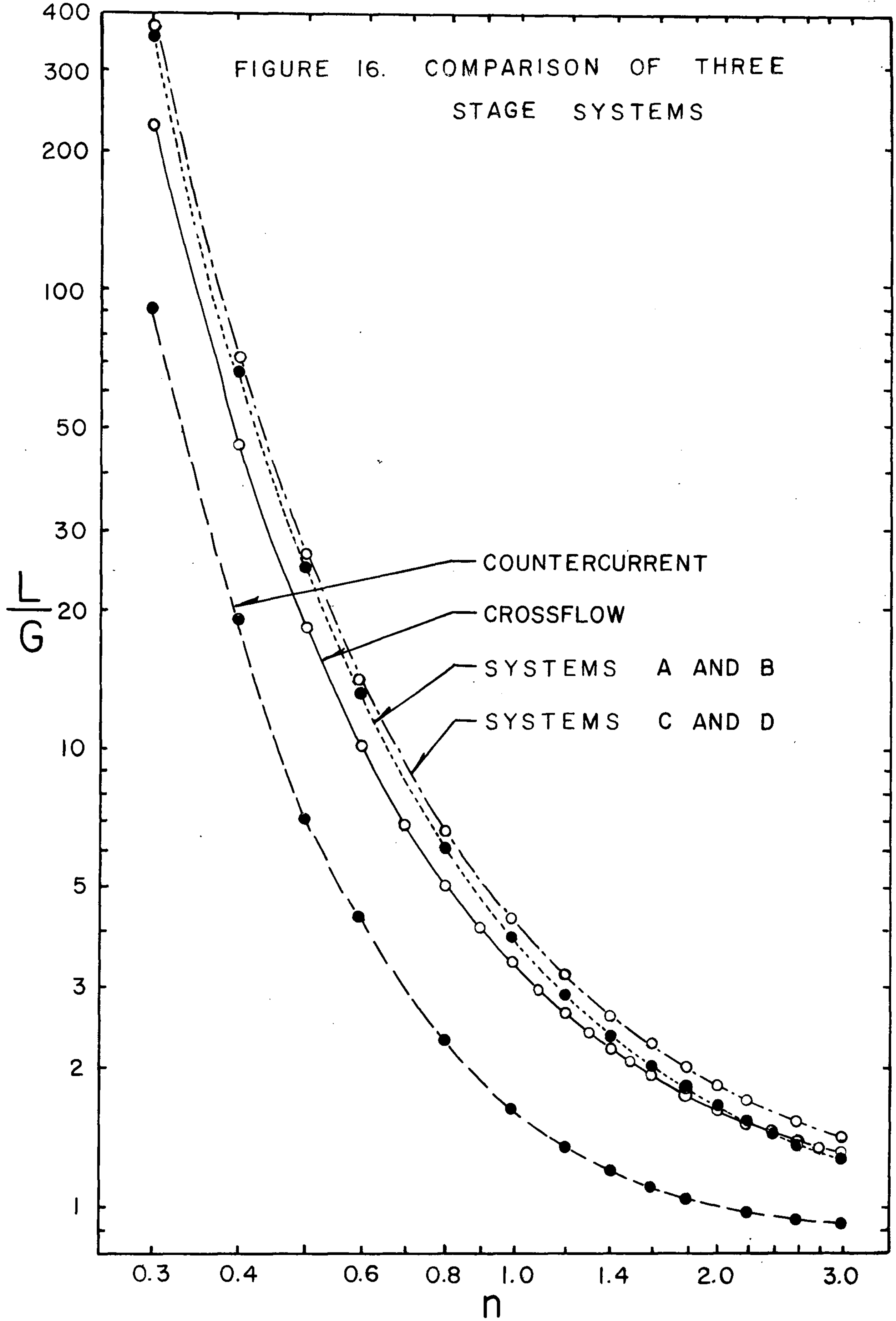
FIGURE 16. COMPARISON OF THREE STAGE SYSTEMS
400
300
200
100
50
30
20
10
5
3
2
1
L/G
COUNTERCURRENT
CROSSFLOW
SYSTEMS A AND B
SYSTEMS C AND D
0.3
0.4
0.6
0.8
1.0
1.4
2.0
3.0
n

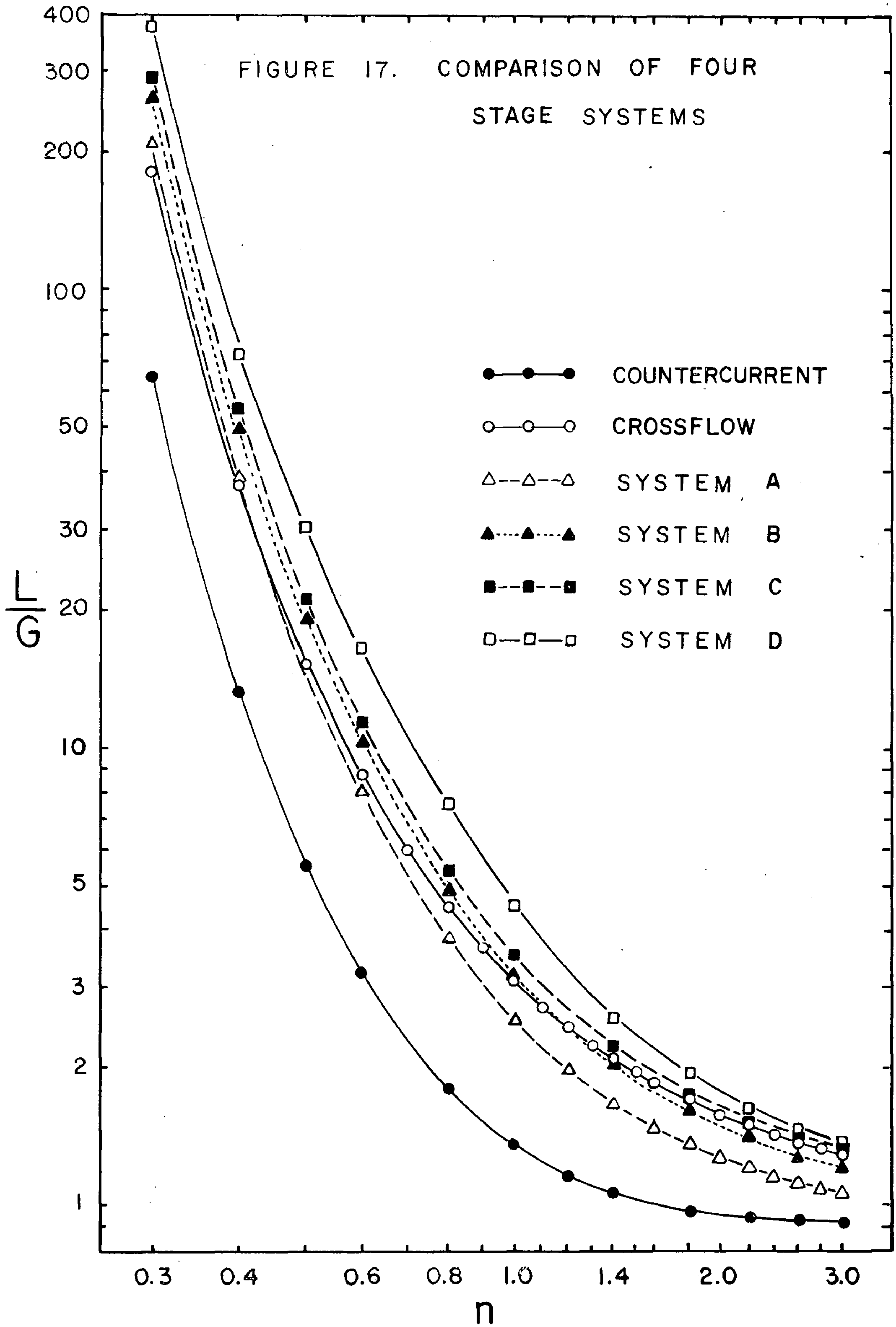
FIGURE 17. COMPARISON OF FOUR STAGE SYSTEMS
COUNTERCURRENT
CROSSFLOW
SYSTEM A
SYSTEM B
SYSTEM C
SYSTEM D
400
300
200
100
50
30
20
10
5
3
2
1
L/G
0.3
0.4
0.6
0.8
1.0
1.4
2.0
3.0
n

points of crossover mentioned are only valid for the conditions used. Systems A, B, C, and D are effective in descending order over the range of isotherms tested. System E (see fig. 14) does not have a feasible solution. It can be shown that the feasible values for the independent variables are $X_1 = X_2 = X_3 = X_4$ which contradict the initial conditions of the problem. A possible explanation for the relative effectiveness may lie in the resemblances of the various configurations to countercurrent flow. System A, equivalent to two-stage countercurrent when $X_{01} = X_{02}$ and $Y_{01} = Y_{02}$, is the most effective. The effectiveness of systems B, C, and D decrease in that order, which is also the order of decreasing similarity of these systems to countercurrent flow. Some of the solutions for system D may be in doubt since there was difficulty in obtaining consistent results. The results for four-stage systems are in tables 14 - 19.

5. Effect of Number of Stages on Crossflow Systems

Fig. 18 shows the general effect of stages on effectiveness in crossflow systems. For large values of the Freundlich exponent the effectiveness is nearly independent of the number of stages. The separation is easy enough so that the first few stages accomplish it. As the exponent decreases the number of stages has more and more effect on the effectiveness of the system. The near independence of the effectiveness on the number of stages is shown analytically in APPENDIX VII for the case when the Freundlich isotherm exponent is 1 .

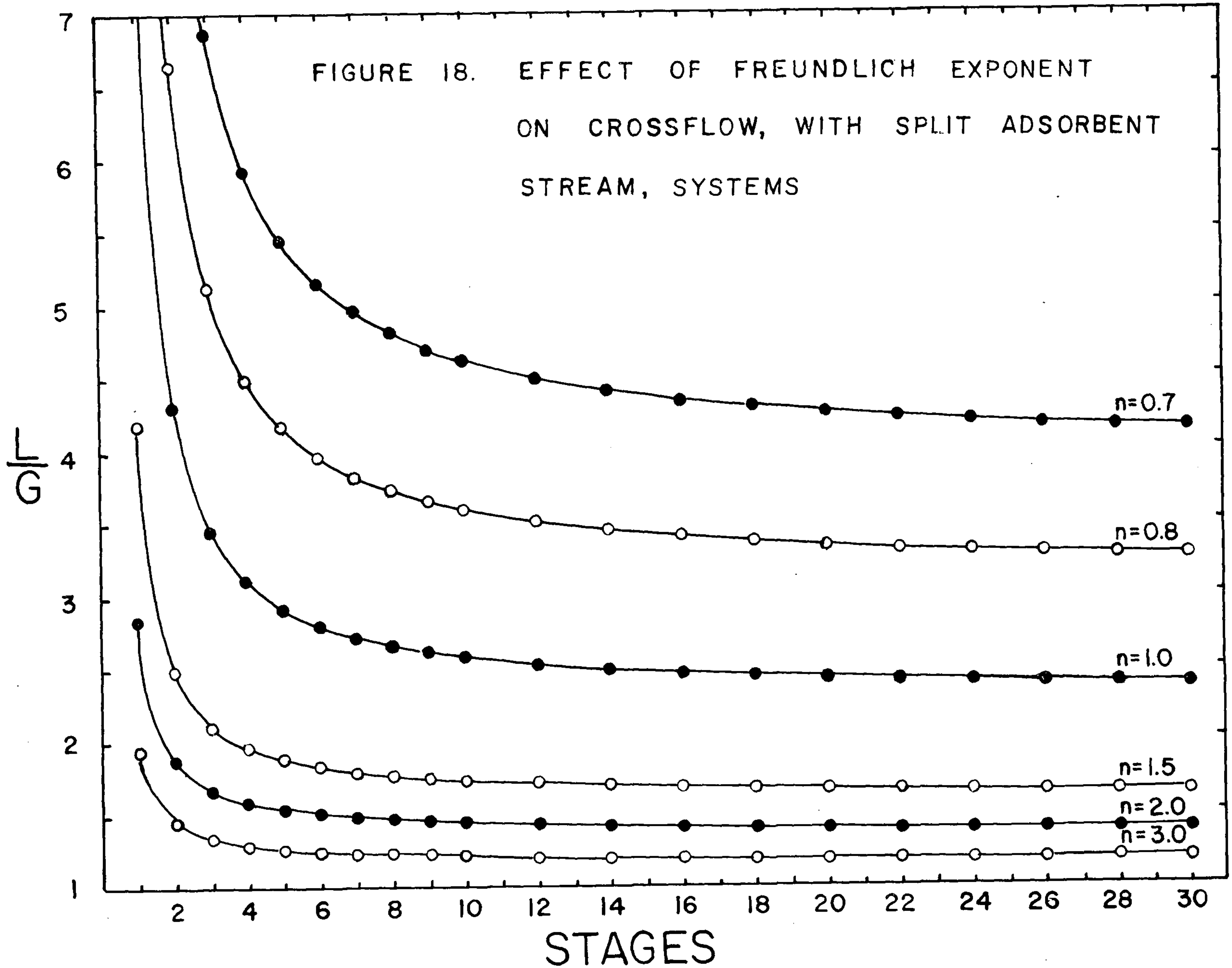

FIGURE 18. EFFECT OF FREUNDLICH EXPONENT ON CROSSFLOW, WITH SPLIT ADSORBENT STREAM, SYSTEMS

The limiting L/G factor as the number of stages approach infinity is the natural logarithm of 10 , for $Y_F = 0.10$, which is the only value considered in this study.

CONCLUSIONS

The deflected gradient method of Fletcher and Powell (5) using either Fibonacci or pattern search one-dimensional minimization subroutines is found to be a successful method of optimizing constrained adsorption systems. On a test problem the solutions obtained by this method are more accurate and can be computed faster than either the pattern search or conjugate direction methods. Therefore, the deflected gradient method was used on all remaining problems.

A general two-stage network converges to the countercurrent network at optimum, confirming the superiority of countercurrent over other two-stage networks.

Countercurrent networks are superior to any others having the same number of stages for Freundlich isotherms between 0.3 and 3.0 . In three-stage systems, excluding countercurrent, crossflow is superior to all others for low exponent values of the Freundlich isotherm but becomes inferior to some of the other networks at very high exponent values. A combination of two types of three-stage networks gives solutions identical to the superior system of the two.

In four-stage networks, excluding countercurrent, the crossflow was most effective at low Freundlich exponent, but became inferior to some of the others as the exponent increased. The other systems are believed to be effective according to their resemblance to countercurrent networks.

The effect of varying the number of stages in crossflow networks is only appreciable for small Freundlich exponent. For exponent values over 2.0 there is virtually no change in effectiveness after a few stages.

In all problems considered in this study the final solute concentration was taken to be one-tenth of the initial concentration. This resulted in "cross-over" points where one configuration becomes superior or inferior to another at certain specific values of the Freundlich exponent. For other values of the final concentration these cross-over points would undoubtedly be shifted.

RECOMMENDATION FOR FURTHER STUDY

A more practical problem would involve the optimization of a real adsorption system taking into account the following considerations:

i. Cost of adsorbent.

ii. Operation of stages at different temperatures.

iii. Equipment and maintenance cost.

iv. Purity of final product.

Zangwill's (12) modification to Powell's (8) procedure should be investigated to see if the modified method becomes suitable for constrained adsorption systems.

NOMENCLATURE

The following symbols hold in all sections except Appendices II to VI.

F	constrained objective function
G_{sj}	quantity of solute-free solvent in solvent stream j, lb.
G	$\Sigma\, G_{sj}$, total quantity of solute-free solvent, lb.
L_{si}	quantity of solute-free adsorbent in adsorbent stream i, lb.
L	$\Sigma\, L_{si}$, total quantity of solute-free adsorbent, lb.
m	parameter in Koble-Corrigan isotherm
M	number of equality constraints
M'	number of inequality constraints
n	number of independent variables (section A of Theory only)
n	parameter in Freundlich and Koble-Corrigan isotherms
N	total number of stages in a configuration
S	space of real lower semi-continuous functions
t_{mk}	constraint penalty factor
X_i	dimensionless concentration of solute in adsorbent stream leaving stage i
X_{0i}	dimensionless initial concentration of solute in adsorbent stream i
Y_i	dimensionless concentration of solute in solvent stream leaving stage i
Y_{0j}	dimensionless initial concentration of solute in solvent stream j

Subscripts

F	index indicating final concentration
i	index for a stage or a stream
j	index for a stream
k	index for number of iterations

Continued....

Subscripts (Continued)

m	index denoting constraint number
O	index indicating initial concentration

Greek

α	solvent-adsorbent ratio
Φ	objective function
ψ	constraint function

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APPENDIX I

Tables of Results

The Freundlich Isotherm is used for the results in all tables except table 4 where the Koble-Corrigan Isotherm is used. The Deflected Gradient procedure has test results in table 16.

Table

TABLE 1.

Four Stage System A - Pattern Search Method

Freundlich exponent = 1.0

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

								True Optimum
Acceleration Factor		1.1111	2.0000	2.0000	2.0000	2.0000	2.5000	
Deceleration Factor		0.1000	0.1000	0.1000	0.2000	0.2500	0.2500	
Initial Step Size		0.1000	0.1000	0.1500	0.1500	0.1500	0.2000	
	X_1	0.8000	0.8000	0.8000	0.8000	0.8000	0.5100	
Starting	X_2	0.2000	0.2000	0.2000	0.2000	0.2000	0.4900	
Variables	X_3	0.8000	0.8000	0.8000	0.8000	0.8000	0.5100	
	X_4	0.2000	0.2000	0.2000	0.2000	0.2000	0.4900	
	X_1	0.4667	0.3844	0.3868	0.3551	0.3646	0.3548	0.3541
Final	X_2	0.1365	0.1046	0.1014	0.1023	0.1020	0.1031	0.1000
Variables	X_3	0.3899	0.3291	0.3276	0.3548	0.3451	0.3541	0.3541
	X_4	0.1467	0.0971	0.1004	0.0986	0.0986	0.0973	0.1000
	L/G	2.0380	2.5416	2.5454	2.5341	2.5376	2.5383	2.5410
Time (sec.)		65.6	99.6	80.4	73.5	108.4	92.1	

TABLE 2.

Four Stage System A - Conjugate Direction Method

Freundlich exponent = 1.0.
Penalty factors increase with each iteration.
Different end tolerances used.

Initial $X_{01} = X_{02} = 0.0$ $Y_{01} = Y_{02} = 1.0$

Final $Y_F = 0.1$

Iteration	F*	X_1	X_2	X_3	X_4	time
Start	101.0444	0.40000	0.20000	0.40000	0.20000	
1	23.3618	0.70008	0.36728	0.70043	0.37317	
2	5.1705	0.50429	0.18060	0.46426	0.17954	
3	2.89820	0.34915	0.11800	0.39351	0.10044	
4	2.59301	0.33728	0.10749	0.37351	0.09355	
5	2.56189	0.33628	0.10661	0.37177	0.09295	
6	2.56506	0.33628	0.10660	0.37176	0.09295	
7	2.55866	0.33611	0.10650	0.37155	0.09286	32.38 sec.
Start	101.04444	0.40000	0.20000	0.40000	0.20000	
1	23.34164	0.70071	0.36772	0.69997	0.37310	
2	5.17166	0.50608	0.18068	0.46279	0.17959	
3	2.87804	0.37632	0.11583	0.36117	0.10055	
4	2.58010	0.36511	0.10654	0.34536	0.09468	
5	2.55011	0.36389	0.10556	0.34367	0.09403	
6	2.54746	0.36369	0.10541	0.34341	0.09393	32.05 sec.
Start	101.04444	0.40000	0.20000	0.40000	0.20000	
1	23.34069	0.70327	0.37079	0.70022	0.37327	
2	5.16573	0.48847	0.17921	0.48567	0.18509	
3	2.86796	0.37215	0.10673	0.36702	0.11082	
4	2.57100	0.35988	0.10023	0.35451	0.10308	
5	2.54491	0.35735	0.09859	0.35134	0.10163	
6	2.54991	0.35737	0.09846	0.35102	0.10161	
7	2.58468	0.35768	0.09859	0.35113	0.10166	
8	2.89342	0.35785	0.09877	0.35136	0.10169	
9	9.72683	0.35796	0.09885	0.35147	0.10172	90.5 sec.
Start	101.04444	0.40000	0.20000	0.40000	0.20000	
1	23.34083	0.70235	0.37056	0.69955	0.37175	
2	5.16340	0.48447	0.17958	0.48250	0.17968	
3	2.86753	0.37118	0.10822	0.36715	0.10875	
4	2.57103	0.35869	0.10083	0.35443	0.10179	
5	2.54473	0.35551	0.09923	0.35319	0.10100	
6	2.57575	0.35609	0.09943	0.35345	0.10122	
7	3.27035	0.35644	0.09957	0.35360	0.10135	
8	12.44869	0.35670	0.09962	0.35366	0.10145	65.5 sec.

*F is the constrained objective function (= L/G plus constraints)

TABLE 3.

General Two Stage System

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.0	0.4
X_1	0.09999	0.00316
X_2	0.35406	0.02326
k_1	0.99994	0.99973
k_2	0.99995	0.99988
β_1	0.00006	0.00008
β_2	0.00027	0.00021
Y_1	0.09999	0.10002
Y_2	0.35406	0.22216
$(L/G)_1$	2.54149	38.62495
$(L/G)_2$	2.54149	38.69969
L/G	2.54204	38.68355

TABLE 4.

General Two Stage System

Koble-Corrigan Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	0.3	0.2	0.2
m	1.7	2.0	1.9
X_1	0.00318	0.00033	0.00026
X_2	0.02959	0.00438	0.00342
k_1	1.00000	1.00000	1.00000
k_2	1.00000	1.00000	1.00000
β_1	0.00000	0.00000	0.00000
β_2	0.00000	0.00000	0.00000
Y_1	0.100000	0.10000	0.10000
Y_2	0.19665	0.16784	0.16781
$(L/G)_1$	30.42287	205.7613	263.1578
$(L/G)_2$	30.42287	205.7613	263.1578
L/G	30.42055	205.62225	263.26908

TABLE 5.

Two Stage Countercurrent

Freundlich Isotherm Used

(The Values marked with a * are due to Lerch)

Initial $Y_{01} = 1.0$ $X_{01} = 0.0$

Final $Y = 0.1$

n	X_1	X_2	Y_1	Y_2	L/G
3.0	0.84170	0.46416	0.59631	0.10000	1.06927
2.8	0.82336	0.43940	0.58030	0.10000	1.09308
2.6	0.80176	0.41246	0.56300	0.10000	1.12253
2.4	0.77611	0.38312	0.54427	0.10000	1.15963
2.2	0.74542	0.35112	0.52393	0.10000	1.20738
2.0*	0.7084	0.3162	0.5018	0.10000	1.271
1.8*	0.6633	0.2783	0.4776	0.1000	1.357
1.6*	0.6080	0.2371	0.4510	0.1000	1.480
1.4*	0.5399	0.1931	0.4219	0.1000	1.667
1.2*	0.4560	0.1467	0.3897	0.1000	1.974
1.0*	0.3541	0.1000	0.3541	0.1000	2.541
0.8*	0.2357	0.0562	0.3147	0.1000	3.818
0.6*	0.1134	0.0215	0.2709	0.1000	7.934
0.4*	0.0233	0.0032	0.2222	0.10000	38.655
0.3	0.00436	0.00046	0.19581	0.10000	206.42484
0.2	0.00014	0.00001	0.16976	0.10000	6339.28312

TABLE 6.

Two Stage Crossflow - Algorithm Solution

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	X_1	X_2	L/G
3.0	0.72146	0.46416	1.45920
2.8	0.70315	0.43940	1.51302
2.6	0.68236	0.41246	1.57809
2.4	0.65854	0.38312	1.65809
2.2	0.63101	0.35112	1.75873
2.0	0.59891	0.31623	1.88885
1.8	0.56110	0.27826	2.06306
1.6	0.51606	0.23714	2.30692
1.5	0.49025	0.21544	2.46874
1.4	0.46186	0.19307	2.66930
1.3	0.43059	0.17013	2.92362
1.2	0.39610	0.14678	3.25475
1.1	0.35807	0.12328	3.70016
1.0	0.31623	0.10000	4.32456
0.9	0.27048	0.07743	5.24724
0.8	0.22106	0.05623	6.70930
0.7	0.16893	0.03728	9.25827
0.6	0.11624	0.02154	14.35702
0.5	0.06717	0.01000	26.94636
0.4	0.02820	0.00316	71.20493
0.3	0.00607	0.00046	379.57169

TABLE 7.

Three Stage Crossflow - Algorithm Solution

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$ $X_{03} = 0.0$

Final $Y_F = 0.1$

n	X_1	X_2	X_3	L/G
3.0	0.81507	0.63564	0.46416	1.34827
2.8	0.80170	0.61386	0.43940	1.39028
2.6	0.78630	0.58943	0.41246	1.44085
2.4	0.76836	0.56185	0.38312	1.50276
2.2	0.74722	0.53055	0.35112	1.58022
2.0	0.72199	0.49478	0.31623	1.67976
1.8	0.69139	0.45368	0.27826	1.81199
1.6	0.65362	0.40619	0.23714	1.99542
1.5	0.63127	0.37970	0.21544	2.11616
1.4	0.60606	0.35119	0.19307	2.26494
1.3	0.57744	0.32054	0.17013	2.45217
1.2	0.54480	0.28767	0.14678	2.69402
1.1	0.50734	0.25258	0.12328	3.01631
1.0	0.46416	0.21544	0.10000	3.46331
0.9	0.41423	0.17665	0.07743	4.11549
0.8	0.35657	0.13702	0.05623	5.13341
0.7	0.29053	0.09799	0.03728	6.87596
0.6	0.21674	0.06184	0.02154	10.28372
0.5	0.13886	0.03180	0.01000	18.46101
0.4	0.06664	0.01132	0.00316	46.15715
0.3	0.01708	0.00190	0.00046	229.56334

TABLE 8.

Three Stage Countercurrent

Freundlich Isotherm Used

Initial $X_0 = 0.0$ $Y_0 = 1.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	1.8	1.6	1.4	1.2
X_1	0.95373	0.93379	0.90110	0.84442	0.80052	0.74026	0.65687
X_2	0.81332	0.76453	0.69610	0.59820	0.53410	0.45761	0.36779
X_3	0.46416	0.41246	0.35112	0.27826	0.23714	0.19367	0.14678
Y_1	0.86751	0.83686	0.79525	0.73757	0.70048	0.65636	0.60392
Y_2	0.53801	0.49754	0.45061	0.39657	0.36661	0.33473	0.30111
Y_3	0.10000	0.10000	0.10000	0.10000	0.10000	0.10000	0.10000
L/G	0.94367	0.96381	0.99878	1.06582	1.12427	1.21578	1.37013

Continued.....

TABLE 8. (CONTINUED)

Three Stage Countercurrent

Freundlich Isotherm Used

Initial $X_0 = 0.0$ $Y_0 = 1.0$

Final $Y_F = 0.1$

n	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.54191	0.38856	0.20512	0.11676	0.04733	0.00952
X_2	0.26608	0.15950	0.06531	0.03136	0.01026	0.00156
X_3	0.10000	0.05623	0.02154	0.01000	0.00316	0.00046
Y_1	0.54191	0.46943	0.38655	0.34170	0.29515	0.24753
Y_2	0.26608	0.23025	0.19453	0.17708	0.16014	0.14388
Y_3	0.10000	0.10000	0.10000	0.10000	0.10000	0.10000
L/G	1.66080	2.31622	4.38763	7.08010	19.01685	91.50349

TABLE 9.

Three Stage System A

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	2.0	1.8	1.6	1.4
X_1	0.43813	0.37943	0.31740	0.27268	0.23849	0.20247	0.16378
X_2	0.75904	0.71776	0.66244	0.62876	0.58743	0.53861	0.47946
X_3	0.48394	0.43341	0.36948	0.33590	0.29493	0.25051	0.20324
Y_1	0.08410	0.08049	0.08008	0.07436	0.07576	0.07765	0.07943
Y_2	0.43732	0.42222	0.40413	0.39534	0.38381	0.37156	0.35732
Y_3	0.11334	0.11375	0.11187	0.11283	0.11105	0.10917	0.10745
L_{s1}/G_{s1}	2.09047	2.42341	2.89829	3.39454	3.87551	4.55560	5.62081
L_{s1}/G_{s2}	1.75334	1.70773	1.72696	1.69808	1.76587	1.86957	2.03587
L_{s2}/G_{s2}	0.66948	0.71173	0.79100	0.84107	0.92484	1.04744	1.22939
L/G	1.31767	1.41931	1.57782	1.69251	1.84845	2.06823	2.39705

Continued.....

TABLE 9. (CONTINUED)

Three Stage System A

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.2	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.12400	0.08415	0.04718	0.01806	0.00839	0.00266	0.00039
X_2	0.40835	0.32271	0.22265	0.11508	0.02739	0.02739	0.00692
X_3	0.15376	0.10414	0.05814	0.02209	0.01020	0.00322	0.00047
Y_1	0.08168	0.08415	0.08690	0.08995	0.09161	0.09161	0.09507
Y_2	0.34138	0.32271	0.30067	0.27325	0.25655	0.23714	0.21372
Y_3	0.10573	0.10414	0.10276	0.10150	0.10100	0.10068	0.10031
L_{s1}/G_{s1}	7.40576	10.8837	19.3498	50.4032	108.225	340.136	2325.58
L_{s1}/G_{s2}	2.31620	2.8391	3.9857	7.4906	12.947	30.855	144.51
L_{s2}/G_{s2}	1.53259	2.0988	3.4033	7.7767	15.246	42.427	241.55
L/G	2.93187	3.91637	6.12855	13.2918	25.1815	67.1987	363.461

TABLE 10.

Three Stage System B

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	2.0	1.8	1.6	1.4
X_1	0.74603	0.70617	0.65314	0.61962	0.57981	0.53232	0.47458
X_2	0.37246	0.33950	0.29140	0.26205	0.23051	0.19630	0.16020
X_3	0.62704	0.56921	0.49718	0.45500	0.40754	0.35444	0.29518
Y_1	0.41521	0.40472	0.39176	0.38392	0.37490	0.36466	0.35224
Y_2	0.05167	0.06028	0.06636	0.06867	0.07126	0.07390	0.07701
Y_3	0.24654	0.23105	0.21494	0.20703	0.19875	0.19022	0.18119
L_{s1}/G_{s1}	0.78387	0.84297	0.93125	0.99428	1.07812	1.19353	1.36490
L_{s2}/G_{s1}	0.97605	1.01456	1.11669	1.20303	1.31723	1.48117	1.71805
L_{s2}/G_{s2}	2.95953	3.34749	3.81507	4.10964	4.52610	5.12058	6.06621
L/G	1.32345	1.42549	1.58428	1.69975	1.85538	2.07461	2.40252

Continued.....

TABLE 10. (CONTINUED)

Three Stage System B

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.2	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.40589	0.32280	0.22448	0.11751	0.06690	0.02831	0.00602
X_2	0.12233	0.08409	0.04798	0.01838	0.00827	0.00287	0.00042
X_3	0.23005	0.16109	0.09335	0.03774	0.01872	0.00555	0.00087
Y_1	0.33891	0.32280	0.30265	0.27672	0.25865	0.24031	0.21572
Y_2	0.08036	0.08409	0.08807	0.09090	0.09096	0.09622	0.09708
Y_3	0.17147	0.16109	0.15000	0.13998	0.13684	0.12523	0.12057
L_{s1}/G_{s1}	1.62874	2.09789	3.10645	6.15521	11.0816	26.8312	130.264
L_{s2}/G_{s1}	2.11353	2.83938	4.47168	10.1124	20.2666	50.1756	282.136
L_{s2}/G_{s2}	7.68112	10.8920	18.7195	44.4134	82.5970	326.797	1960.78
L/G	2.93559	3.91637	6.11852	13.2506	25.1718	66.7575	360.841

TABLE 11

Three Stage System C

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	2.0	1.8	1.6	1.4
X_1	0.00660	0.00310	0.00025	0.00021	0.00011	0.00015	0.00011
X_2	0.73175	0.69150	0.63651	0.59947	0.56557	0.51611	0.46208
X_3	0.46481	0.41280	0.35115	0.31626	0.27827	0.23716	0.19309
Y_1	0.0000	0.00000	0.00000	0.00000	0.00000	0.00000	0.00000
Y_2	0.39183	0.38323	0.37014	0.35936	0.35849	0.34705	0.33931
Y_3	0.10042	0.10021	0.10002	0.10002	0.10001	0.10002	0.10001
L_{s2}/G_{s1}	151.515	322.580	4000.00	4761.90	9090.90	6666.67	9090.90
L_{s1}/G_{s2}	0.83112	0.89193	0.98956	1.06868	1.13427	1.26515	1.42981
L_{s2}/G_{s2}	0.63597	0.69079	0.76979	0.82059	0.92925	1.04227	1.24001
L/G	1.46096	1.57933	1.75901	1.88894	2.06330	2.30706	2.66948

Continued.....

TABLE 11. (CONTINUED)

Three Stage System C

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.2	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.00009	0.00006	0.00003	0.00001	0.00001	0.00000	0.00000
X_2	0.39619	0.31623	0.22045	0.11623	0.06718	0.02823	0.00605
X_3	0.14680	0.10001	0.05624	0.02155	0.01000	0.00317	0.00046
Y_1	0.00001	0.00006	0.00027	0.00122	0.00241	0.00526	0.01434
Y_2	0.32922	0.31623	0.29830	0.27492	0.25920	0.24005	0.21604
Y_3	0.10001	0.10001	0.10001	0.10001	0.10001	0.10008	0.10003
L_{s2}/G_{s1}	11111.1	16666.7	33333.3	100000.	100000.	-------*	-------*
L_{s1}/G_{s2}	1.69307	2.16225	3.18299	6.23830	11.0265	26.9179	129.533
L_{s2}/G_{s2}	1.56232	2.16314	3.52771	8.12215	15.9261	44.1891	250.000
L/G	3.25493	4.32483	6.70987	14.3586	26.9493	71.1088	379.556

*These figures exceeded the output format

TABLE 12.

Three Stage System D

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	2.0	1.8	1.6	1.4
X_1	0.61105	0.60231	0.56919	0.59562	0.54669	0.51560	0.44863
X_2	0.39116	0.36421	0.31416	0.31438	0.26846	0.23707	0.19265
X_3	0.69824	0.66340	0.61587	0.59830	0.59994	0.51570	0.44924
Y_1	0.21118	0.26763	0.28945	0.35477	0.33724	0.34650	0.32557
Y_2	0.05985	0.07236	0.07829	0.09884	0.09375	0.09995	0.09970
Y_3	0.34043	0.34405	0.34426	0.35796	0.35209	0.34661	0.32619
L_{s1}/G_{s1}	1.26315	1.21593	1.24836	1.08329	1.21232	1.26745	1.50330
L_{s2}/G_{s1}	0.43026	0.53614	0.67212	0.81408	0.90698	1.03997	1.17248
L_{s2}/G_{s2}	7.56428	10.7371	14.0470	239.808	48.9017	6700.00	1114.11
L/G	1.45109	1.57384	1.76374	1.88884	2.06803	2.30699	2.67217

Continued.....

TABLE 12. (CONTINUED)

Three Stage System D

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.2	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.39590	0.31603	0.22096	0.11622	0.06712	0.02827	0.00604
X_2	0.14675	0.09998	0.05623	0.02154	0.01000	0.00317	0.00046
X_3	0.39590	0.31606	0.22097	0.11623	0.06712	0.02827	0.00604
Y_1	0.32893	0.31603	0.29884	0.27490	0.25907	0.24019	0.21595
Y_2	0.09997	0.09998	0.09999	0.09999	0.09999	0.10006	0.10001
Y_3	0.32898	0.31606	0.29886	0.27491	0.25908	0.24019	0.21595
L_{s1}/G_{s1}	1.69505	2.16422	3.17228	6.23883	11.0393	26.8744	129.776
L_{s2}/G_{s1}	1.56018	2.16094	3.53669	8.12033	15.9103	44.2477	249.700
L_{s2}/G_{s2}	14000.0	25000.0	50000.0	89314.8	188745.	-------*	-------*
L/G	3.25486	4.32475	6.70954	14.3582	26.9481	71.1232	379.465

*These figures exceeded the output format.

TABLE 13.

Combination of Three Stage Systems B and D

Streams from Stages 1 and 2 are joined for stage 3

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$, $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.0
X_1	0.32256
X_2	0.08403
X_3	0.16118
X_4	0.00010
Y_1	0.32256
Y_2	0.08403
Y_3	0.16118
L_{s1}/G_{s1}	2.10018
L_{s2}/G_{s1}	2.83849
L_{s2}/G_{s2}	10.87547
L/G	3.91651

TABLE 14.

Four Stage Crossflow - Algorithm Solution

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$ $X_{03} = 0.0$ $X_{04} = 0.0$

Final $Y_F = 0.1$

n	X_1	X_2	X_3	X_4	L/G
3.0	0.86205	0.72624	0.59326	0.46416	1.29968
2.8	0.85164	0.70807	0.57021	0.43940	1.33668
2.6	0.83957	0.68738	0.54453	0.41246	1.38110
2.4	0.82538	0.66362	0.51578	0.38312	1.43535
2.2	0.80851	0.63612	0.48344	0.35112	1.50309
2.0	0.78814	0.60396	0.44690	0.31623	1.58987
1.8	0.76306	0.56592	0.40543	0.27826	1.70471
1.6	0.73154	0.52044	0.35828	0.23714	1.86336
1.5	0.71258	0.49428	0.33234	0.21544	1.96740
1.4	0.69089	0.46544	0.30471	0.19307	2.09519
1.3	0.66588	0.43356	0.27537	0.17013	2.25549
1.2	0.63682	0.39830	0.24433	0.14678	2.46174
1.1	0.60272	0.35928	0.21172	0.12328	2.73546
1.0	0.56234	0.31623	0.17783	0.10000	3.11312
0.9	0.51412	0.26903	0.14317	0.07743	3.66094
0.8	0.45611	0.21798	0.10864	0.05623	4.50997
0.7	0.38622	0.16422	0.07564	0.03728	5.95130
0.6	0.30289	0.11031	0.04618	0.02154	8.74140
0.5	0.20729	0.06110	0.02277	0.01000	15.35217
0.4	0.10863	0.02374	0.00768	0.00316	37.38514
0.3	0.03126	0.00441	0.00120	0.00046	180.26419

TABLE 15.

Four Stage Countercurrent

Freundlich Isotherm Used

Initial $Y_0 = 1.0$ $X_0 = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	1.8	1.4	1.2
X_1	0.98629	0.97751	0.96076	0.92622	0.84888	0.77679
X_2	0.94184	0.91516	0.87078	0.79361	0.65555	0.55110
X_3	0.80597	0.75390	0.68060	0.57592	0.42789	0.33591
X_4	0.46416	0.41246	0.35112	0.27826	0.19307	0.14678
Y_1	0.95943	0.94259	0.91571	0.87114	0.79503	0.73852
Y_2	0.83546	0.79412	0.73756	0.65961	0.55366	0.48919
Y_3	0.52355	0.47976	0.42891	0.37038	0.30470	0.27006
Y_4	0.10000	0.10000	0.10000	0.10000	0.10000	0.10000
L/G	0.91249	0.92209	0.93675	0.97169	1.06023	1.15862

Continued.....

TABLE 15. (CONTINUED)

Four Stage Countercurrent

Freundlich Isotherm Used

Initial $Y_0 = 1.0$ $X_0 = 0.0$

Final $Y_F = 0.1$

n	1.0	0.8	0.6	0.5	0.4	0.3
X_1	0.66549	0.49920	0.27753	0.16243	0.06774	0.01403
X_2	0.41814	0.26270	0.11207	0.05468	0.01811	0.00278
X_3	0.23524	0.13491	0.05210	0.02415	0.00760	0.00111
X_4	0.10000	0.05623	0.02154	0.01000	0.00316	0.00046
Y_1	0.66549	0.57362	0.46343	0.40301	0.34066	0.27805
Y_2	0.41814	0.34322	0.26896	0.23384	0.20100	0.17100
Y_3	0.23524	0.20138	0.16987	0.15541	0.14202	0.12978
Y_4	0.10000	0.10000	0.10000	0.10000	0.10000	0.10000
L/G	1.35239	1.80287	3.24293	5.54132	13.28698	64.14546

TABLE 16.

Four Stage System A

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	2.0	1.8	1.6	1.4	1.2
X_1	0.70837	0.66392	0.60803	0.55283	0.45611
X_2	0.31632	0.27833	0.23695	0.19446	0.14689
X_3	0.70836	0.66262	0.60792	0.52706	0.45586
X_4	0.31613	0.27819	0.23733	0.19163	0.14667
Y_1	0.50178	0.47841	0.45111	0.43614	0.38984
Y_2	0.10006	0.10005	0.09987	0.10101	0.10009
Y_3	0.50178	0.47673	0.45097	0.40795	0.38958
Y_4	0.09994	0.09995	0.10013	0.09896	0.09991
L_{s2}/G_{s1}	1.27019	1.35222	1.48069	1.56017	1.97182
L_{s1}/G_{s1}	1.26998	1.35939	1.48234	1.72339	1.97256
L_{s1}/G_{s2}	1.27084	1.36165	1.47998	1.78007	1.97566
L_{s2}/G_{s2}	1.27112	1.35440	1.47828	1.61243	1.97498
L/G	1.27053	1.35692	1.48033	1.66881	1.97375

Continued.,...

TABLE 16. (CONTINUED)

Four Stage System A

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	1.0*	1.0*	1.0*	1.0*	0.8	0.6
X_1	0.35320	0.35462	0.35394	0.35421	0.23551	0.11372
X_2	0.09987	0.09999	0.09999	0.10001	0.05618	0.02164
X_3	0.35508	0.35366	0.35434	0.35407	0.23592	0.11319
X_4	0.10013	0.10001	0.10001	0.09999	0.05628	0.02145
Y_1	0.35320	0.35462	0.35394	0.35421	0.31449	0.27133
Y_2	0.09987	0.09999	0.09999	0.10001	0.09994	0.10028
Y_3	0.35508	0.35366	0.35434	0.35407	0.31493	0.27058
Y_4	0.10013	0.10001	0.10001	0.09999	0.10006	0.09975
L_{s2}/G_{s1}	2.55581	2.53478	2.54404	2.54028	3.82467	7.89764
L_{s1}/G_{s1}	2.53659	2.54655	2.53975	2.54175	3.81810	7.90263
L_{s1}/G_{s2}	2.52702	2.54796	2.53847	2.54243	3.81170	7.96749
L_{s2}/G_{s2}	2.54619	2.53625	2.54325	2.54105	3.81825	7.96241
L/G	2.54139	2.54138	2.54138	2.54138	3.81816	7.93237

*Test solutions for Conjugate Direction method.

TABLE 17.

Four Stage System B

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	1.8	1.4	1.0
X_1	0.63657	0.59419	0.54007	0.46779	0.37104	0.23831
X_2	0.31504	0.26937	0.22369	0.17184	0.11634	0.05702
X_3	0.83758	0.80807	0.76300	0.69557	0.58875	0.41899
X_4	0.60456	0.54536	0.47243	0.38175	0.27059	0.14324
Y_1	0.25795	0.25835	0.25786	0.25474	0.24957	0.23831
Y_2	0.03127	0.03303	0.03709	0.04200	0.04921	0.05702
Y_3	0.58760	0.57460	0.55151	0.52025	0.47633	0.41890
Y_4	0.22097	0.20672	0.19211	0.17668	0.16041	0.14324
L_{s1}/G_{s1}	1.16570	1.24815	1.37415	1.59314	2.02249	3.19622
L_{s2}/G_{s1}	0.71953	0.83648	0.98695	1.23805	1.72217	3.17941
L_{s1}/G_{s2}	2.05161	1.98898	2.01178	2.10619	2.40534	3.21569
L_{s2}/G_{s2}	1.26635	1.33294	1.44494	1.63674	2.04817	3.19714
L/G	1.20217	1.28085	1.40288	1.61192	2.03422	3.19733

Continued.....

TABLE 17. (CONTINUED)

Four Stage System B

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	0.8	0.6	0.5	0.4	0.3
X_1	0.16209	0.08062	0.04655	0.01819	0.00377
X_2	0.03401	0.01248	0.00584	0.00172	0.00025
X_3	0.29073	0.15712	0.09327	0.03924	0.00873
X_4	0.08120	0.03138	0.01460	0.00463	0.00068
Y_1	0.23324	0.22073	0.21576	0.20135	0.18741
Y_2	0.06688	0.07208	0.07640	0.07847	0.08249
Y_3	0.37222	0.32941	0.30541	0.27523	0.24113
Y_4	0.13416	0.12532	0.12082	0.11651	0.11215
L_{s1}/G_{s1}	4.73059	9.66650	16.8471	43.8981	215.715
L_{s2}/G_{s1}	4.88615	11.9075	23.8748	71.2250	420.906
L_{s1}/G_{s2}	4.87971	8.76577	14.8659	33.6360	153.032
L_{s2}/G_{s2}	5.04489	10.7979	21.0671	54.5553	298.598
L/G	4.88510	10.2599	19.0890	49.9436	264.201

TABLE 18.

Four Stage System C

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	1.8	1.4	1.0
X_1	0.44077	0.46250	0.54338	0.48971	0.38599	0.25371
X_2	0.73662	0.68899	0.74031	0.68228	0.56404	0.39242
X_3	0.45652	0.42968	0.39185	0.31623	0.22307	0.11839
X_4	0.47139	0.38501	0.25769	0.19904	0.13976	0.07262
Y_1	0.08563	0.13467	0.26135	0.27663	0.26376	0.25371
Y_2	0.39970	0.37962	0.40694	0.50249	0.44858	0.39242
Y_3	0.09514	0.11122	0.12731	0.12589	0.12241	0.11839
Y_4	0.10475	0.08361	0.05063	0.05471	0.06361	0.07262
L_{s1}/G_{s1}	2.07477	1.87097	1.35936	1.47712	1.90745	2.94151
L_{s2}/G_{s2}	2.02905	2.73905	2.45736	2.58357	3.09693	4.38022
L_{s1}/G_{s1}	0.63970	0.52519	0.99913	1.28623	1.69649	2.95652
L_{s2}/G_{s2}	0.62569	0.76885	1.80619	2.24971	2.75444	4.40622
L/G	1.34207	1.42369	1.51850	1.75816	2.23029	3.52885

Continued.....

TABLE 18. (CONTINUED)

Four Stage System C

Freundlich Isotherm Used.

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	0.8	0.6	0.5	0.4	0.3
X_1	0.17550	0.09228	0.05333	0.02171	0.00481
X_2	0.28037	0.15014	0.08822	0.03707	0.00809
X_3	0.06771	0.02654	0.01247	0.00399	0.00060
X_4	0.04072	0.01517	0.00700	0.00224	0.00031
Y_1	0.24885	0.23936	0.23093	0.21610	0.20162
Y_2	0.36156	0.32055	0.29702	0.26796	0.23572
Y_3	0.11601	0.11333	0.11168	0.10974	0.10785
Y_4	0.07724	0.08102	0.08369	0.08718	0.08902
L_{s1}/G_{s1}	4.28192	8.24334	14.4208	36.1141	166.113
L_{s1}/G_{s2}	6.08753	11.7426	20.1470	47.6644	232.730
L_{s2}/G_{s1}	4.91136	11.0840	21.8048	60.9384	332.405
L_{s2}/G_{s2}	6.98275	15.7903	30.4631	80.4505	465.719
L/G	5.39708	11.3557	21.1132	55.2124	290.892

TABLE 19.

Four Stage System D

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	3.0	2.6	2.2	1.8	1.4	1.0
X_1	0.59502	0.57171	0.50460	0.42470	0.29692	0.17512
X_2	0.21381	0.30613	0.25256	0.19321	0.11797	0.07211
X_3	0.62500	0.59456	0.52526	0.43907	0.31612	0.18049
X_4	0.67508	0.66699	0.60469	0.50967	0.42809	0.25342
Y_1	0.21067	0.23370	0.22207	0.21413	0.18268	0.17512
Y_2	0.00977	0.04606	0.04844	0.05186	0.05017	0.07211
Y_3	0.24414	0.25877	0.24257	0.22728	0.19943	0.18049
Y_4	0.30765	0.34891	0.33065	0.29725	0.30489	0.25342
L_{s1}/G_{s1}	1.32655	1.34035	1.54168	1.85013	2.75269	4.71031
L_{s2}/G_{s1}	0.93958	0.61295	0.68749	0.83982	1.12325	1.42853
L_{s1}/G_{s2}	2.11915	3.94535	4.26261	4.89214	5.49285	13.5759
L_{s2}/G_{s2}	1.50098	1.80424	1.90086	2.22067	2.24138	4.11760
L/G	1.39370	1.45798	1.63708	1.95181	2.58199	4.55766

Continued.....

TABLE 19. (CONTINUED)

Four Stage System D

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $Y_{02} = 1.0$ $X_{01} = 0.0$ $X_{02} = 0.0$

Final $Y_F = 0.1$

n	0.8	0.6	0.5	0.4	0.3
X_1	0.07955	0.02402	0.05211	0.00749	0.00000
X_2	0.03306	0.00674	0.00771	0.00133	0.00000
X_3	0.09234	0.03285	0.05136	0.00766	0.00046
X_4	0.26427	0.14753	0.01241	0.01213	0.00792
Y_1	0.13198	0.10676	0.22829	0.14117	0.01023
Y_2	0.06537	0.04980	0.08779	0.07071	0.00302
Y_3	0.14869	0.12881	0.22664	0.14246	0.10002
Y_4	0.34486	0.31720	0.11140	0.17119	0.21642
L_{s1}/G_{s1}	10.9115	37.1804	14.8079	114.701	-------*
L_{s2}/G_{s1}	2.01500	8.45133	18.2289	53.0049	-------*
L_{s1}/G_{s2}	15.3437	21.3355	153.489	166.130	250.760
L_{s2}/G_{s2}	2.83349	4.84969	188.948	76.7702	128.764
L/G	7.55434	16.6378	30.1230	99.2087	379.501

*These figures exceeded the output format.

TABLE 20.

L/G For Multi-Stage Crossflow Systems

Freundlich Isotherm Used

Initial $Y_0 = 1.0$ $X_{0i} = 0.0$, $i = 1,\ldots,40$

Final $Y_F = 0.1$

Stages	n=3.0	n=2.8	n=2.6	n=2.4	n=2.2
2	1.45920	1.51302	1.57809	1.65809	1.75873
3	1.34827	1.39028	1.44085	1.50276	1.58022
4	1.29968	1.33668	1.38110	1.43535	1.50309
5	1.27248	1.30674	1.34774	1.39782	1.46022
6	1.25515	1.28763	1.32654	1.37392	1.43298
8	1.23427	1.26464	1.30099	1.34529	1.40036
10	1.22220	1.25138	1.28628	1.32875	1.38150
12	1.21433	1.24270	1.27662	1.31794	1.36922
14	1.20874	1.23661	1.26985	1.31037	1.36058
16	1.20463	1.23203	1.26485	1.30470	1.35421
18	1.20147	1.22859	1.26098	1.30037	1.34927
20	1.19893	1.22581	1.25790	1.29694	1.34535
24	1.19520	1.22164	1.25333	1.29180	1.33953
28	1.19249	1.21875	1.25007	1.28817	1.33541
32	1.19051	1.21657	1.24765	1.28544	1.33231
36	1.18895	1.21486	1.24577	1.28337	1.32994
40	1.18773	1.21350	1.24429	1.28166	1.32804

Continued.....

TABLE 20. (CONTINUED)

L/G For Multi-stage Crossflow Systems

Freundlich Isotherm Used

Initial $Y_0 = 1.0$ $X_{0i} = 0.0$, $i = 1,\ldots,40$

Final $Y_F = 0.1$

Stages	n=2.0	n=1.8	n=1.6	n=1.5	n=1.4
2	1.88885	2.06306	2.30692	2.46874	2.66930
3	1.67976	1.81199	1.99542	2.11616	2.26494
4	1.58987	1.70471	1.86336	1.96740	2.09519
5	1.54004	1.64547	1.79069	1.88569	2.00224
6	1.50840	1.60793	1.74475	1.83418	1.94370
8	1.47058	1.56307	1.69005	1.77284	1.87410
10	1.44876	1.53725	1.65855	1.73761	1.83422
12	1.43457	1.52048	1.63814	1.71473	1.80832
14	1.42458	1.50870	1.62381	1.69869	1.79019
16	1.41722	1.49996	1.61318	1.68684	1.77678
18	1.41151	1.49322	1.60501	1.67770	1.76645
20	1.40696	1.48789	1.59851	1.67045	1.75826
24	1.40024	1.47994	1.58889	1.65970	1.74609
28	1.39546	1.47433	1.58204	1.65206	1.73748
32	1.39192	1.47012	1.57696	1.64638	1.73107
36	1.38916	1.46689	1.57302	1.64198	1.72608
40	1.38697	1.46430	1.56989	1.63850	1.72215

Continued.....

TABLE 20. (CONTINUED)

L/G For Multi-stage Crossflow Systems

Freundlich Isotherm Used

Initial $Y_{0i} = 1.0$ $X_{0i} = 0.0$, $i = 1,\ldots,40$

Final $Y_F = 0.1$

Stages	n=1.3	n=1.2	n=1.1	n=1.0	n=0.9	n=0.8
2	2.92362	3.25475	3.70016	4.32456	5.24724	6.70930
3	2.45217	2.69402	3.01631	3.46331	4.11549	5.13341
4	2.25549	2.46174	2.73546	3.11312	3.66094	4.50997
5	2.14183	2.33544	2.58338	2.92445	3.41756	4.17878
6	2.08066	2.25620	2.48822	2.80679	3.26634	3.97398
8	2.00057	2.16238	2.37578	2.66816	3.08881	3.73451
10	1.95469	2.10873	2.31164	2.58926	2.98805	3.59910
12	1.92498	2.07401	2.27018	2.53834	2.92314	3.51208
14	1.90419	2.04973	2.24119	2.50276	2.87787	3.45148
16	1.88880	2.03177	2.21978	2.47652	2.84447	3.40684
18	1.87695	2.01795	2.20333	2.45634	2.81883	3.37260
20	1.86755	2.00700	2.19028	2.44037	2.79856	3.34552
24	1.85362	1.99074	2.17090	2.41666	2.76844	3.30537
28	1.84376	1.97923	2.15723	2.39990	2.74720	3.27704
32	1.83641	1.97069	2.14703	2.38745	2.73140	3.25599
36	1.83072	1.96407	2.13915	2.37782	2.71919	3.23974
40	1.82617	1.95879	2.13287	2.37016	2.70947	3.22679

Continued.....

TABLE 20. (CONTINUED)

L/G For Multi-stage Crossflow Systems

Freundlich Isotherm Used

Initial $Y_{01} = 1.0$ $X_{0i} = 0.0$, $i = 1,\ldots,40$

Final $Y_F = 0.1$

Stages	n=0.7	n=0.6	n=0.5	n=0.4	n=0.3
2	9.25827	14.35702	26.94636	71.20493	379.57169
3	6.87596	10.28372	18.46101	46.15715	229.56334
4	5.95130	8.74140	15.35217	37.38514	180.26419
5	5.46492	7.94046	13.76512	33.01127	156.48170
6	5.16593	7.45199	12.80748	30.41069	142.63016
8	4.81826	6.88790	11.71204	27.47484	127.28229
10	4.62258	6.57240	11.10458	25.86619	119.01552
12	4.49721	6.37107	10.71891	24.85265	113.86353
14	4.41007	6.23146	10.45251	24.15611	110.34996
16	4.34598	6.12901	10.25757	23.64819	107.80231
18	4.29688	6.05062	10.10862	23.26154	105.87124
20	4.25805	5.98872	9.99123	22.95736	104.35749
24	4.20058	5.89719	9.81795	22.50956	102.13763
28	4.16006	5.83276	9.69621	22.19580	100.58859
32	4.12998	5.78497	9.60601	21.96376	99.44643
36	4.10676	5.74809	9.53649	21.78520	98.56959
40	4.08829	5.71879	9.48128	21.64355	97.87524

APPENDIX II

FORTRAN IV List For Pattern Search Method

PLEASE RETURN TO THE CHEMICAL ENGINEERING BUILDING

JOB NUMBER 16096 CATEGORY F USER'S NAME- J P LUCAS USER'S

*****THE COMPUTING CENTRE WILL BE CLOSED FRI., SAT., SUN., AND MON., - APRIL 12, 13,

JOB START 15HRS 57MIN 09.4SEC V9M011 OFF-LI

```
$JOB     16096    J.P. LUCAS

$TIME             1

$FORTRAN

          COMMON X,XMAX,XMIN,SN,TOL,ALPHA,BETA,D,SC,C,DEL,N,LA,K,KK,LT,LSN,
         1NCT,AKI,NS
          DIMENSION X(50),Y(50),XMAX(50),XMIN(50),C(50),D(50),AKI(50),ALP(9)
        1 FORMAT (2I2,2E10.5,3F10.5)
        4 FORMAT (8F10.5)
          READ (5,1) N,NC,TO,RI,ALPHA,BETA,DEL
          READ (5,4) Q,Y01,Y02,YF,X01,X02
       25 READ (5,4) (X(I),I=1,N)
          READ (5,4) (XMIN(I),XMAX(I), I = 1,N)
          WRITE (6,60)
          WRITE (6,51) N,NC,TO,ALPHA,BETA,DEL
          WRITE (6,56) (X(I),XMIN(I),XMAX(I), I = 1,N)
       60 FORMAT (1H1,90H RUN NO.         DATE          DESCRIPTION
         1                                           )
       51 FORMAT (1H0,24H INDEPENDENT VARIABLES =,I2//14H CONSTRAINTS =,I2,/
         1/20H INITIAL TOLERANCE =,E12.5//,22H ACCELERATION FACTOR =,F6.3//1
         29H REDUCTION FACTOR =,F6.3//50H FRACTION OF INTERVAL USED FOR INIT
         3IAL STEP SIZE =,F6.3)
       56 FORMAT(1H0,43H  INITIAL VALUE    MIN. VALUE    MAX. VALUE//(4X,F9.
         14,7X,F8.4,6X,F8.4))
          R = RI
          RR = RI
          TOL = TO
          LSW = 1
          NS = 1
          LA = 1
          LSN = 0
          LT = 0
       11 DO 10 I = 1,N
       10 Y(I) = X(I)**Q
          ALP(1) = (X(1) - X(4))/(Y01 - Y(1))
          ALP(2) = (X(2) - X01)/(Y(1) - Y(2))
          ALP(3) = (X(3) - X(2))/(Y02 - Y(3))
          ALP(4) = (X(4) - X02)/(Y(3) - Y(4))
          AL1 = 1./(ALP(1) + ALP(4))
          CON1 = YF - (ALP(1)*Y(2) + ALP(4)*Y(4))*AL1
          CON2 = ALP(1)/ALP(2) - ALP(4)/ALP(3)
          CC = (1. + ALP(1)/ALP(2))*AL1
          SN = CC + R*CON1**2 + RR*CON2**2
          CALL STEP50
          GO TO (11,49,50), NS
       50 WRITE (6,19) CC,SN
          WRITE (6,22) (X(I), I = 1,N)
          WRITE (6,23) (Y(I), I = 1,N)
          WRITE (6,20) R,CON1,RR,CON2
          WRITE (6,21) NCT
```

```
   19 FORMAT (1H0,16H OPTIMAL - L/G =,F14.5, 6H SN = ,F14.5/)
   20 FORMAT (20H PENALTY FACTOR C1 =,E15.5,4HC1 =,E15.5,20H PENALTY FAC
     1TOR C2 =,E15.5,4HC2 =,E15.5/)
   21 FORMAT (1H0,26H RETURNS TO MAIN PROGRAM =,I5//)
   22 FORMAT (38H INDEPENDENT VARIABLES AT OPTIMUM ARE ,8F10.6/)
   23 FORMAT (38H   DEPENDENT VARIABLES AT OPTIMUM ARE ,8F10.6/)
      C1 = ABS(CON1)
      C2 = ABS(CON2)
      C3 = C2/C1
      IF (C1.GT.C2) GO TO 206
      RR = RR*10.
      IF (C3.LT.10.0) R = 10.*R/C3
      GO TO 207
  206 R = R*10.
      IF (C3.GT.0.1) RR = 10.*RR*C3
  207 IF(R.GT.1.E8.AND.RR.GT.1.E8) GO TO 49
      NS = 1
      LA = 1
      GO TO 11
   49 STOP
      END
```

```
      SUBROUTINE STEP50
      COMMON X,XMAX,XMIN,SN,TOL,ALPHA,BETA,D,SC,C,DEL,N,LA,K,KK,LT,LSN,
     1NCT,AKI,NS
      DIMENSION X(50),XMAX(50),XMIN(50),C(50),P(50),D(50),AKI(50)
      GO TO (100,280,460,580,510),LA
C  FIRST ENTRY TO SUBROUTINE
C  INITIALIZE VARIABLES
  100 SP = SN
      NCT=1
      SC = SN
      M1=1
      M2=1
      NPF=0
      K=1
      KK=1
      IF (TOL.LE.0.0) TOL = 1.E-5
      IF (ALPHA.LE.0.0) ALPHA = 1.1111111
      IF (BETA.LE.0.0) BETA = 0.1
      IF (DEL.LE.0.0) DEL = 0.1
      DO 180 I = 1,N
C  SET INDEPENDENT VARIABLES AT LAST BASE POINT
      C(I)=X(I)
C  SET INITIAL STEP SIZES
  180 D(I)=DEL*(XMAX(I)-XMIN(I))
C  SET SWITCH FOR FIRST SET OF PATTERN MOVES - PREVENTS STEP SIZE FROM
C  BECOMING TOO LARGE
  190 JSW = 1
C  JSW = 1 INITIALLY AND AFTER EACH TIME THROUGH THE LAST VARIABLE WITH
C      NO IMPROVEMENT WITHIN TOLERANCE OF LAST PATTERN MOVE OVER OLD BASE
C      POINT.
  200 LA = 2
      IF (D(K).EQ.0.0) GO TO 490
      X(K) = X(K) + D(K)
C  MAKES FIRST PATTERN MOVE
C  CHECK TO SEE IF VARIABLES ARE WITHIN BOUNDARIES
  230 IF (X(K).LE.XMAX(K).AND.XMIN(K).LE.X(K)) GO TO 270
      GO TO (500,360,480,500,500) , LA
C  ADD TO SUBROUTINE ENTRY COUNTER
  270 NCT = NCT + 1
      RETURN
C  CHECK TO SEE IF PATTERN MOVE WAS SUCCESSFUL
  280 IF (SN.GE.SP) GO TO 360
C  SUCCESSFUL PATTERN MOVE
C  IF INITIAL BASE POINT REMAINS DO NOT INCREASE STEP SIZE
      IF (JSW.EQ.2) D(K) = D(K) * ALPHA
C  SET SP TO BE BEST VALUE FOUND ON THIS SEARCH
  300 SP = SN
      NPF=0
      M2=1
      M1=1
C  GO TO NEXT VARIABLE
  305 K = K + 1
C  IF PREVIOUS VARIABLE WAS THE LAST - RETURN TO FIRST
      IF (K.GT.N) K = 1
C  LT = 1 FOR TRUNCATED SEARCH
      IF (LT.GT.0) GO TO 340
C  ADD TO COUNTER OF VARIABLES STUDIED SINCE LAST TEST FOR BASE POINT
      KK = KK + 1
      IF (KK.LE.N) GO TO 200
```

```
C  IF KK EXCEEDS THE NO. OF VARIABLES DO TOLERANCE CHECK ON LAST BASE PT
   340 IF (SP + TOL*ABS(SC).GE.SC) GO TO 400
C  IF THE OBJECTIVE FUNCTION ON THE LAST PATTERN MOVE IS GREATER THAN
C      THE OBJECTIVE FUNCTION FOR THE LAST BASE POINT OR IS LESS THAN A
C      TOLERANCE OF THE BASE POINT - CHECK TO SEE IF LAST VARIABLE HAS
C      BEEN USED - OTHERWISE SET THE CURRENT VALUES TO BE THE NEW BASE PT
       IF (JSW.EQ.2) GO TO 353
   352 LA = 5
C  DO BASE POINT CALCULATION
       M1=1
       GO TO 270
C  IF THE NUMBER OF SUCCESSIVE PATTERN MOVES FOLLOWED BY FAILURE OF
C      INDIVIDUAL MOVES EXCEEDS 5 CHECK TO SEE IF LAST VARIABLE HAS BEEN
C      USED - OTHERWISE SET CURRENT VALUES TO BE NEW BASE POINT
   353 IF(NPF-5) 352,400,400
   360 LA = 3
       X(K) = X(K) - 2.*D(K)
       GO TO 230
   400 IF (LT.GT.0) KK = KK + 1
       IF (KK.LE.N) GO TO 200
       IF (JSW.NE.2) GO TO 425
       SP = SC
C  SET THE CURRENT VALUES TO BE NEW BASE POINT
       DO 420 I=1,N
   420 X(I) = C(I)
       NPF=0
   440 KK = 1
       M1=1
       M2=1
       GO TO 190
   425 JSW = 2
       IF (M1.LE.N) GO TO 440
C  DURING THE SECOND SWEEP THROUGH THE VARIABLES WITH NO IMPROVEMENT IN
C      THE OBJECTIVE FUNCTION THE COUNTER M1 IS TESTED TO SEE IF IT
C      EXCEEDS THE NUMBER OF VARIABLES - IF IT HAS GO TO END OF JOB AND
C      PRINT OUT LOCAL OPTIMUM  - IF NOT SET M1 = 1,KK=1 AND RETURN FOR
C      ANOTHER SWEEP THROUGH VARIABLES
       DO 1015 I=1,N
  1015 X(I) = C(I)
       NS = 3
       RETURN
   460 IF (SN.GE.SP)  GO TO 480
C  IF THE OBJECTIVE FUNCTION HAS IMPROVED OVER THE ORIGINAL PATTERN
C      POINT BY A REVERSE MOVE CHANGE THE SIGN OF THE STEP SO THAT THE
C      FIRST MOVE ON THE VARIABLE DURING THE NEXT PATTERN MOVE WILL BE IN
C      THE SAME DIRECTION AS THE CURRENT SUCCESS
       D(K) = - D(K)
C  THE STEP SIZE IS NOT INCREASED ON A REVERSE SUCCESS - ONLY WHEN THE
C      FIRST MOVE SUCCEEDS IS THE STEP SIZE INCREASED BY ALPHA FACTOR
       GO TO 300
   480 X(K)=X(K)+D(K)
       D(K) = D(K)*BETA
C  PREVENTS INTERVALS FROM BECOMING TOO SMALL
       DX =ABS(X(K)/D(K)*TOL)
C  WHEN DX RISES ABOVE UNITY THE MINIMUM STEP SIZE HAS BEEN REACHED
       IF (1.-DX) 481,482,484
   481 D(K) =D(K)*DX
   482 DX= ABS(1.E-30/D(K))
       IF (1. - DX) 485,490,490
```

```
      484 DX=ABS(1.E-30/D(K))
    C  ADDITIONAL CHECK TO PREVENT THE STEP SIZES FROM GOING BELOW E-30
          IF(1.-DX) 485,490,492
      485 D(K) =D(K)*DX
    C  AS SOON AS THE STEP SIZE BECOMES MINIMUM THE COUNTER M1 BEGINS TO
    C     FUNCTION
      490 M1 = M1 + 1
      492 IF (JSW.EQ.1) GO TO 305
      493 M2 = M2 + 1
          IF (M2.LE.N) GO TO 305
          M2 = 1
          NPF=NPF+1
          GO TO 305
      500 WRITE(6,1001)
     1001 FORMAT (52H  PROGRAM HAS CRAPPED OUT - RESET STARTING VARIABLES)
          NS = 2
          RETURN
      510 KK = 1
          IF (LSN.GT.0) SP = SN
      530 SC = SP
    C  MAKES PATTERN MOVE JUDGING FROM LAST RUN THRU ALL VARIABLES
          LA=4
          DO 570 I=1,N
    C  STORE LAST BASE POINT VARIABLES IN P ARRAY
          P(I)=C(I)
    C  SETS VARIABLES FROM LAST PATTERN MOVE TO BE NEW BASE POINT
          C(I)=X(I)
    C  NEW INDEPENDENT VARAIBLES FORMED FOR NEXT PATTERN MOVE BY INCREASING
    C     THE VARIABLES BY AN EQUAL AMOUNT IN THE DIRECTION OF THEIR LAST
    C     SUCCESS
          X(I) =2.*X(I)-P(I)
    C  CHECK TO SEE THAT VARIABLES REMAIN WITHIN BOUNDS
          IF(X(I)-XMAX(I)) 550,570,540
      540 X(I)=XMAX(I)
          GO TO 570
      550 IF (XMIN(I).GT.X(I)) X(I) = XMIN(I)
      570 CONTINUE
          GO TO 270
      580 SP = SN
          JSW = 2
          GO TO 200
          END
    $ENTRY
```

APPENDIX III

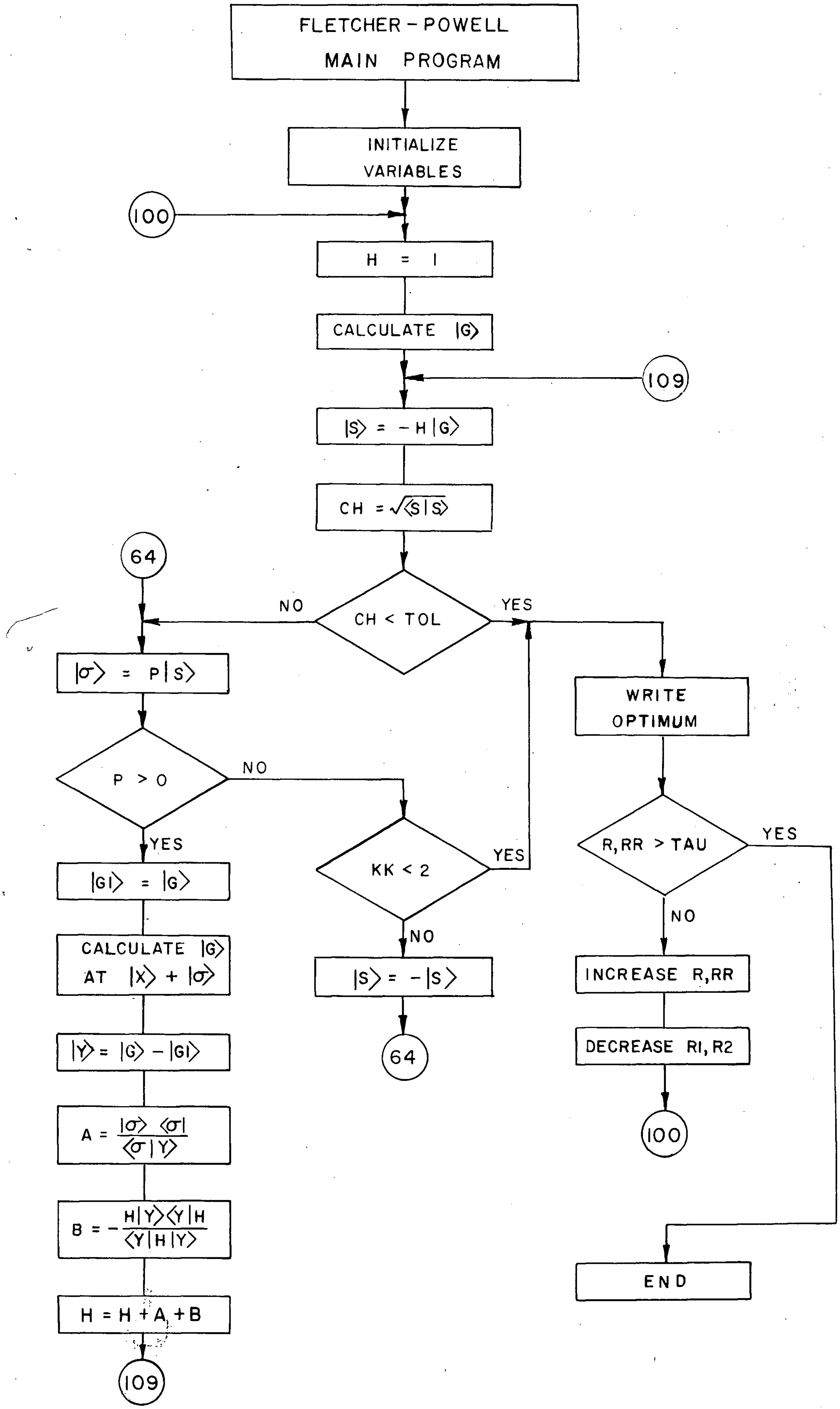
Deflected Gradient Optimization Method (Fletcher-Powell)

1. Nomenclature
2. Flow Diagram
3. FORTRAN IV List

NOMENCLATURE

Dirac bra - ket notation is used as applied to real vectors.

$\lvert x\rangle$	Column vector
$\langle x\rvert$	Row vector
$\langle x\lvert x\rangle$	Scalar product
$\lvert x\rangle\langle x\rvert$	Matrix operator
$\langle x\lvert M\rvert x\rangle$	Quadratic form, M matrix of coefficients
I	Identity matrix
H	Matrix determining direction of search
$\lvert G\rangle$	Gradient vector determined before search for minima
$\lvert S\rangle$	Direction of search for minima
CH	Magnitude of $\lvert S\rangle$
$\lvert \sigma\rangle$	Vector between starting point and minima
P	Step along $\lvert S\rangle$ to minima
$\lvert GI\rangle$	Gradient vector determined after minima found
$\lvert Y\rangle$	Gradient difference vector
A	Matrix computed to make an improvement on H
B	Matrix computed to correct initial guess of H
K	KK = 1 move in positive $\lvert S\rangle$ direction
	KK = 2 move in negative $\lvert S\rangle$ direction
R,RR	Penalty factors on equality constraints
R1,R2	Penalty factors on inequality constraints
TOL	Minimum magnitude of $\lvert S\rangle$
TAU	Maximum final value for equality penalty factors


FLETCHER - POWELL
MAIN PROGRAM
INITIALIZE
VARIABLES
100
H = I
CALCULATE |G>
109
|S> = − H |G>
CH = √<S|S>
64
NO
CH < TOL
YES
|σ> = P|S>
WRITE
OPTIMUM
P > 0
NO
YES
KK < 2
YES
R,RR > TAU
YES
NO
|GI> = |G>
CALCULATE |G>
AT |X> + |σ>
NO
|S> = − |S>
INCREASE R,RR
DECREASE RI, R2
|Y> = |G> − |GI>
64
100
A = |σ> <σ| / <σ|Y>
B = − H|Y><Y|H / <Y|H|Y>
END
H = H + A + B
109

PLEASE RETURN TO THE CHEMICAL ENGINEERING BUILDING

3.4

JOB NUMBER 16096 CATEGORY F USER'S NAME- J P LUCAS USER'S

*****THE COMPUTING CENTRE WILL BE CLOSED FRI., SAT., SUN., AND MON., - APRIL 12, 13,

JOB START 15HRS 13MIN 49.3SEC V9M011 OFF-LI

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$JOB    16096    J.P.LUCAS

$TIME            1

$FORTRAN

      C      DESCENT METHOD FOR MINIMIZATION
             COMMON G,R,R1,R2,RR,W,YF,X01,X02,ALP,S,SO,JJ,Q,Y01,Y02,P,CON1,CON2
            1,CC,Z,KCT
             DIMENSION H(20,20),A(20,20),B(20,20),X(20),S(20),SIG(20),Y(20),G(2
            10),GI(20),YT1(20),YT2(20),ALP(20),W(50),SO(20),Z(20)
             READ (5,1) MM,N,TOL,RI
           1 FORMAT (2I2,2E10.5)
             JJ= 1
             DO 400 M = 1,MM
             READ (5,2) (X(I), I = 1,N)
             READ (5,2) X01,X02,Y01,Y02,YF,Q
             READ (5,2) (W(I),I=1,13),R1,R2
             WRITE (6,3)
             WRITE (6,4) X01,X02
             WRITE (6,5) Y01,Y02
             WRITE (6,6) YF
             WRITE (6,7) Q
           2 FORMAT (8F10.5)
           3 FORMAT (1H1,20H FOUR STAGE SYSTEM A//)
           4 FORMAT (48H INITIAL CONCENTRATION IN ADSORBENT - STREAM 1 =,F12.5,
            113H , STREAM 2 =,F12.5/)
           5 FORMAT (47H INITIAL CONCENTRATION IN SOLUTION - STREAM 1 =,F12.5,1
            13H , STREAM 2 =,F12.5/)
           6 FORMAT (34H FINAL CONCENTRATION IN SOLUTION =,F12.5/)
           7 FORMAT (37H VALUE OF N FOR FREUNDLICH ISOTHERM =,F12.5//)
             R = RI
             RR = RI
             KCT = 1
             KK = 1
         100 WRITE (6,101) (X(I), I = 1,N)
         101 FORMAT (12H VARIABLES =,10F10.5)
             DO 105 J = 1,N
             DO 105 I = 1,N
         105 H(I,J) = 0.0
             DO 106 I = 1,N
         106 H(I,I) = 1.0
             CALL GRAD (X,N)
         109 DO 110 I = 1,N
             S(I) = 0.0
             DO 110 J = 1,N
         110 S(I) = S(I) - H(I,J)*G(J)
             CHK = 0.0
             DO 111 I = 1,N
         111 CHK = CHK + S(I)**2
             CH = SQRT(CHK)
             IF (CH.LT.TOL) GO TO 200
```

```
   64 CALL PARMIN(X,N)
      GO TO (65,210),KCT
   65 KK = 1
      DO 115 I = 1,N
  115 SIG(I) = P*SO(I)
      DO 107 I = 1,N
  107 GI(I) = G(I)
      CALL GRAD (X,N)
  120 DO 121 I = 1,N
  121 Y(I) = G(I) - GI(I)
      DEN1 = 0.0
      DO 125 I = 1,N
  125 DEN1 = DEN1 + SIG(I)*Y(I)
      DO 130 I = 1,N
      DO 130 J = 1,N
  130 A(I,J) = SIG(I)*SIG(J)/DEN1
      DO 135 J = 1,N
      YT1(J) = 0.0
      DO 135 I = 1,N
  135 YT1(J) = Y(I)*H(I,J) + YT1(J)
      DEN2 = 0.0
      DO 140 I = 1,N
  140 DEN2 = DEN2 + YT1(I)*Y(I)
      DO 145 I = 1,N
      YT2(I) = 0.0
      DO 145 J = 1,N
  145 YT2(I) = YT2(I) + H(I,J)*Y(J)
      DO 150 I = 1,N
      DO 150 J = 1,N
  150 B(I,J) =-YT2(I)*YT1(J)/DEN2
      DO 155 I = 1,N
      DO 155 J = 1,N
  155 H(I,J) = H(I,J) + A(I,J) + B(I,J)
      GO TO 109
  200 WRITE (6,201) (X(I),I=1,N)
      WRITE (6,202) (Z(I),I=1,N)
      WRITE (6,203) CC
      WRITE (6,303) (ALP(I), I = 1,N)
      WRITE (6,305) R,RR
      WRITE (6,306) CON1,CON2
  201 FORMAT (1H0,27H VARIABLES AT OPTIMUM ARE =,10F10.5)
  202 FORMAT (35H DEPENDENT VARIABLES AT OPTIMUM ARE,8F12.5)
  203 FORMAT (1H0,24H OBJECTIVE FUNCTION IS   ,F12.5)
  303 FORMAT (1H0,9H ALPHAS =,10F12.5)
  305 FORMAT(1H0,33H PENALTY FACTORS - CONSTRAINT 1 =,E15.5,15H CONSTRAI
     1NT 2 =,E15.5/)
  306 FORMAT (25H CONSTRAINT VALUES - C1 =,E15.5,5H C2 =,E15.5/)
      DO 310 I = 1,N
  310 ALP(I) = 1./ALP(I)
      WRITE (6,311)(ALP(I) , I = 1,N)
  311 FORMAT (1H0, 6H L/G =,10F12.5/)
      C1 = ABS(CON1)
      C2 = ABS(CON2)
      C3 = C2/C1
      IF (C1.GT.C2) GO TO 206
      RR = RR*10.
      IF (C3.LT.10.0) R = 10.*R/C3
      GO TO 207
  206 R = R*10.
```

```
      IF (C3.GT.0.1) RR = 10.*RR*C3
  207 IF (R.LT.1.E9.AND.RR.LT.1.E9)  GO TO 100
      GO TO 400
  210 IF (KK.GT.2) GO TO 200
      KK = KK + 1
      DO 211 I = 1,N
  211 S(I) = -S(I)
      WRITE (6,212)
  212 FORMAT (1H0,32H REVERSE DIRECTIONS ON PARAMETER/)
      KCT = 1
      GO TO 64
  400 CONTINUE
      STOP
      END
```

```
      SUBROUTINE GRAD (X,N)
      COMMON G,R,R1,R2,RR,W,YF,X01,X02,ALP,S,SO,JJ,Q,Y01,Y02,P,CON1,CON2
     1,CC,Y,KCT
      DIMENSION G(20),ALP(20),W(50),X(20),Y(20),SO(20),S(20)
      DO 5 I = 1,N
    5 Y(I) = X(I)**Q
      ALP(1) = (X(1) - X(4))/(Y01 - Y(1))
      ALP(2) = (X(2) - X02)/(Y(1) - Y(2))
      ALP(3) = (X(3) - X(2))/(Y02 - Y(3))
      ALP(4) = (X(4) - X01)/(Y(3) - Y(4))
      AL1 = 1./(ALP(1) + ALP(4))
      AL2 = AL1**2
      CON1 = YF - (ALP(1)*Y(2) + ALP(4)*Y(4))*AL1
      CON2 = ALP(2)/ALP(1) - ALP(3)/ALP(4)
      DFDA1 = AL2*(ALP(4)/ALP(2) - 1. + 2.*R*CON1*ALP(4)*(Y(4) - Y(2)))-
     1(2.*RR*CON2*ALP(2) + R2)/ALP(1)**2
      DFDA2 =-(ALP(1)*AL1 + R2)/ALP(2)**2 + 2.*RR*CON2/ALP(1)
      DFDA3 = -2.*RR*CON2/ALP(4) - R2/ALP(3)**2
      DFDA4 = -AL2*(1. + ALP(1)/ALP(2) + 2.*R*CON1*ALP(1)*(Y(4) - Y(2)))
     1 + (2.*RR*CON2*ALP(3) - R2)/ALP(4)**2
      DFDY2 = -2.*R*CON1*ALP(1)*AL1
      DFDY4 = DFDY2*ALP(4)/ALP(1)
      DA1DX1 = 1./(Y01 - Y(1))
      DA1DX4 = -DA1DX1
      DA2DX2 = 1./(Y(1) - Y(2))
      DA3DX2 = -1./(Y02 - Y(3))
      DA3DX3 = -DA3DX2
      DA4DX4 = 1./(Y(3) - Y(4))
      DA1DY1 = ALP(1)*DA1DX1
      DA2DY1 = -ALP(2)*DA2DX2
      DA2DY2 = -DA2DY1
      DA3DY3 = ALP(3)*DA3DX3
      DA4DY3 = -ALP(4)*DA4DX4
      DA4DY4 = -DA4DY3
      DO 10 J = 1,N
      J2 = 2*J
   10 G(J) = R1*(W(J2)/(1. - X(J))**2 - W(J2-1)/X(J)**2)
      DX = Q*X(1)**(Q - 1.)
      G(1) = G(1) + DFDA1*(DA1DX1 + DA1DY1*DX) + DFDA2*DA2DY1*DX
      DX = Q*X(2)**(Q - 1.)
      G(2) = G(2) + DFDA2*(DA2DX2 + DA2DY2*DX) + DFDA3*DA3DX2 + DFDY2*DX
      DX = Q*X(3)**(Q - 1.)
      G(3) = G(3) + DFDA3*(DA3DX3 + DA3DY3*DX) + DFDA4*DA4DY3*DX
      DX = Q*X(4)**(Q - 1.)
      G(4) = G(4) + DFDA1*DA1DX4 + DFDA4*(DA4DX4 + DA4DY4*DX) + DFDY4*DX
      RETURN
      END
```

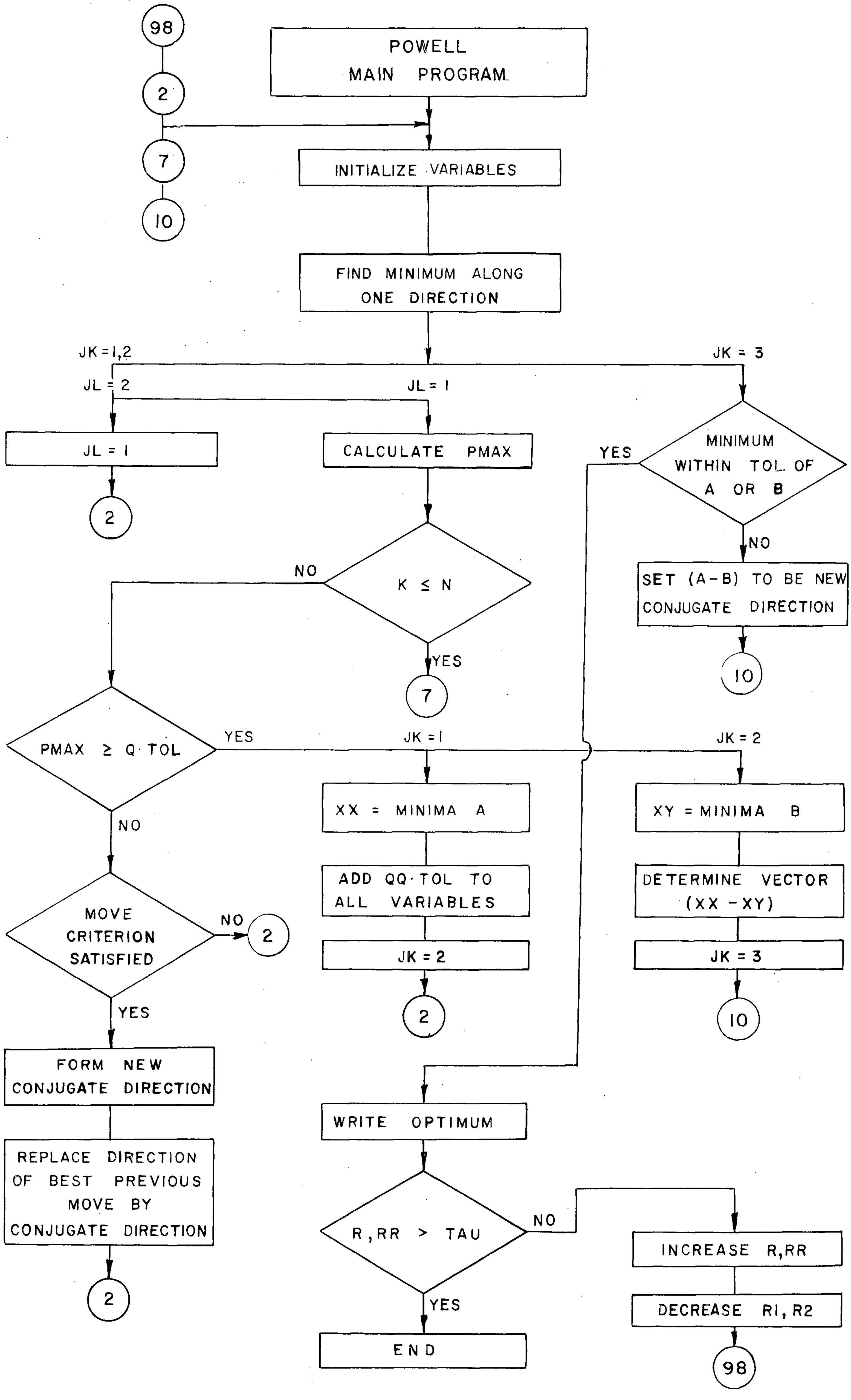
APPENDIX IV

Conjugate Direction Method (Powell)

1. Nomenclature
2. Flow Diagram
3. FORTRAN IV List

NOMENCLATURE

JK	JK = 1 Determine initial minima
	JK = 2 Determine second minima
	JK = 3 Determine possible minima between previous pair
JL	JL = 1 Normal variable minimization
	JL = 2 Minimize along conjugate direction
PMAX	Maximum distance moved by any variable in a set of variable moves
K	Variable index
N	Number of variables
Q	Factor on minimum step size TOL (< 1)
TOL	Minimum step size
XX	Coordinates of initial minima
XY	Coordinates of second minima
QQ	Factor on minimum step size (> 1)
R,RR	Penalty factors on equality constraints
R1,R2	Penalty factors on inequality constraints
TAU	Maximum penalty factor on equality constraints



98
2
7
10
POWELL
MAIN PROGRAM
INITIALIZE VARIABLES
FIND MINIMUM ALONG ONE DIRECTION
JK = 1,2
JK = 3
JL = 2
JL = 1
JL = 1
2
CALCULATE PMAX
YES
MINIMUM WITHIN TOL. OF A OR B
NO
NO
K ≤ N
YES
7
SET (A−B) TO BE NEW CONJUGATE DIRECTION
10
PMAX ≥ Q·TOL
YES
JK = 1
JK = 2
XX = MINIMA A
XY = MINIMA B
NO
ADD QQ·TOL TO ALL VARIABLES
DETERMINE VECTOR (XX − XY)
MOVE CRITERION SATISFIED
NO
2
JK = 2
JK = 3
2
10
YES
FORM NEW CONJUGATE DIRECTION
WRITE OPTIMUM
REPLACE DIRECTION OF BEST PREVIOUS MOVE BY CONJUGATE DIRECTION
R, RR > TAU
NO
INCREASE R, RR
DECREASE R1, R2
YES
2
END
98

```
ISN      SOURCE STATEMENT

  0 ‡ $IBFTC POWELL                                                       ‡
    ‡ C       POWELL MINIMIZATION WITHOUT DERIVITIVES                       ‡
  1 ‡         COMMON ALP,YF,Y01,Y02,X01,X02,W,R,RR,R1,R2,Q,CON1,CON2         ‡
  2 ‡         DIMENSION XI(20,20),XO(20),X(20),XJ(20),S(20),XN(20),ALP(20),W(50) ‡
    ‡        1,XX(20),XY(20)                                                  ‡
  3 ‡         READ (5,4) N,RI                                                 ‡
  5 ‡       4 FORMAT (I2,E10.5)                                               ‡
  6 ‡         READ (5,1) (X(I),I=1,N)                                         ‡
 13 ‡         READ (5,1) X01,X02,Y01,Y02,YF,Q                                 ‡
 14 ‡         READ (5,1) (W(I), I = 1,12),R1,R2                               ‡
 21 ‡       1 FORMAT (8F10.5)                                                 ‡
 22 ‡         R = RI                                                          ‡
 23 ‡         RR = RI                                                         ‡
 24 ‡         READ (5,3) TOL,TAU,DEL                                          ‡
 25 ‡       3 FORMAT (2E10.5,F10.5)                                           ‡
 26 ‡         STL = TOL*10.                                                   ‡
 27 ‡         FAC = 5.0                                                       ‡
 30 ‡         NP1 = N + 1                                                     ‡
 31 ‡         NM1 = N - 1                                                     ‡
 32 ‡      98 JL = 1                                                          ‡
 33 ‡         JK = 1                                                          ‡
 34 ‡         FACI = 1./FAC                                                   ‡
 35 ‡         DO 5 J = 1,N                                                    ‡
 36 ‡         DO 5 I = 1,N                                                    ‡
 37 ‡       5 XI(J,I) = 0.0                                                   ‡
 42 ‡         DO 6 I = 1,N                                                    ‡
 43 ‡       6 XI(I,I) = 1.0                                                   ‡
 45 ‡         SN = F(X,N)                                                     ‡
 46 ‡         WRITE (6,11) SN,(X(I),I = 1,N)                                  ‡
 53 ‡      11 FORMAT (1H0,22H STARTING VALUES - F =,F10.5,12H VARIABLES =,8F10.5 ‡
    ‡        1/)                                                              ‡
 54 ‡       2 SO = F(X,N)                                                     ‡
 55 ‡         K = 1                                                           ‡
 56 ‡      10 DO 15 I = 1,N                                                   ‡
 57 ‡      15 XN(I) = X(I)                                                    ‡
 61 ‡         DELTA = 0.0                                                     ‡
 62 ‡         PMAX = 0.0                                                      ‡
 63 ‡       7 P = 0.0                                                         ‡
 64 ‡         DO 14 I = 1,N                                                   ‡
 65 ‡      14 XO(I) = X(I)                                                    ‡
 67 ‡         LA = 1                                                          ‡
 70 ‡         KY = 1                                                          ‡
 71 ‡         DELT = DEL                                                      ‡
 72 ‡       9 DO 8 I = 1,N                                                    ‡
 73 ‡       8 X(I) = XO(I) + P*XI(K,I)                                        ‡
 75 ‡         GO TO (100,120,140,160,180,200,240),LA                          ‡
 76 ‡     100 FX = F(X,N)                                                     ‡
 77 ‡         FF = FX                                                         ‡
100 ‡         P = DELT                                                        ‡
101 ‡         LA = 2                                                          ‡
102 ‡         GO TO 9                                                         ‡
103 ‡     120 FXP = F(X,N)                                                    ‡
104 ‡         IF (FXP.GT.FF) GO TO 125                                        ‡
107 ‡         FF = FXP                                                        ‡
110 ‡         DELT = DELT*1.618                                               ‡
```

POWELL
ISN SOURCE STATEMENT

```
          P = P + DELT
          KY = 2
          GO TO 9
      125 GO TO (130,135),KY
      130 P = -DELT
          LA = 3
          PF = DELT
          GO TO 9
      135 PO = 0.0
          PM = 0.381*P
          PD = 0.619*P
          GO TO 150
      140 FXMP = F(X,N)
          IF (FXMP.LT.FF) GO TO 155
          PO = -DELT
          PP = 2.*DELT
          PM = 0.381*PP + PO
          PD = 0.619*PP + PO
      150 LA = 5
          P = PM
          GO TO 9
      155 LA = 4
          DELT = -DELT
      156 DELT = DELT*1.618
          P = P + DELT
          GO TO 9
      160 FXP = F(X,N)
          IF (FXP.LT.FF) GO TO 156
          PO = P
          PF = 0.0
          PM = 0.619*PO
          PD = 0.381*PO
          GO TO 150
      180 FPM = F(X,N)
          P = PD
          LA = 6
          GO TO 9
      200 FPD = F(X,N)
      205 IF (FPD.GT.FPM) GO TO 210
          PO = PM
          PM = PD
          FPM = FPD
          FPO = FPM
          PD = 0.619*(PF - PO) + PO
          P = PD
          LA = 6
          GO TO 215
      210 PF = PD
          PD = PM
          FPD = FPM
          FPF = FPD
          PM = 0.381*(PF - PO) + PO
          P = PM
          LA = 7
      215 IF ((ABS(PF - PO)).GT.TAU) GO TO 9
```

ISN SOURCE STATEMENT

```
      GO TO 260
  240 FPM = F(X,N)
      GO TO 205
  260 WRITE (6,270) SN, (X(I),I = 1,N)
  270 FORMAT (23H MINIMUM ON PARAMETER =,F10.5,12H VARIABLES =,8F10.5)
      S(K) = SN
      GO TO (399,399,490),JK
  399 GO TO (13,12),JL
   12 JL = 1
      GO TO 2
   13 IF (K.EQ.1) GO TO 400
      DELT = S(K - 1) - S(K)
      GO TO 410
  400 DELT = SO - S(K)
  410 IF (DELT.LT.DELTA) GO TO 420
      DELTA = DELT
      M = K
  420 PP = ABS (X(K) - XO(K))
      K = K + 1
      IF (PMAX.LT.PP) PMAX = PP
      IF (K.LE.N) GO TO 7
      IF (PMAX.LT.TOL*FACI) GO TO 450
      DO 430 I = 1,N
  430 XJ(I) = 2.*X(I) - XN(I)
      F1 = SO
      F2 = SN
      F3 = F(XJ,N)
      FF = (F1 - 2.*F2 + F3)*(F1 - F2 - DELTA)**2
      FG = 0.5*DELTA*(F1 - F3)**2
      IF (F3.GE.F1.OR.FF.GE.FG) GO TO 2
      DO 440 I = 1,N
  440 XI(M,I) = X(I) - XN(I)
      SUM = 0.0
      DO 441 I = 1,N
  441 SUM = SUM + XI(M,I)**2
      DO 442 I = 1,N
  442 XI(M,I) = XI(M,I)/SQRT(SUM)
      IF (M.EQ.N) GO TO 82
      DO 79 I = 1,N
   79 XI(NP1,I) = XI(M,I)
      DO 80 KK = M,N
      DO 80 I = 1,N
   80 XI(KK,I) = XI(KK+1,I)
   82 K = N
      JL = 2
      GO TO 7
  450 GO TO (455,470),JK
  455 DO 460 I = 1,N
      XX(I) = X(I)
      SA = SN
  460 X(I) = X(I) + FAC*TOL
      JK = 2
      GO TO 2
  470 PMAX = 0.0
      DO 471 I = 1,N
```

```
ISN       SOURCE STATEMENT

 331 ‡          PP = ABS(XX(I) - X(I))                                          ‡
 332 ‡      471 IF (PP.GT.PMAX) PP = PMAX                                       ‡
 336 ‡          IF (PMAX.LT.TOL*FACI) GO TO 500                                 ‡
 341 ‡          DO 475 I = 1,N                                                  ‡
 342 ‡          XY(I) = X(I)                                                    ‡
 343 ‡          SB = SN                                                         ‡
 344 ‡          IF (ABS((SA-SB)/SB).LT.STL) GO TO 500                           ‡
 347 ‡      475 XI(NP1,I) = X(I) - XX(I)                                        ‡
 351 ‡          SUM = 0.0                                                       ‡
 352 ‡          DO 480 I = 1,N                                                  ‡
 353 ‡      480 SUM = SUM + XI(NP1,I)**2                                        ‡
 355 ‡          DO 481 I = 1,N                                                  ‡
 356 ‡      481 XI(NP1,I) = XI(NP1,I)/SQRT(SUM)                                 ‡
 360 ‡          JK = 3                                                          ‡
 361 ‡          GO TO 10                                                        ‡
 362 ‡      490 PMAX = 0.0                                                      ‡
 363 ‡          DO 491 I = 1,N                                                  ‡
 364 ‡          PP = ABS(XX(I) - X(I))                                          ‡
 365 ‡          IF (PP.GT.PMAX) PMAX = PP                                       ‡
 370 ‡          PP = ABS (XY(I) - X(I))                                         ‡
 371 ‡      491 IF (PP.GT.PMAX) PMAX = PP                                       ‡
 375 ‡          IF (PMAX.LT.TOL*0.25) GO TO 500                                 ‡
 400 ‡          SC = SN                                                         ‡
 401 ‡          IF (ABS((SA-SC)/SC).LT.STL.OR.ABS((SB-SC)/SC).LT.STL) GO TO 500 ‡
 404 ‡          JK = 1                                                          ‡
 405 ‡          K = 2                                                           ‡
 406 ‡          DO 492 I = 1,N                                                  ‡
 407 ‡      492 XI(1,I) = XI(NP1,I)                                             ‡
 411 ‡          GO TO 10                                                        ‡
 412 ‡      500 WRITE (6,501) SO,(X(I),I = 1,N)                                 ‡
 417 ‡      501 FORMAT(1H0,10H OPTIMUM =,F10.5,12H VARIABLES =,8F10.5/)         ‡
 420 ‡          CC = (1. + ALP(1)/ALP(2))/(ALP(1) + ALP(4))                     ‡
 421 ‡          WRITE (6,502) CC                                                ‡
 422 ‡      502 FORMAT (6H L/G =,F12.5/)                                        ‡
 423 ‡          WRITE (6,503) CON1,CON2                                         ‡
 424 ‡      503 FORMAT (4H C1=,E12.5,5H C2 =,E12.5/)                            ‡
 425 ‡          FAC = FAC*0.8                                                   ‡
 426 ‡          DO 505 I = 1,12                                                 ‡
 427 ‡      505 W(I) = W(I)*0.1                                                 ‡
 431 ‡          C1 = ABS(CON1)                                                  ‡
 432 ‡          C2 = ABS(CON2)                                                  ‡
 433 ‡          C3 = C2/C1                                                      ‡
 434 ‡          IF (C1.GT.C2) GO TO 206                                         ‡
 437 ‡          RR = RR*10.                                                     ‡
 440 ‡          IF (C3.LT.10.0) R = 10.*R/C3                                    ‡
 443 ‡          GO TO 207                                                       ‡
 444 ‡      206 R = R*10.                                                       ‡
 445 ‡          IF (C3.GT.0.1) RR = 10.*RR*C3                                   ‡
 450 ‡      207 IF (R.LT.1.E8.AND.RR.LT.1.E8) GO TO 98                          ‡
 453 ‡          STOP                                                            ‡
 454 ‡          END                                                             ‡
```

```
      FUNCTION F(X,N)
      COMMON ALP,YF,Y01,Y02,X01,X02,W,R,RR,R1,R2,Q,CON1,CON2
      DIMENSION X(20),Y(20),W(50),ALP(20)
      DO 5 I = 1,N
    5 Y(I) = X(I)**Q
      ALP(1) = (X(1) - X(4))/(Y01 - Y(1))
      ALP(2) = (X(2) - X01)/(Y(1) - Y(2))
      ALP(3) = (X(3) - X(2))/(Y02 - Y(3))
      ALP(4) = (X(4) - X02)/(Y(3) - Y(4))
      CC = (1. + ALP(1)/ALP(2))/(ALP(1) + ALP(4))
      CON1 = YF - (ALP(1)*Y(2) + ALP(4)*Y(4))/(ALP(1) + ALP(4))
      CON2 = ALP(2)/ALP(1) - ALP(3)/ALP(4)
      F  = CC + R*CON1**2 + RR*CON2**2 + R1*(W(1)/X(1) + W(2)/(1. -X(1))
     1+ W(3)/X(2) + W(4)/(1. - X(2)) + W(5)/X(3) + W(6)/(1. - X(3)) + W(
     27)/X(4) + W(8)/(1. - X(4))) + R2*(W(9)/ALP(1) + W(10)/ALP(2) + W(1
     31)/ALP(3) + W(12)/ALP(4))
      RETURN
      END
 $ENTRY
```

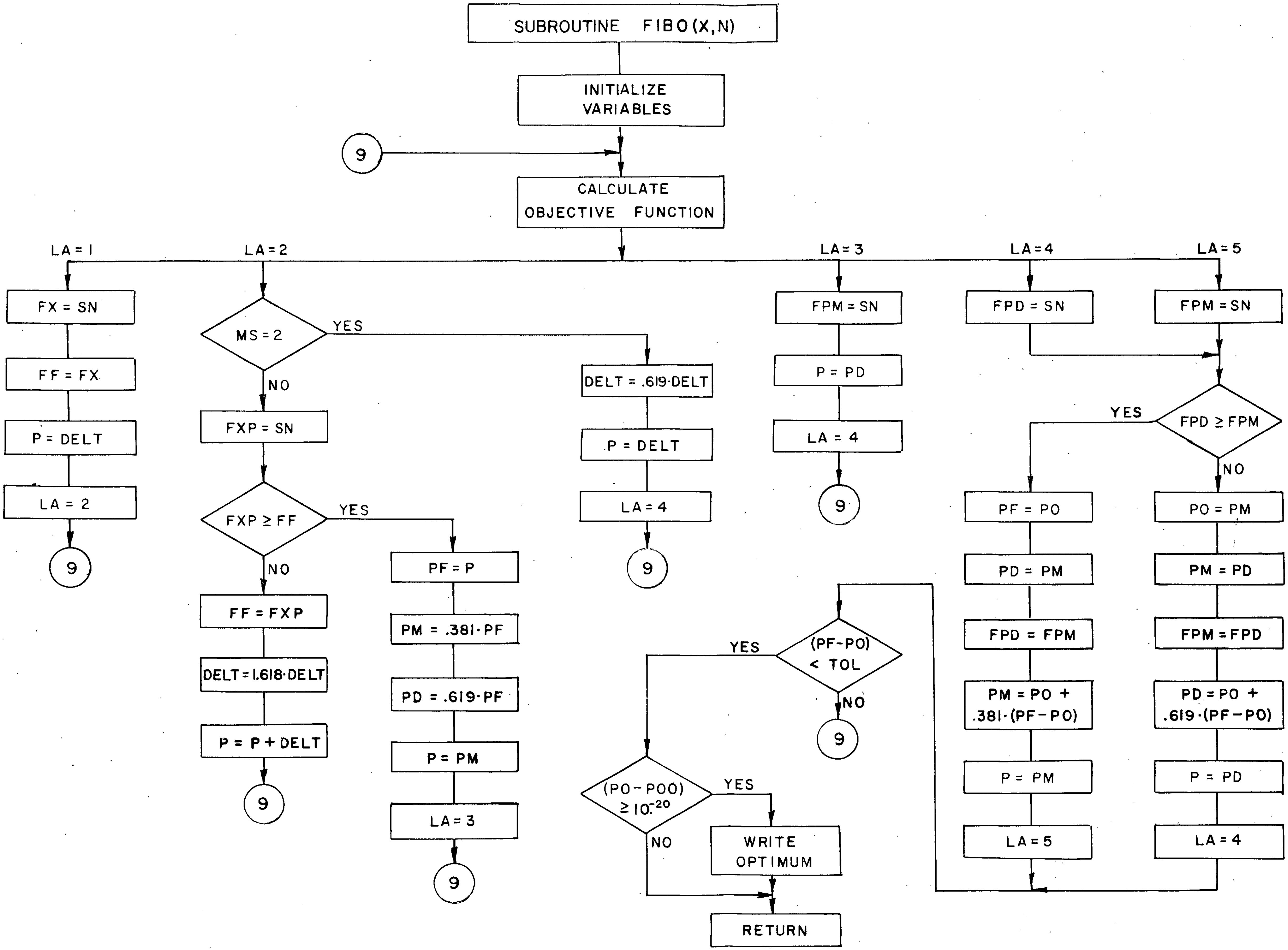
APPENDIX V.

Fibonnaci Sub-program for One-dimensional Minimization

1. Nomenclature
2. Flow Diagram
3. FORTRAN IV List

NOMENCLATURE

LA	LA = 1 Set starting point
	LA = 2 Set end point so that a minima occurs between the two points
	LA = 3 Determine objective function at points PP and PF in interval.
	LA = 4,5 Shrink interval until minimum step size is reached
SN	Current value of objective function
FX	Value of objective function at starting point
FF	Minimum value of objective function during end point search
DELT	Step size
P	Step taken from initial point
MS	MS = 1 Bounds of problem have not been exceeded
	MS = 2 Bounds of problem have been exceeded
FXP	Objective function calculated a distance P from start
PF	End point of interval for Fibonacci search
PM	Point within interval closest to initial point
PD	Point within interval closest to end point
FPM	Value of objective function at PM
FPD	Value of objective function at PD
PO	End point of interval nearest initial point
POO	Starting point of interval


SUBROUTINE FIBO(X,N)
INITIALIZE VARIABLES
9
CALCULATE OBJECTIVE FUNCTION
LA = 1
LA = 2
LA = 3
LA = 4
LA = 5
FX = SN
FF = FX
P = DELT
LA = 2
9
MS = 2
YES
NO
FXP = SN
FXP ≥ FF
YES
NO
FF = FXP
DELT = 1.618·DELT
P = P + DELT
9
PF = P
PM = .381·PF
PD = .619·PF
P = PM
LA = 3
9
DELT = .619·DELT
P = DELT
LA = 4
9
FPM = SN
P = PD
LA = 4
9
FPD = SN
FPM = SN
FPD ≥ FPM
YES
NO
PF = PO
PD = PM
FPD = FPM
PM = PO + .381·(PF − PO)
P = PM
LA = 5
PO = PM
PM = PD
FPM = FPD
PD = PO + .619·(PF − PO)
P = PD
LA = 4
(PF-PO) < TOL
YES
NO
9
(PO − POO) ≥ 10^-20
YES
NO
WRITE OPTIMUM
RETURN

```
      SUBROUTINE FIBO(X,N)
      COMMON G,R,R1,R2,RR,W,YF,X01,X02,ALP,S,SO,JJ,Q,Y01,Y02,P,CON1,CON2
     1,CC,Y,KCT
      DIMENSION G(20),ALP(20),W(50),X(20),Y(20),SO(20),XI(20),S(20)
      GO TO (2,1) , JJ
    2 JJ= 2
      READ (5,3) TOL,DEL,XMAX,XMIN,ALPMN
    3 FORMAT (E10.5,4F10.5)
    1 LA = 1
      MS = 1
      DELT = DEL
      POO = 0.0
      PO = 0.0
      P = PO
      DO 4 I = 1, N
    4 XI(I) = X(I)
      SS = 0.0
      DO 7 I = 1,N
    7 SS = SS + S(I)**2
      SS = SQRT(SS)
      DO 8 I = 1,N
    8 SO(I) = S(I)/SS
    9 DO 10 I = 1,N
   10 X(I) = XI(I) + P*SO(I)
      DO 11 I = 1,N
      IF (X(I).GT.XMIN.AND.X(I).LT.XMAX) GO TO 11
      MS = 2
      GO TO 99
   11 CONTINUE
      DO 5 I = 1,N
    5 Y(I) = X(I)**Q
      ALP(1) = (X(1) - X(4))/(Y01 - Y(1))
      ALP(2) = (X(2) - X02)/(Y(1) - Y(2))
      ALP(3) = (X(3) - X(2))/(Y02 - Y(3))
      ALP(4) = (X(4) - X01)/(Y(3) - Y(4))
      DO 14 I = 1,N
      IF (ALP(I).GT.ALPMN) GO TO 14
      MS = 2
      GO TO 99
   14 CONTINUE
      AL1 = 1./(ALP(1) + ALP(4))
      CC = (1. + ALP(1)/ALP(2))*AL1
      CON1 = YF - (ALP(1)*Y(2) + ALP(4)*Y(4))*AL1
      CON2 = ALP(2)/ALP(1) - ALP(3)/ALP(4)
      S1 = 0.0
      S2 = 0.0
      DO 15 I = 1,N
      S1 = S1 + 1./X(I) + 1./(1. - X(I))
   15 S2 = S2 + 1./ALP(I)
      SN = CC + R*CON1**2 + RR*CON2**2 + R1*S1 + R2*S2
   99 GO TO (200,250,300,350,400),LA
  200 FX = SN
      FF = FX
      P = DELT
      LA = 2
      GO TO 9
  250 IF (MS.EQ.2) GO TO 260
      FXP = SN
      IF (FXP.GE.FF) GO TO 25
```

```
      FF = FXP
      DELT = 1.618*DELT
      P = P + DELT
      GO TO 9
   25 PF = P
      PM = 0.381*PF
      PD = 0.619*PF
      P = PM
      LA = 3
      GO TO 9
  260 DELT = 0.619*DELT
      P = DELT
      MS = 1
      GO TO 9
  300 FPM = SN
      P = PD
      LA = 4
      GO TO 9
  350 FPD = SN
      GO TO 410
  400 FPM = SN
  410 IF (FPD.GT.FPM) GO TO 45
      PO = PM
      PM = PD
      FPM = FPD
      PD = 0.619*(PF - PO) + PO
      P = PD
      LA = 4
   40 IF ((PF - PO).LT.TOL) GO TO 100
      GO TO 9
   45 PF = PD
      PD = PM
      FPD = FPM
      PM = 0.381*(PF - PO) + PO
      P = PM
      LA = 5
      GO TO 40
  100 IF ((PO - POD).GT.1.E-20) GO TO 105
      RETURN
  105 WRITE (6,320) CC,SN,(X(I),I=1,N)
  320 FORMAT (6H L/G =,F10.5,6H  SN =,F14.5,12H VARIABLES =,8F10.5)
      RETURN
      END
$ENTRY
```

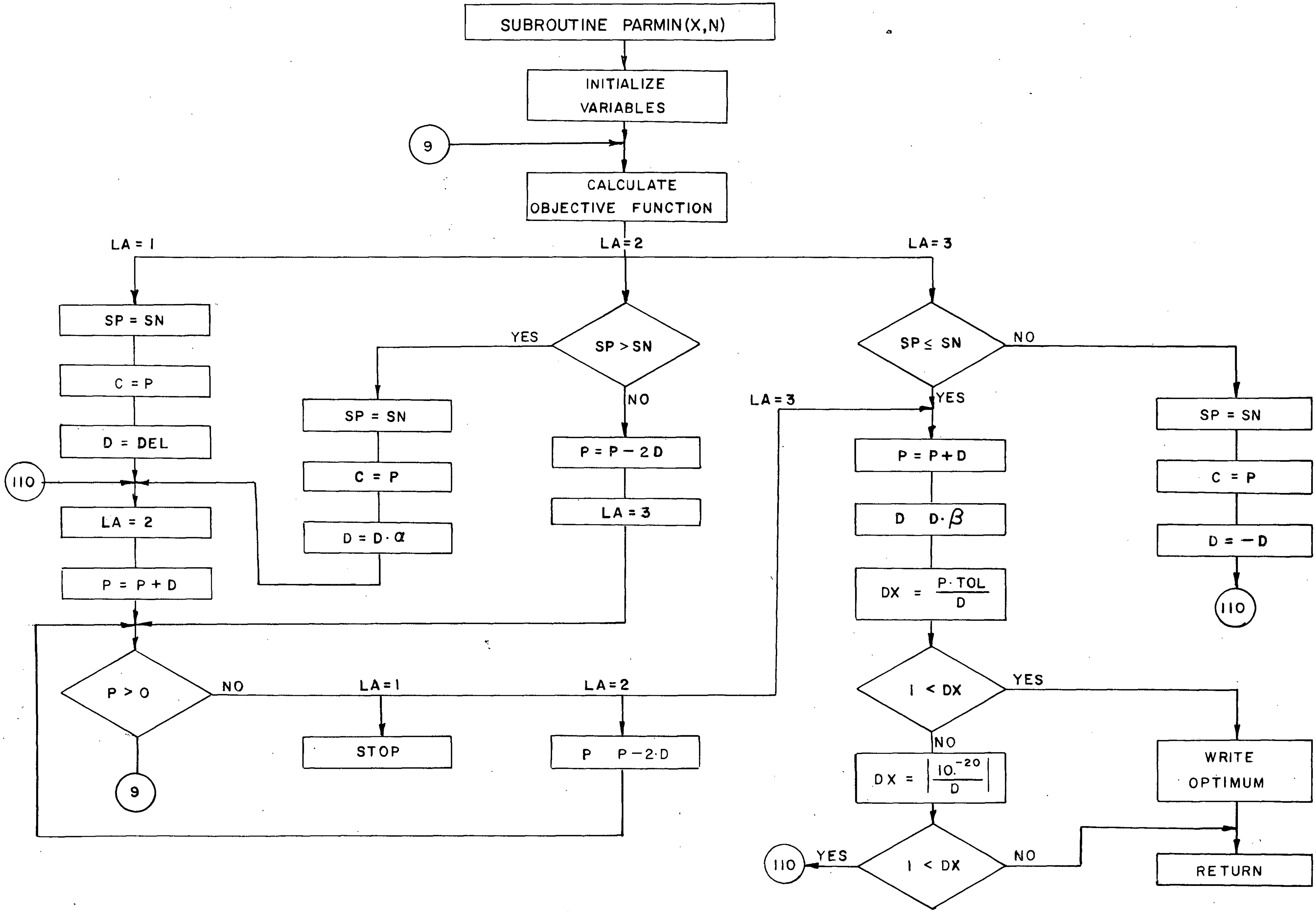
APPENDIX VI.

One Dimensional Pattern Search Sub-program

1. Nomenclature
2. Flow Diagram
3. FORTRAN IV List

NOMENCLATURE

LA	LA = 1	Make forward step.
	LA = 2	Make backward step if forward step unsuccessful. If forward step successful increase step size.
	LA = 3	Shrink step size if both forward and backward steps are failures. Return to forward step procedure if a success occurs in an immediately previous forward or backward step.
SN	Current value of objective function.	
SP	Minimum value of objective function from previous calculations.	
C	Step from initial point that corresponds to SP.	
P	Step from initial point that corresponds to SN.	
DEL	Initial step size.	
D	Step size after initialization.	
α	Step size acceleration factor.	
β	Step size deceleration factor.	
TOL	Minimum step - total move ratio [illegible]	
DX	Normalized step - total move ratio.	


SUBROUTINE PARMIN(X,N)
INITIALIZE VARIABLES
9
CALCULATE OBJECTIVE FUNCTION
LA = 1
LA = 2
LA = 3
SP = SN
C = P
D = DEL
110
LA = 2
P = P + D
SP > SN
YES
NO
SP = SN
C = P
D = D·α
P = P − 2D
LA = 3
SP ≤ SN
NO
YES
LA = 3
SP = SN
C = P
D = −D
110
P = P + D
D D·β
DX = P·TOL / D
P > 0
NO
LA = 1
LA = 2
STOP
P P − 2·D
9
I < DX
YES
NO
DX = |10.^−20 / D|
WRITE OPTIMUM
110
YES
I < DX
NO
RETURN

```
      SUBROUTINE PARMIN (X,N)
      COMMON G,R,R1,R2,RR,W,YF,X01,X02,ALP,S,SO,JJ,Q,Y01,Y02,P,CON1,CON2
     1,CC,Y,KCT
      DIMENSION G(20),ALP(20),W(50),X(20),Y(20),SO(20),XI(20),S(20)
      GO TO (2,1) , JJ
    2 JJ= 2
      READ (5,3) TOL,ALPHA,BETA,DEL,XMAX,XMIN,ALPMN
    3 FORMAT (E10.5,6F10.5)
    1 LA = 1
      DO 4 I = 1, N
    4 XI(I) = X(I)
      P = 0
      SS = 0.0
      DO 7 I = 1,N
    7 SS = SS + S(I)**2
      SS = SQRT(SS)
      DO 8 I = 1,N
    8 SO(I) = S(I)/SS
    9 DO 10 I  = 1,N
   10 X(I) = XI(I) + P*SO(I)
      DO 11 I = 1,N
      IF (X(I).GT.XMIN.AND.X(I).LT.XMAX) GO TO 11
      SN = 1.E20
      GO TO 99
   11 CONTINUE
      DO 5 I = 1,N
    5 Y(I) = X(I)**Q
      ALP(1) = (X(1) - X(4))/(Y01 - Y(1))
      ALP(2) = (X(2) - X02)/(Y(1) - Y(2))
      ALP(3) = (X(3) - X(2))/(Y02 - Y(3))
      ALP(4) = (X(4) - X01)/(Y(3) - Y(4))
      DO 14 I = 1,N
      IF (ALP(I).GT.ALPMN) GO TO 14
      SN = 1.E20
      GO TO 99
   14 CONTINUE
      AL1 = 1./(ALP(1) + ALP(4))
      CC = (1. + ALP(1)/ALP(2))*AL1
      CON1 = YF - (ALP(1)*Y(2) + ALP(4)*Y(4))*AL1
      CON2 = ALP(2)/ALP(1) - ALP(3)/ALP(4)
      S1 = 0.0
      S2 = 0.0
      DO 15 I = 1,N
      S1 = S1 + 1./X(I) + 1./(1. - X(I))
   15 S2 = S2 + 1./ALP(I)
      SN = CC + R*CON1**2 + RR*CON2**2 + R1*S1 + R2*S2
   99 GO TO (100,200,300), LA
  100 SP = SN
      C = P
      D = DEL
  110 LA = 2
      P = P + D
  120 IF (P.GT.0.0) GO TO 9
      GO TO (121,122,310),LA
  121 STOP
  122 P = P - 2.*D
      GO TO 120
  200 IF (SN.LT.SP) GO TO 220
      P = P - 2.*D
```

```
      LA = 3
      GO TO 120
  220 SP = SN
      C = P
      D = D*ALPHA
      GO TO 110
  300 IF (SN.GE.SP) GO TO 310
      SP = SN
      C = P
      D = -D
      GO TO 110
  310 P = P + D
      D = D*BETA
      DX = ABS(P*TOL/D)
      IF (1.0.LT.DX) GO TO 311
      DX = ABS(1.E-20/D)
      IF (1.0.LT.DX) GO TO 312
      GO TO 110
  312 WRITE (6,313)
  313 FORMAT (1H0,27HSTOP OCCURS ON DX UNDERFLOW/)
      KCT = 2
      RETURN
  311 SN = SP
      WRITE (6,320) CC,SN, (X(I),I=1,N)
  320 FORMAT(6H L/G =,F10.5,6H  SN =,F14.5,12H VARIABLES =,8F10.5)
      RETURN
      END
```

APPENDIX VII.

1. Derivation of Crossflow Algorithm.
2. Formulation of Derivatives for Deflected Gradient Method.

1. Derivation of Crossflow Algorithm.

Referring to fig. 3 and using the notation of Treybal (9) the adsorbent-solvent ratios for an N stage crossflow network may be written as follows:

$$\frac{L_{si}}{G_s} = \frac{Y_{i-1}-Y_i}{X_i-X_{0i}} \qquad i = 1,\ldots,N \qquad \text{(A1)}$$

and the total adsorbent-solvent ratio as:

$$F = \frac{L}{G} = \sum_{i=1}^{N} \frac{L_{si}}{G_s} = \sum_{i=1}^{N} \frac{Y_{i-1}-Y_i}{X_i-X_{0i}} \qquad \text{(A2)}$$

Providing an equilibrium relation (isotherm) exists between the Y_i and the X_i, the following equations hold at optimum:

$$\frac{dF}{dX_i} = \frac{\partial F}{\partial X_i} + \frac{\partial F}{\partial Y_i}\frac{dY_i}{dX_i} = 0 \qquad i = 1,\ldots,N-1 \qquad \text{(A3)}$$

Performing the above operations on (A2) and writing in terms of Y_i:

$$Y_i = Y_{i-1} + (X_i-X_{0i})\left[1 - \frac{X_i-X_{0i}}{X_{i+1}-X_{0i+1}}\right]\frac{dY_i}{dX_i} \qquad \text{(A4)}$$

$$i = 1,\ldots,N-1$$

(A4) is the most general form of the crossflow algorithm for an N stage system.

If all initial adsorbent streams have zero solute, then (A4) simplifies to:

$$Y_i = Y_{i-1} + X_i \left[1 - \frac{X_i}{X_{i+1}}\right] \frac{dY_i}{dX_i} \qquad i = 1,\ldots,N-1 \quad (A5)$$

If the Freundlich isotherm: $Y_i = X_i^n$, $i = 1,\ldots,N$, is used then (A5) may be written in terms of X_i , X_{i+1}, X_{i+2} :

$$X_i^n = X_{i+1}^n \left\{1 - n\left[1 - \frac{X_{i+1}}{X_{i+2}}\right]\right\} \qquad i = 0,1,\ldots,N-2 \quad (A6)$$

where X_0 is a pseudo - concentration in equilibrium with the Y_0 of the entering solvent stream. Knowing the final concentration in the solvent stream, Y_F , and hence the concentration X_N in equilibrium with Y_F , the X_i , $i = 0,1,\ldots,N-3$ can be computed in the following manner:

i. Guess an appropriate value for X_{N-1} $(X_{N-1} > X_N)$.

ii. Using (A6) compute X_{N-2} . Use the equation again to compute X_{N-3} . Continue in this manner until X_0 is computed.

iii. If the computed X_0 is less than a certain tolerance of the expected value assume that the solution is correct and go to iv. If not, reset X_{N-1} according to whether the computed X_0 was larger or smaller than the expected value. Return to ii. and re-iterate until a new X_0 is computed.

iv. Determine F from (A2) where $X_{0i} = 0$, $Y_i = X_i^n$, $i = 1,\ldots,N$.

For the special case when $n = 1$ an analytical solution for the optimum may be determined. (A6) becomes:

$$X_i = \frac{X_{i+1}^2}{X_{i+2}} \qquad i = 0,1,\ldots,N-2 \qquad \text{(A7)}$$

with $Y_0 = X_0$ and $Y_F = X_N$. It can be shown that (A7) may be written in terms of Y_F and Y_0 as follows:

$$X_i = Y_F^{\frac{i}{N}} \cdot Y_0^{\frac{N-i}{N}} \qquad i = 0,1,\ldots,N \qquad \text{(A8)}$$

Substituting (A8) into (A2) and simplifying, noting that all initial adsorbent streams have zero solute:

$$F = F_N = N \frac{Y_0^{\frac{1}{N}}}{Y_F} - 1 \qquad \text{(A9)}$$

(A9) gives the optimum value of F for any value of N. Taking the limit as N approaches infinity:

$$\lim_{N \to \infty} F_N = \ln \frac{Y_0}{Y_F} \qquad \text{(A10)}$$

When $Y_0 = 1.0$ and $Y_F = 0.1$, the limiting optimum is $\ln(10)$.

2. Formulation of Derivatives For Deflected Gradient Method

Using the nomenclature of Treybal (9) and referring to fig. 10 the mass balances for each stage are written as follows:

$$
\begin{aligned}
&\text{i.} && Y_{01}G_{s1} + X_4L_{s2} = Y_1G_{s1} + X_1L_{s2} \\
&\text{ii.} && Y_1G_{s1} + X_{02}L_{s1} = Y_2G_{s1} + X_2L_{s1} \\
&\text{iii.} && Y_{02}G_{s2} + X_2L_{s1} = Y_3G_{s2} + X_3L_{s1} \\
&\text{iv.} && Y_3G_{s2} + X_{01}L_{s2} = Y_4G_{s2} + X_4L_{s2}
\end{aligned}
\qquad \text{(B1)}
$$

Rearranging equations (B1) as solvent - adsorbent ratios it is possible to obtain:

$$
\begin{aligned}
&\text{i.} && \alpha_1 = \frac{G_{s1}}{L_{s2}} = \frac{X_1-X_4}{Y_{01}-Y_1} \\
&\text{ii.} && \alpha_2 = \frac{G_{s1}}{L_{s1}} = \frac{X_2-X_{02}}{Y_1-Y_2} \\
&\text{iii.} && \alpha_3 = \frac{G_{s2}}{L_{s1}} = \frac{X_3-X_2}{Y_{02}-Y_3} \\
&\text{iv.} && \alpha_4 = \frac{G_{s2}}{L_{s2}} = \frac{X_4-X_{01}}{Y_3-Y_4}
\end{aligned}
\qquad \text{(B2)}
$$

Since it is desired that the average concentration in the output solution be a fixed quantity the first constraint is:

$$
Y_F = \frac{Y_2G_{s1}+Y_4G_{s2}}{G_{s1}+G_{s2}} \qquad \text{(B3)}
$$

By using equations (B2) and rearranging, (B3) becomes:

$$Y_F - \frac{\alpha_1 Y_2 + \alpha_4 Y_4}{\alpha_1 + \alpha_4} = \text{CON1} \tag{B4}$$

(B4) is the first equality constraint and is denoted as CON1 . The second equality constraint comes from the inter-relation of equations (B2):

$$\frac{\alpha_2}{\alpha_1} - \frac{\alpha_3}{\alpha_4} = \text{CON2} \tag{B5}$$

(B5) is denoted as CON2 . Inequality constraints are of a more arbitrary nature, depending on the user's direction. However, to prevent absurd solutions the following contraints are necessary:

$$\alpha_i \geq 0 \qquad i = 1, \ldots, 4 \tag{B6}$$

$$0 \leq X_i \leq 1 \qquad i = 1, \ldots, 4 \tag{B7}$$

Occasionally it is necessary to put a bound on the α_i to prevent the numerator and denominator from changing sign at $+\infty$.

The standard function to be minimized is the adsorbent-solvent ratio:

$$\frac{L}{G} = \frac{L_{s1} + L_{s2}}{G_{s1} + G_{s2}} \tag{B8}$$

using equations (B2), (B8) may be written in terms of α_i :

$$\frac{L}{G} = \frac{1 + \alpha_1/\alpha_2}{\alpha_1 + \alpha_4} \tag{B9}$$

According to the theorem of Courant (3) and the method prescribed by Carroll (2) the total objective function is written:

$$F = \frac{L}{G} + R\cdot CON1^2 + RR\cdot CON2^2 + R1\cdot \sum_{i=1}^{4}\left(\frac{1}{X_i} - \frac{1}{1-X_i}\right) + R2\cdot \sum_{i=1}^{4} \frac{1}{\alpha_i} \tag{B10}$$

where R,RR,R1 , and R2 are penalty factors, the first pair to be varied according to the theorem of Courant, and the second pair according to the method prescribed by Carroll.

Using the chain rule the general formulation of the gradient vector is as follows:

$$\frac{dF}{dX_i} = \sum_{k=1}^{4} \frac{\partial F}{\partial \alpha_k}\left(\frac{\partial \alpha_k}{\partial X_i} + \frac{\partial \alpha_k}{\partial Y_i}\,\frac{dY_i}{dX_i}\right) + \frac{\partial F}{\partial Y_i}\,\frac{dY_i}{dX_i} + \frac{\partial F}{\partial X_i} \qquad i = 1,\ldots,4 \tag{B11}$$

where the dF/dX_i are the components of the gradient vector. For the problem of fig. 10 most of the components of (B11) vanish. The partial derivatives of F with respect to the α_i are:

$$\frac{\partial F}{\partial \alpha_1} = \frac{1}{(\alpha_1+\alpha_4)^2}\left[\frac{\alpha_4}{\alpha_2} - 1 + 2\cdot R\cdot CON1\cdot\alpha_4\cdot(Y_4-Y_2)\right] - \frac{2\cdot RR\cdot CON2.\alpha_2 - R2}{\alpha_1^2}$$

$$\frac{\partial F}{\partial \alpha_2} = -\frac{1}{\alpha_2^2}\left(\frac{\alpha_1}{\alpha_1+\alpha_4} + R2\right) + \frac{2RRCON2}{\alpha_1}$$

$$\frac{\partial F}{\partial \alpha_3} = -\frac{2\cdot RR\cdot CON2}{\alpha_4} - \frac{R2}{\alpha_3^2} \tag{B12}$$

$$\frac{\partial F}{\partial \alpha_4} = -\frac{1}{(\alpha_1+\alpha_4)^2}\left[1 + \frac{\alpha_1}{\alpha_2} + 2\cdot R\cdot CON1\alpha_1\cdot(Y_4-Y_2)\right] + \frac{2\cdot RR\cdot CON2\cdot\alpha_3 - R2}{\alpha_4^2}$$

The partials of F with respect to the Y_i are:

$$\frac{\partial F}{\partial Y_2} = - \frac{2 \cdot R \cdot CON1 \cdot \alpha_1}{\alpha_1+\alpha_4}$$

$$\frac{\partial F}{\partial Y_4} = - \frac{2 \cdot R \cdot CON1 \cdot \alpha_4}{\alpha_1+\alpha_4} = \frac{\partial F}{\partial Y_2} \frac{\alpha_4}{\alpha_1} \tag{B13}$$

The other $\partial F/\partial Y_i$ are zero. The partials of the α_k with respect to the X_i are:

$$\frac{\partial \alpha_1}{\partial X_1} = \frac{1}{Y_{01}-Y_1}$$

$$\frac{\partial \alpha_1}{\partial X_4} = - \frac{1}{Y_{01}-Y_1} = - \frac{\partial \alpha_1}{\partial X_1}$$

$$\frac{\partial \alpha_2}{\partial X_2} = \frac{1}{Y_1-Y_2} \tag{B14}$$

$$\frac{\partial \alpha_3}{\partial X_2} = - \frac{1}{Y_{02}-Y_3}$$

$$\frac{\partial \alpha_3}{\partial X_3} = \frac{1}{Y_{02}-Y_3} = - \frac{\partial \alpha_3}{\partial X_2}$$

$$\frac{\partial \alpha_4}{\partial X_4} = \frac{1}{Y_3-Y_4}$$

The other $\partial \alpha_k/\partial X_i$ are zero. The partials of the α_k with respect to the Y_i are:

$$\frac{\partial \alpha_1}{\partial Y_1} = \frac{X_1-X_4}{(Y_{01}-Y_1)^2} = \alpha_1 \frac{\partial \alpha_1}{\partial X_1}$$

$$\frac{\partial\alpha_2}{\partial Y_1} = -\frac{X_2-X_{02}}{(Y_1-Y_2)^2} = -\alpha_2\frac{\partial\alpha_2}{\partial X_2}$$

$$\frac{\partial\alpha_2}{\partial Y_2} = \frac{X_2-X_{02}}{(Y_1-Y_2)^2} = -\frac{\partial\alpha_2}{\partial Y_1}$$

$$\frac{\partial\alpha_3}{\partial Y_3} = \frac{X_3-X_2}{(Y_{02}-Y_3)^2} = \alpha_3\frac{\partial\alpha_3}{\partial X_3} \tag{B15}$$

$$\frac{\partial\alpha_4}{\partial Y_3} = -\frac{X_4-X_{01}}{(Y_3-Y_4)^2} = -\alpha_4\frac{\partial\alpha_4}{\partial X_4}$$

$$\frac{\partial\alpha_4}{\partial Y_4} = \frac{X_4-X_{01}}{(Y_3-Y_4)^2} = -\frac{\partial\alpha_4}{\partial Y_3}$$

The other $\partial\alpha_k/\partial Y_i$ are zero. The partials of F with respect to the X_i are:

$$\frac{\partial F}{\partial X_i} = R1\cdot\left[\frac{1}{(1-X_i)^2} - \frac{1}{X_i^2}\right] \qquad i = 1,\ldots,4 \tag{B16}$$

Eliminating the elements of equations (B12) to (B16) that are zero, equations (B11) become:

$$\frac{dF}{dX_1} = \frac{\partial F}{\partial X_1} + \frac{\partial F}{\partial\alpha_1}\left[\frac{\partial\alpha_1}{\partial X_1} + \frac{\partial\alpha_1}{\partial Y_1}\frac{dY_1}{dX_1}\right] + \frac{\partial F}{\partial\alpha_2}\frac{\partial\alpha_2}{\partial Y_1}\frac{dY_1}{dX_1}$$

$$\frac{dF}{dX_2} = \frac{\partial F}{\partial X_2} + \frac{\partial F}{\partial\alpha_2}\left[\frac{\partial\alpha_2}{\partial X_2} + \frac{\partial\alpha_2}{\partial Y_2}\frac{dY_2}{dX_2}\right] + \frac{\partial F}{\partial\alpha_3}\frac{\partial\alpha_3}{\partial X_2} + \frac{\partial F}{\partial Y_2}\frac{dY_2}{dX_2} \tag{B17}$$

$$\frac{dF}{dX_3} = \frac{\partial F}{\partial X_3} + \frac{\partial F}{\partial\alpha_3}\left[\frac{\partial\alpha_3}{\partial X_3} + \frac{\partial\alpha_3}{\partial Y_3}\frac{dY_3}{dX_3}\right] + \frac{\partial F}{\partial\alpha_4}\frac{\partial\alpha_4}{\partial Y_3}\frac{dY_3}{dX_3}$$

$$\frac{dF}{dX_4} = \frac{\partial F}{\partial X_4} + \frac{\partial F}{\partial \alpha_1}\frac{\partial \alpha_1}{\partial X_4} + \frac{\partial F}{\partial \alpha_4}\left[\frac{\partial \alpha_4}{\partial X_4} + \frac{\partial \alpha_4}{\partial Y_4}\frac{dY_4}{dX_4}\right] + \frac{\partial F}{\partial Y_4}\frac{dY_4}{dX_4} \qquad \text{(B17)}$$

Equations (B17) now are the components of the gradient vector. When the Freundlich isotherm is used:

$$\frac{dY_i}{dX_i} = n \cdot X_i^{n-1} \qquad \text{(B18)}$$

When the Koble - Corrigan isotherm is used:

$$\frac{dY_i}{dX_i} = \frac{n \cdot Y_i (1 + m^{\frac{1}{n}})}{X_i (1 + m^{\frac{1}{n}}(1 - X_i))} \qquad \text{(B19)}$$

APPENDIX VIII

Equations of Isotherms Used.

Freundlich Isotherm

The general equation for the Freundlich Isotherm is written:

$$\xi = c\eta^{n}$$

where

ξ = concentration of solute in fluid, lb. solute/lb. solvent.

η = concentration of adsorbate, lb. solute adsorbed/lb. adsorbent.

n = exponent ($n > 0$)

If $\eta' = \max \eta$, then $\xi' = \max \xi$. The Freundlich Isotherm may then be written in the dimensionless form:

$$Y = \frac{\xi}{\xi'} = \frac{c\eta^{n}}{c\eta'^{n}} = X^{n}$$

where:

Y = concentration of solute in fluid, dimensionless ($0 \leq Y \leq 1$).

X = concentration of adsorbate, dimensionless ($0 \leq X \leq 1$).

Koble - Corrigan Isotherm

Applying the same procedure as for the Freundlich isotherm, the dimensionless Koble - Corrigan isotherm becomes:

$$Y = \left[\frac{X}{1+m^{1/n}(1-X)}\right]^{n}$$

where:

m = parameter ($m > 0$)

n = exponent ($n > 0$)