OPTIMAL TEMPERATURE CONTROL IN BATCH REACTORS

by

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

ABSTRACT

A general study is made of means of approximating optimal temperature control in batch reactors by simple conventional control equipment.

The performance of standard three mode - proportional, integral, derivative - linear controllers that sense product concentration and manipulate temperature is investigated for common classes of reaction systems. These controllers are found to be capable of giving essentially optimal performance over broad ranges of kinetic parameters.

Simple proportional controllers designed to track a pre-set temperature curve are studied as well. For reversible exothermic reactions, these are also found to be capable of giving near-optimal performance.

The sensitivity of the reactor performance to choice of controller constants is studied. Stability analysis for the closed loop systems are made.

The study as a whole shows the feasibility of approximating the optimal performance of batch reactors with conventional, relatively inexpensive, control equipment.

CHAPTER 1 INTRODUCTION

1.1 HISTORICAL BACKGROUND

A great deal of work has been done in recent years to develop means for determining best temperature sequences in batch reactors. Broadly speaking, this work proceeded along two fronts: by applying methods of the classical calculus of variations, which led to the a priori determination of best temperature schedules; and by applying the newer methods of dynamic programming, which led to the determination of best strategies for controlling the temperature.

A pioneering study based on variational methods was carried out by Bilous and Amundson(1956). They studied a number of reaction mechanisms, finding, for example, for consecutive reactions $A \rightarrow B \rightarrow C$, that if the activation energy of the first step is less than that of the second, it is to be expected that a decreasing temperature will favor production of B. In the early part of the reaction a high temperature is needed to speed up the reaction, whereas towards the end a lower temperature will minimize the formation of C. For a complex reaction scheme, Denbigh (1958) found that the proper choice of temperature sequence can greatly improve the product yield. Horn (1961 a,b,c) and Katz (1960) developed general formulations

for this class of problems. The decisive mathematical step in the development of these variational methods was taken by Pontrjagin (1962) in his development of the maximum principle. This has been given an engineering formulation by Fan (1966), and has been explicitly applied by Siebenthal and Aris (1964), by Lee (1964, 1968) and by many others to the solution of reactor optimization problems.

The variational methods discussed in the preceding paragraph, which are designed in the first instance to characterize best temperature schedules, can of course be applied to characterize best temperature control actions as well. Control problems indeed furnished the immediate motivation for Pontrjagin's work, and studies along these lines have been carried out by Kipiniak (1961), Katz (1964) and many others. The methods of dynamic programming developed by Bellman (1957) give a more natural formulation of the control problem, and have been applied to reactor optimization by Aris (1960,1961) and others.

Now the methods of optimal control - whether formulated in the manner of Bellman or of Pontrjagin - call for massive amounts of computing control equipment if they are to be applied on-line. The main idea of the present work is to see how well these exact optimal controllers can be approximated by simple conventional control equipment.

1.2 SCOPE AND MAIN RESULTS OF THIS WORK

In any exact formulation of the optimal temperature control problem for a batch reactor, the best temperature to apply at any time during the batch cycle depends not only on the composition of the reacting mass at that time, but also on the time remaining until the end of the cycle. To incorporate these dependencies into a control system calls either for massive on-line computing capacity, or for on-line access to very large memories. This work addresses itself to the problem of approximating such exact optimal control systems by simple conventional controllers.

The principal conventional control equipment studied here are standard three mode - proportional, integral, derivative - controllers, arranged so as to sense product concentration in the reactor and manipulate the reactor temperature. For a given reaction scheme, the controller constants are to be set once and for all to give the best reactor performance, although in the absence of full knowledge of the kinetics, they may of course be adjusted from one run to another so as to improve the performance. The measure of reactor performance used throughout this work is the product yield, and these conventional controllers are applied primarily to a class of common reaction mechanisms where the best product yield attainable

by arbitrary manipulation of the reactor temperature is known by prior calculation (Evangelista and Katz, 1968).

For these common reaction mechanisms - consecutive and parallel first order kinetics - the best controller constants can be evaluated (Millman and Katz, 1967), and turn out to give essentially the maximum theoretical yield over a broad range of kinetic parameters. Using only proportional control will give almost the maximum theoretical yield. An analysis of these suboptimal proportional controllers shows that the reactor behavior is quite insensitive to changes in controller settings in certain directions, but moderately sensitive to changes in other directions. Further analysis reveals that these suboptimal controllers tend to have local instabilities over a good part of the batch cycle, although their time constants will not in general lead to undue amplification of disturbances. This whole order of ideas is applied to a complex reaction scheme that has been exhaustively studied by Rosenbrock and Storey (1966) and others, where it turns out to work very well.

The "sense yield, manipulate temperature" controllers discussed above assume of course that an arbitrarily large heat duty is available to implement the desired control action. And, the sensitivity and stability problems noted above provide a further impetus for the study

of other methods of approximating optimal control. Accordingly, an analysis is made of a proportional controller that senses reactor temperature and manipulates the flow rate of heat exchanger fluid so as to drive the temperature toward a pre-set schedule. This study is carried out for a reversible, first order, exothermic reaction, where optimal behavior can be achieved as well by forcing the temperature to track a pre-set function of product concentration. It turns out that essentially optimal behavior can be attained with quite reasonable configurations of control equipment, and that the sensitivity and stability problems noted above by and large disappear. These tracking controllers can of course be used in cascade with the suboptimal controllers discussed above, although no detailed analysis of such systems are carried out here.

The study as a whole shows the direct feasibility of approximating the optimal performance of batch reactors with conventional, relatively inexpensive, controllers.

1.3 PLAN OF THIS STUDY

The bulk of the present study is devoted to an analysis of the "sense yield, manipulate temperature" controllers described in the preceding section. Chapter 2 is given over to the appropriate mathematical formulations, and Chapter 3 to a discussion of the numerical methods by which the best controller settings are found. Chapter 4 presents the results of these calculations for consecutive and parallel first order reactions.

In Chapter 5, the sensitivity of the reactor performance to departures from the best controller settings is systematically analysed, and contours of constant yield in the space of the controller settings are developed. Chapter 6 analyzes the stability of the reactor system, and investigates numerically its performance to such random disturbances as may be expected to occur in practice. Chapter 7 studies the application of controllers of this type to a complex reaction scheme.

Chapter 8 shifts to a study of the tracking controllers described in the last section, and analyzes their behavior for first order reversible reactions.

CHAPTER 2 FORMULATION OF THE OPTIMIZATION PROBLEM

2.1 GENERAL SOLUTION

The course of a reaction in a batch chemical reactor may be described by the differential equations

$$\frac{dx_i}{dt} = f_i(\underline{x},u); \quad i = 1,2,\cdots n; \qquad (2.1)$$

$$0 \leq t \leq 1$$

where t, the normalized reaction time, runs from zero at the start of the reaction to unity at the end of the reaction, $\underline{x}$, the state vector

$$\underline{x} = \underline{x}(t) = \left[x_1(t),x_2(t),\cdots x_n(t)\right] \qquad (2.2)$$

consists essentially of the composition variables along the reaction path, and u(t), $0 \leq t \leq 1$, is the temperature schedule. The functions, f_i, contain the reaction mechanism and kinetic parameters. In this work, the desired product concentration will be $x_2(t)$, and the criterion for reactor performance will be taken as the level of product yield. Thus the optimal situation is one for which

$$x_2(1) = \text{maximum} \qquad (2.3)$$

The optimal temperature schedule problem is to choose $u(t)$, $0 \leq t \leq 1$, subject to bounds, which maximizes $x_2(1)$. Either the schedules themselves, or various techniques for obtaining them, are available.

The question asked in this work is - with the theoretical optimum temperature schedule available, how well can it be approximated by commercial controllers? The course of the reaction in the batch reactor is now described by the differential equations

$$\frac{dx_i}{dt} = f_i(\underline{x},\underline{c}); \quad i = 1,2,\cdots n; \qquad (2.4)$$

$$0 \leq t \leq 1$$

where the state vector, $\underline{x}$, is enlarged to contain variables associated with the controller and $\underline{c}$ is a vector of controller settings. The problem here is to choose the vector of controller settings, $\underline{c}$, to maximize $x_2(1)$. Then the results are studied to see both how closely a controller result approaches the theoretical optimum and how it compares with other controllers.

The type of controller studied is a conventional three-term (proportional-integral-derivative) controller of the form

$$m = M + K_c e + (K_c/T_i)\int_0^t (e)dt + K_c T_d \frac{de}{dt} \qquad (2.5)$$

where m is the manipulated variable with set point M, e is the deviation of the measured variable from its set point, K_c is the proportional sensitivity, T_i is the integral time and T_d is the derivative time. It was decided that the controller would measure desired product concentration and control temperature directly. This simplified controller model was chosen as showing most directly how closely conventional temperature control could be made to approximate optimality. Later on in this work, certain studies are presented which indicate how more complex, realistic control systems perform.

The controller is

$$u = u_o + K_c(x_2 - x_2^d) + (K_c/T_i)\int_0^t (x_2 - x_2^d)dt + K_c T_d \frac{dx_2}{dt} \qquad (2.6)$$

where u is temperature, x_2 is product concentration, u_o

and x_2^d are constants. At the start of the batch,

$$u(0) = u_o + K_c(x_2(0) - x_2^d) + K_c T_d \left(\frac{dx_2}{dt} \right)_{t=0} \tag{2.7}$$

Since the model of the batch reactor is in the form of a set of differential equations, it is convenient to have the controller as a differential equation also. It is also much easier computationally, as will be shown a little further on, to handle the controller as a differential equation. Differentiating Equation 2.6 gives

$$\frac{du}{dt} = K_c \frac{dx_2}{dt} + (K_c/T_i)x_2 - (K_c/T_i)x_2^d + \frac{d^2x_2}{dt^2} \quad ;$$
$$u(0) = u_o + K_c x_2(0) - K_c x_2^d + K_c T_d \left(\frac{dx_2}{dt} \right)_{t=0} \tag{2.8}$$

A more convenient set of constants is defined as

$$\begin{aligned} c_2 &= K_c \\ c_3 &= K_c/T_i \\ c_4 &= -K_c x_2^d/T_i \\ c_5 &= K_c T_d \end{aligned} \tag{2.9}$$

where

$$\begin{aligned} K_c &= c_2 \\ T_i &= c_2/c_3 \\ x_2^d &= -c_4/c_3 \\ T_d &= c_5/c_2 \end{aligned} \tag{2.10}$$

Now c_1 may be defined as

$$c_1 = u(0) = u_o + c_2c_4/c_3 + c_2x_2(0) + c_5\left(\frac{dx_2}{dt}\right)_{t=0} \tag{2.11}$$

and

$$u_o = c_1 - c_2c_4/c_3 - c_2x_2(0) - c_5\left(\frac{dx_2}{dt}\right)_{t=0} \tag{2.12}$$

Thus the controller is specified by five parameters - either K_c, T_i, T_d, x_2^d and u_o or, more conveniently, c_1, c_2, c_3, c_4 and c_5. The values of $x_2(0)$ and $(dx_2/dt)_{t=0}$ come from the reactor model.

The three-term controller model is thus

$$\frac{du}{dt} = c_2(dx_2/dt) + c_3x_2 + c_4 + c_5(d^2x_2/dt^2) \qquad (2.13)$$

$$u(0) = c_1$$

For proportional control, $c_3 = c_4 = c_5 = 0$, and

$$\frac{du}{dt} = c_2(dx_2/dt) \qquad (2.14)$$

$$u(0) = c_1$$

Specifying c_1 and c_2 is sufficient to define the controller and obviously $T_i \rightarrow \infty$ and $T_d = 0$, but only two of the three constants K_c, u_o and x_2^d can be recovered. Fixing c_1 can only determine the specific relationship between u_o and x_2^d.

For proportional-integral control, $c_5 = 0$, and

$$\frac{du}{dt} = c_2(dx_2/dt) + c_3x_2 + c_4 \qquad (2.15)$$

$$u(0) = c_1$$

Through Equations 2.10 and 2.12, a complete transformation

of variables can be made.

For proportional-derivative control, $c_3 = c_4 = 0$, and

$$\frac{du}{dt} = c_2(dx_2/dt) + c_5(d^2x_2/dt^2) \qquad (2.16)$$

$$u(0) = c_1$$

In this case, as with proportional control, there is a relationship between u_o and x_2^d when c_1 is specified, but they cannot be determined independently.

It may be noted here in summary that the suboptimal control study which follows is carried out in terms of the(mathematically convenient)parameters c_1, c_2, c_3, c_4, c_5 rather than in terms of the physical controller parameters u_o, x_2^d, K_c, T_i, T_d. The relationship between these two controller descriptions is only one-to-one when the integral mode of control is included. For proportional or proportional-derivative control, only the sensitivity parameter K_c, the derivative time T_d and a linear relationship between u_o and x_2^d are uniquely determined by the c's.

A block diagram of the suboptimal controller is shown in Figure 2.1.

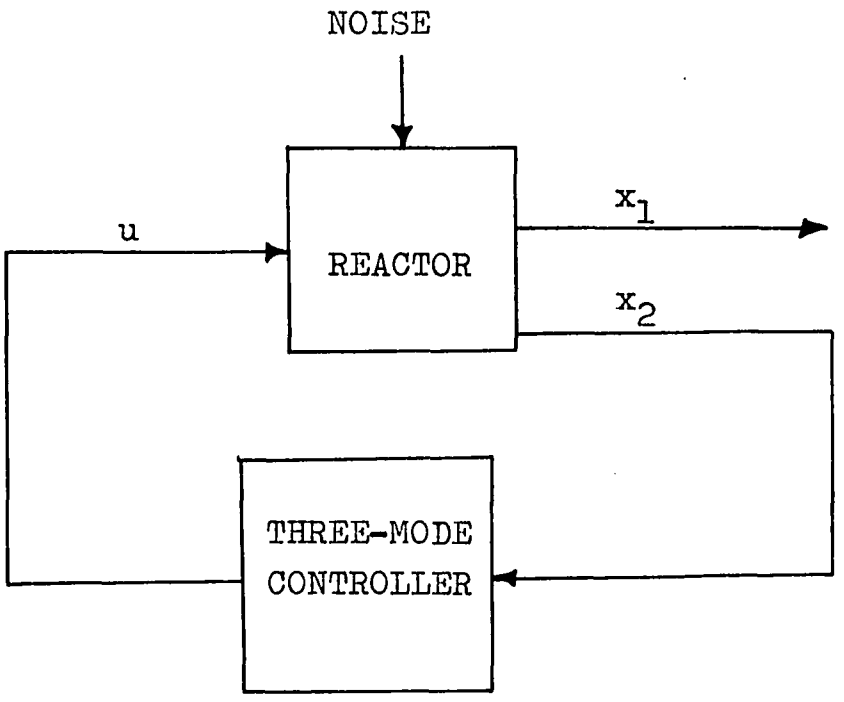


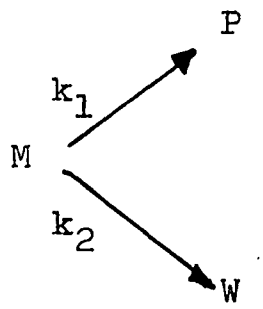
FIGURE 2.1 THREE-MODE SUBOPTIMAL CONTROLLER

2.2 CONSECUTIVE AND PARALLEL REACTION SCHEMES

The case studies in this work consist of a survey of consecutive

$$M \xrightarrow{k_1} P \xrightarrow{k_2} W \tag{2.17}$$

and parallel



(2.18)

first order reactions, where raw material M makes product P and waste W. For both reaction schemes, the rate constants are taken in the standard Arrhenius form

$$\begin{aligned} k_1 &= A_1 \exp(- E_1/R\theta) \\ k_2 &= A_2 \exp(- E_2/R\theta) \end{aligned} \tag{2.19}$$

where A_1, A_2 are the frequency factors of the two reactions, E_1, E_2 their activation energies, R the gas constant and θ the absolute temperature. The kinetic equa-

tions for the two reaction schemes are as follows. For the consecutive reactions,

$$\begin{aligned} dM/dT &= -k_1 M \quad ; \quad M(0) = M_i \\ dP/dT &= k_1 M - k_2 P; \quad P(0) = 0 \end{aligned} \tag{2.20}$$

For the parallel reactions,

$$\begin{aligned} dM/dT &= -(k_1 + k_2)M; \quad M(0) = M_i \\ dP/dT &= k_1 M \quad ; \quad P(0) = 0 \end{aligned} \tag{2.21}$$

Here the chemical symbols M and P of the reacting species stand for their concentrations as well, and T is the time.

The Equations 2.20 and 2.21 may be taken as describing the courses of the various reactions in batch, starting with pure raw material at concentration M_i. From this point of view, the situation may be conveniently described in terms of the variables

$$\begin{aligned} x_1 &= M/M_i \\ x_2 &= P/M_i \end{aligned} \tag{2.22}$$

rather than in terms of the concentration M and P directly.

The schemes can now be written in the new variables. The consecutive scheme becomes,

$$\begin{aligned} dx_1/dT &= -k_1x_1 \quad ; \quad x_1(0) = 1 \\ dx_2/dT &= k_1x_1 - k_2x_2 \quad ; \quad x_2(0) = 0 \end{aligned} \tag{2.23}$$

and the parallel scheme is

$$\begin{aligned} dx_1/dT &= -(k_1 + k_2)x_1 \quad ; \quad x_1(0) = 1 \\ dx_2/dT &= k_1x_1 \quad ; \quad x_2(0) = 0 \end{aligned} \tag{2.24}$$

It is convenient to carry out the calculations and present the results in terms of certain dimensionless quantities. In place of time T is introduced the variable

$$t = T/T_f \tag{2.25}$$

where T_f is the total batch time. A dimensionless temperature is defined as

$$u = T_f A_1 \exp(-E_1/R\theta) \tag{2.26}$$

Also introduced are the dimensionless parameters

$$\alpha = E_2/E_1 \tag{2.27}$$

and

$$\beta = (T_f A_2)/(T_f A_1)^{\gamma} \tag{2.28}$$

The batch equations for the two reaction schemes may be written as follows. For the consecutive reactions,

$$\begin{aligned} dx_1/dt &= -ux_1 \quad ; \quad x_1(0) = 1 \\ dx_2/dt &= ux_1 - \beta u^{\gamma} x_2; \quad x_2(0) = 0 \end{aligned} \tag{2.29}$$

For the parallel reactions,

$$\begin{aligned} dx_1/dt &= -(u + \beta u^{\gamma})x_1; \quad x_1(0) = 1 \\ dx_2/dt &= ux_1 \quad ; \quad x_2(0) = 0 \end{aligned} \tag{2.30}$$

The parameters α and β completely characterize the batch and there are only two variables to survey rather than five.

The optimal temperature question is to choose u as a function of t, $0 \leq t \leq 1$ to make $x_2(1)$ a maximum, subject to the condition $0 \leq u \leq \gamma$, where

$$\gamma = T_f A_1 \exp(- E_1/R\theta_m) \qquad (2.31)$$

and θ_m is the maximum allowable absolute temperature. Once the optimal schedule is obtained, the various controllers described previously are studied to see how closely they approximate the behavior of the optimal schedule. That is, using the same α and β values, a controller type (such as proportional-integral) is chosen, and the problem is to choose $\underline{c}$, the vector of controller settings, to maximize $x_2(1)$, also subject to the same temperature bounds. The various controller types are each studied. Then one can see both how a particular controller compares with the others and how closely it approximates the optimal temperature schedule. This study is carried out over a broad range of kinetic parameters for both reaction schemes.

CHAPTER 3 SEEKING THE OPTIMUM

3.1 INTRODUCTION

As the problem has now been stated, the basic task is to choose certain free parameters contained in a set of nonlinear differential equations to maximize a certain final value. Since the set of nonlinear equations cannot be solved analytically and must be solved by numerical techniques, the choice of the free parameters must also be carried out numerically with an optimization procedure. The optimization procedures employed here are variations of steepest ascent methods, and accordingly require gradient calculations.

First the method of obtaining the gradients will be shown, then the hill-climbing problem will be discussed, and finally such computational matters as choosing starting values for the parameters and actual numerical schemes will be presented.

3.2 COMPUTATION OF THE GRADIENTS

In deriving the gradient equations, it is convenient and economical to do it all at once for the optimal controller setting problem and the optimal schedule problem. The set of differential equations

$$dx_i/dt = f_i(\underline{x},u,c); \quad x_i(0) = a_i$$
$$i = 1,2,\cdots n; \quad 0 \leq t \leq 1 \tag{3.1}$$

contains both problems. The gradient of $x_2(1)$, the quantity to be maximized, is to be found with respect to: $u(t)$, $0 \leq t \leq 1$, the temperature schedule; c, a representative parameter contained in the f's; and a_r, a representative initial condition. Basepoint values - $u^*(t)$, c^* and a_r^* - at which the gradient is sought are chosen and through Equation 3.1, the associated state variables $\underline{x}^*(t)$ are calculated. Then the basepoint values are slightly perturbed

$$\begin{aligned} u(t) &= u^*(t) + \delta u(t) \\ a_r &= a_r^* + \delta a_r \\ c &= c^* + \delta c \end{aligned} \tag{3.2}$$

to give

$$x_i = x_i^* + \delta x_i; \quad i = 1,2,\cdots n \tag{3.3}$$

Putting Equation 3.3 into Equation 3.1 gives

$$\begin{aligned} d(\delta x_i)/dt &= \sum_{j=1}^{n} (\partial f_i/\partial x_j)\delta x_j + \\ &(\partial f_i/\partial u)\delta u + (\partial f_i/\partial c)\delta c \\ \delta x_i(0) &= 0 \text{ if } i \neq r; \\ \delta x_r(0) &= \delta a_r \end{aligned} \tag{3.4}$$

where the partial derivatives are evaluated at the base-point. A set of adjoint differential equations is defined as

$$\begin{aligned} dz_i/dt &= -\sum_{j=1}^{n} (\partial f_j/\partial x_i)z_j; \quad i = 1,2,\cdots n \\ z_2(1) &= 1; \quad z_i(1) = 0 \text{ if } i \neq 2 \end{aligned} \tag{3.5}$$

where the special endpoint condition on z_2 arises because it is the gradient of $x_2(1)$ which is sought.

Multiplying Equation 3.5 by x_i and Equation 3.4 by z_i and adding gives

$$\frac{d}{dt}\left[\sum_{i=1}^{n} z_i \delta x_i\right] = \sum_{i=1}^{n} z_i (\partial f_i/\partial u)\delta u + \sum_{i=1}^{n} z_i (\partial f_i/\partial c)\delta c \tag{3.6}$$

Integrating gives

$$\sum_{i=1}^{n} z_i(1)\delta x_i(1) = \sum_{i=1}^{n} z_i(0)\delta x_i(0) + \int_0^1 \sum_{i=1}^{n} z_i (\partial fi/\partial c)\,\delta c\, dt + \int_0^1 \sum_{i=1}^{n} z_i (\partial f_i/\partial u)\,\delta u\, dt \tag{3.7}$$

Putting in the proper boundary conditions,

$$\delta x_2(1) = z_r(0)\delta a_r + \int_0^1 \sum_{i=1}^{n} z_i (\partial f_i/\partial c)\,\delta c\, dt + \int_0^1 \sum_{i=1}^{n} z_i (\partial f_i/\partial u)\,\delta u\, dt \tag{3.8}$$

From Equation 3.8, it is recognized that the gradient on the initial condition a_r is $z_r(0)$ and the gradient on c is

$$\int_0^1 \sum_{i=1}^{n} z_i (\partial f_i/\partial c)\, dt.$$

The schedule can be represented by dividing the batch in-

terval into a large number of parts and assigning a temperature value to each part. The schedule value at a particular time, t, is $u(t)$ and its gradient is

$$\sum_{i=1}^{h} z_i (\partial f_i / \partial u),$$

where all values are evaluated at t.

Now if the basepoint values at which the gradient is being calculated are trials in the way to maximize $x_2(1)$, then the perturbations - δa_r, δc, $\delta u(t)$ - should be chosen so as to make $\delta x_2(1)$ positive. It can be seen from Equation 3.8 that choosing these perturbations along the gradient

$$\begin{aligned} \delta a_r &= + H \cdot z_r(0) \\ \delta u(t) &= + H \cdot \sum_{i=1}^{h} z_i (\partial f_i / \partial u) \\ \delta c &= + H \cdot \int_0^1 \sum_{i=1}^{h} z_i (\partial f_i / \partial c) \; dt \end{aligned} \qquad (3.9)$$

where H is a positive constant, does just this.

An equivalent and more convenient method for handling parameters such as c is to transform them into initial conditions. An additional differential equation

$$dx_{n+1}/dt = 0 \; ; \quad x_{n+1}(0) = c \qquad (3.10)$$

is added where $x_{n+1} = c$. The adjoint equation is

$$dz_{n+1}/dt = -\sum_{j=1}^{n}(\partial f_j/\partial x_{n+1})z_j;\ z_{n+1}(1) = 0$$

or

$$dz_{n+1}/dt = -\sum_{j=1}^{n}(\partial f_j/\partial c)z_j;\ z_{n+1}(1) = 0 \quad (3.11)$$

Integrating and substituting the boundary condition gives

$$z_{n+1}(0) = \int_0^1 \sum_{j=1}^{n}(\partial f_j/\partial c)z_j\ dt \quad (3.12)$$

Thus $z_{n+1}(0)$ is the gradient on c. By enlarging the adjoint system of equations, there is one less integration scheme required. Integrating the x equations foward in time, then the z equations backwards in time yields all of the gradients.

3.3 THE WORKING DIFFERENTIAL EQUATIONS FOR THE CONSECUTIVE AND PARALLEL REACTION SCHEMES

In setting up the differential equations for the consecutive and parallel reaction schemes with linear sub-optimal control, it is convenient to let x_1 and x_2 be the concentrations of the raw material and desired product, x_3 be the temperature and x_4, x_5, x_6 and x_7 be equal to c_2, c_3, c_4 and c_5. The c_1 parameter is contained in the initial condition $x_3(0)$. It is easier to set down the equations and more convenient to write computer programs if certain factors are defined to simplify the equations.

For the consecutive reaction scheme, the following factors are defined

$$
\begin{aligned}
g_1 &= -x_1 x_3 \\
g_2 &= x_1 x_3 - \beta x_3^{\alpha} x_2 \\
g_3 &= \beta x_3^{\alpha} \\
g_4 &= \alpha \beta x_3^{(\alpha-1)} \\
g_5 &= \alpha(\alpha-1)\beta x_3^{(\alpha-2)} \\
g_6 &= x_3 g_1 - g_2 g_3 \\
g_7 &= c_2 g_2 + c_3 x_2 + c_4 + c_5 g_6
\end{aligned}
\qquad (3.13)
$$

$$g_8 = 1 + c_5(g_4 x_2 - x_1)$$

$$g_9 = - z_3/g_8^2$$

$$g_{10} = - z_3/g_8$$

Then

$$dx_1/dt = g_1$$

$$dx_2/dt = g_2$$

$$dx_3/dt = c_2(dx_2/dt) + c_3 x_2 + c_4 + c_5(d^2x_2/dt^2)$$

where

$$c_5(d^2x_2/dt^2) = c_5(dg_2/dt)$$

$$= c_5(x_1 - \alpha\beta x_3^{(\gamma-1)} x_2)(dx_3/dt)$$

$$+ c_5 x_3(dx_1/dt) - c_5\beta x_3^{\gamma} (dx_2/dt)$$

$$= (1 - g_8)(dx_3/dt) + c_5(g_1 x_3 - g_2 g_3)$$

and

$$dx_3/dt = c_2g_2 + c_3x_2 + c_4 + (1 - g_8)dx_3/dt + c_5(g_1x_3 - g_2g_3)$$

Thus

$$dx_3/dt = g_7/g_8$$

and the complete set of state equations for the consecutive reaction scheme is

$$\begin{aligned} dx_1/dt &= g_1 \quad ; \quad x_1(0) = 1 \\ dx_2/dt &= g_2 \quad ; \quad x_2(0) = 0 \\ dx_3/dt &= g_7/g_8 \quad ; \quad x_3(0) = c_1 \\ dx_{i+3}/dt &= 0 \quad ; \quad x_{i+3}(0) = c_{i+1} \end{aligned} \tag{3.14}$$

$$i = 1,2,3,4$$

where the controller settings c_1, c_2, c_3, c_4 and c_5 are defined by Equations 2.9 and 2.11. The set of adjoint equations for the consecutive scheme is

$$dz_1/dt = (z_1 - z_2)x_3 + g_9(c_2g_8x_3 + c_5(-x_3g_8(x_3 + g_3) + g_7));$$
$$z_1(1) = 0$$

$$dz_2/dt = z_2 g_3 + g_9(- c_2 g_3 g_8 + c_3 g_8 + c_5(g_3^2 g_8 - g_4 g_7)); \; z_2(1) = 1$$

$$dz_3/dt = (z_1 - z_2)x_1 + z_2 x_2 g_4 + g_9(c_2 g_8 (x_1 - g_4 x_2) + c_5(2g_1 - (g_4/\alpha)((\alpha - 1)g_1 + 2\alpha g_2) - x_2 g_5 g_7)); \; z_3(1) = 0 \qquad (3.15)$$

$$dz_4/dt = g_2 g_{10}; \; z_4(1) = 0$$

$$dz_5/dt = x_2 g_{10}; \; z_5(1) = 0$$

$$dz_6/dt = g_{10}; \; z_6(1) = 0$$

$$dz_7/dt = g_9(g_6 g_8 - x_2 g_4 g_7); \; z_7(1) = 0$$

The x_4, x_5, x_6 and x_7 never actually appear in the calculations and are shown only to explain z_4, z_5, z_6 and z_7.

For the parallel reaction scheme, the following factors are defined

$$\begin{aligned} g_1 &= \beta x_3^{\alpha} \\ g_2 &= x_3 + g_1 \\ g_3 &= 1 + \alpha\beta x_3^{(\alpha-1)} \\ g_4 &= 1 - c_5 x_1 \\ g_5 &= - z_3/g_4 \\ g_6 &= - x_1 g_2 \\ g_7 &= x_1 x_3 \end{aligned} \qquad (3.16)$$

Then

$$dx_1/dt = g_6$$

$$dx_2/dt = g_7$$

$$dx_3/dt = c_2(dx_2/dt) + c_3x_2 + c_4 + c_5(d^2x_2/dt^2)$$

where

$$c_5(d^2x_2/dt^2) = c_5(x_3(dx_1/dt) + x_1(dx_3/dt))$$

$$= c_5(g_6x_3 + x_1(dx_3/dt))$$

and

$$dx_3/dt = c_2g_7 + c_3x_2 + c_4 + c_5g_6x_3 + c_5x_1(dx_3/dt)$$

Thus

$$dx_3/dt = (c_2g_7 + c_3x_2 + c_4 + c_5g_6x_3)/(1 - c_5x_1)$$

and the complete set of state equations is

$$
\begin{aligned}
dx_1/dt &= g_6 \quad ; \quad x_1(0) = 1 \\
dx_2/dt &= g_7 \quad ; \quad x_2(0) = 0 \\
dx_3/dt &= (c_2 g_7 + c_3 x_2 + c_4 + c_5 g_6 x_3)/g_4; \quad x_3(0) = c_1 \\
dx_{i+3}/dt &= 0; \quad x_{i+3}(0) = c_{i+1} \\
& \quad i = 1,2,3,4
\end{aligned}
\tag{3.17}
$$

where the controller settings c_1, c_2, c_3, c_4 and c_5 are defined by Equations 2.9 and 2.11. The set of adjoint differential equations for the parallel reaction scheme is

$$
\begin{aligned}
dz_1/dt &= z_1 g_2 - z_2 x_3 - z_3/g_4^2 (c_2 x_3 \\
&\quad + c_5 (c_3 x_2 + c_4 - x_3 g_2)); \\
&\quad z_1(1) = 0 \\
dz_2/dt &= g_5 c_3; \quad z_2(1) = 1 \\
dz_3/dt &= z_1 x_1 g_3 - z_2 x_1 - g_5 (c_2 x_1 \\
&\quad - c_5 x_1 (x_3 g_3 + g_2)); \; z_3(1) = 0 \\
dz_4/dt &= g_5 g_7; \quad z_4(1) = 0 \\
dz_5/dt &= g_5 x_2; \quad z_5(1) = 0
\end{aligned}
\tag{3.18}
$$

$$dz_6/dt = g_5 \; ; \quad z_6(1) = 0$$

$$dz_7/dt = g_5 x_1 (-x_3 g_2 + c_2 g_7 + c_3 x_2 + c_4)/g_4 \; ; \quad z_7(1) = 0$$

With the complete sets of the equations for the linear sub-optimal control now in hand, it is convenient to also set down the equations for obtaining the best temperature schedules. For the consecutive reaction scheme, the equations are

$$\begin{aligned} dx_1/dt &= -ux_1 \; ; \quad x_1(0) = 1 \\ dx_2/dt &= ux_1 - \beta u^{\gamma} x_2 ; \; x_2(0) = 0 \\ dz_1/dt &= u(z_1 - z_2) ; \; z_1(1) = 0 \\ dz_2/dt &= \beta u^{\gamma} z_2 ; \quad z_2(1) = 1 \\ \text{Gradient} &= x_1(z_2 - z_1) - \gamma \beta u^{(\gamma-1)} x_2 z_2 \end{aligned} \tag{3.19}$$

and for the parallel reaction scheme, the equations are

$$
\begin{aligned}
dx_1/dt &= -(u + \beta u^{\alpha})x_1;\ x_1(0) = 1 \\
dx_2/dt &= ux_1 \qquad\qquad ;\ x_2(0) = 0 \\
dz_1/dt &= u(z_1 - z_2) + \beta u^{\alpha} z_1;\ z_1(1) = 0 \qquad (3.20) \\
dz_2/dt &= 0 \qquad ;\ z_2(1) = 1 \\
\text{Gradient} &= x_1(z_2 - z_1) + \alpha\beta u^{(\alpha-1)} z_1
\end{aligned}
$$

These equations are used in a best temperature schedule calculation, which is done as a matter of convenience to avoid the necessity of having to consult and manipulate prior calculations.

3.4 METHOD OF COMPUTATION

With the equations for obtaining the state variable trajectories and gradients now set down, the next task is to develop the means of employing these equations to obtain optimal parameter values with a digital computer. The flow chart shown in Figure 3.1 will be helpful in describing the methods used.

With a knowledge of the product yield and gradient at some basepoint of controller setting values, trials are made along the gradient vector at increasing distances from the basepoint as long as product yield is increasing. When a new trial value is less than the previous trial value, this previous value is chosen as the new basepoint, a new gradient is calculated, and the procedure is continued. If the first trial at a particular basepoint gives a smaller product yield than the basepoint value, then trials are made at progressively smaller distances from the basepoint until an improvement in yield is found. At this point, a new basepoint is chosen, a new gradient vector is calculated and the procedure is continued. The scheme has the means of stopping when the optimum appears to have been reached, and it can also be stopped after a desired number of iterations.

The data required are the kinetic parameters, the boundary conditions, the temperature bounds, an initial

estimate of the controller settings whose optimal values are sought, plus the values of certain parameters required for the steepest ascent section - H, DELTA, MMAX, FMULT and FDIV - whose meanings will shortly be explained. A counting variable, M, is set to unity. This counting variable will be tested against a maximum value, MMAX, after each iteration. Then the state equations are integrated foward in time to give the product yield value, F, corresponding to the initial estimate of the settings. With the state variable trajectories in hand, the adjoint differential equations are integrated backwards in time to give the gradients.

The integrations are performed with a Runge-Kutta-Gill routine (Lapidus, 1962) which has been programmed so that the equations are contained in subroutines while the integration steps are set up in the main program. Any time a knowledge of the equations is required, the appropriate subroutine is called. The equations derived for the consecutive and parallel schemes in Section 3.3 are programmed as subroutines. To use the main program for any new kinetic scheme, one would write a subroutine for the state equations, another subroutine for the adjoint equations, and change the data.

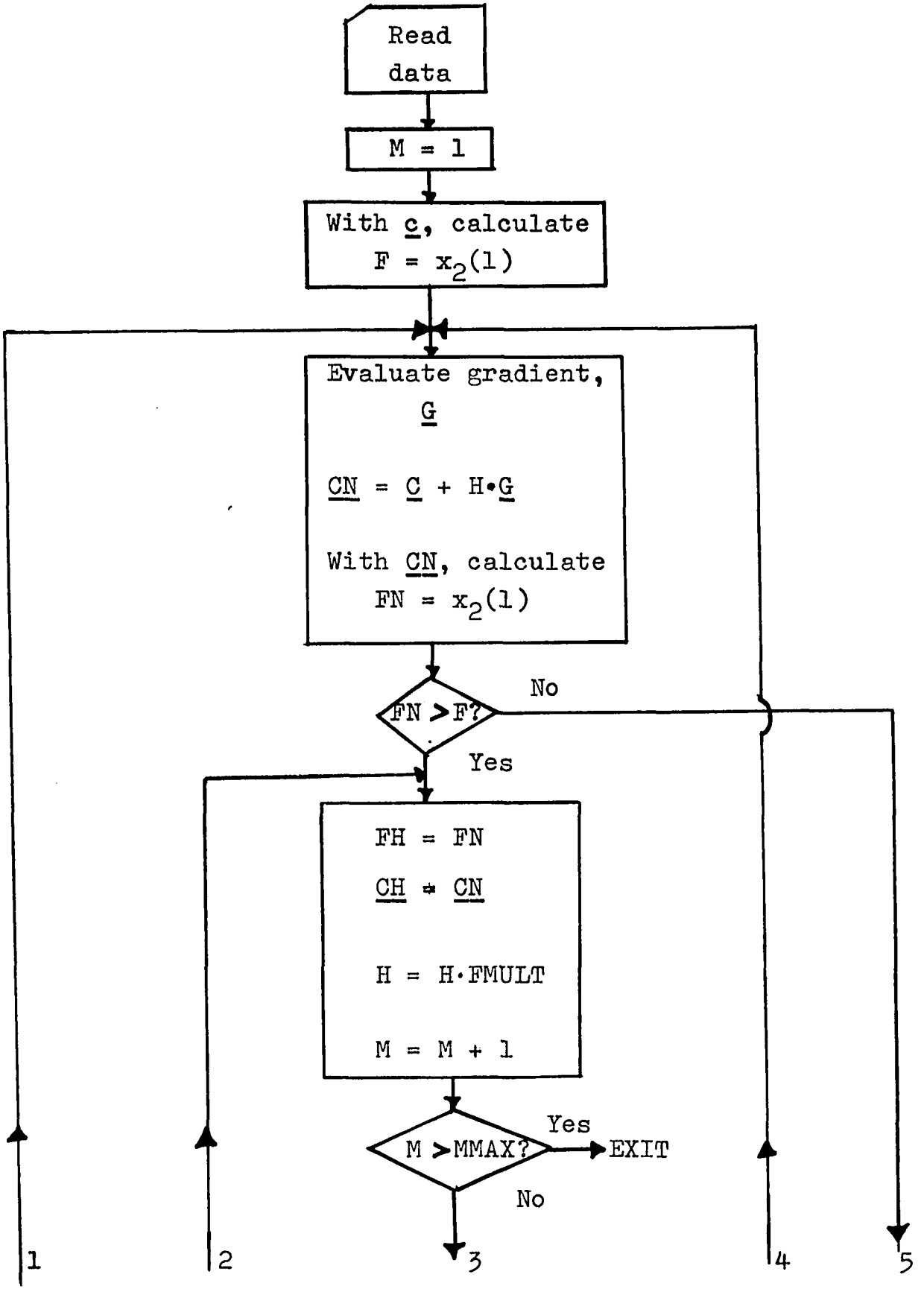
After the gradient vector, $\underline{G}$, is calculated, it is multiplied by a factor, H, and the product is added to the vector of controller settings to give new settings,

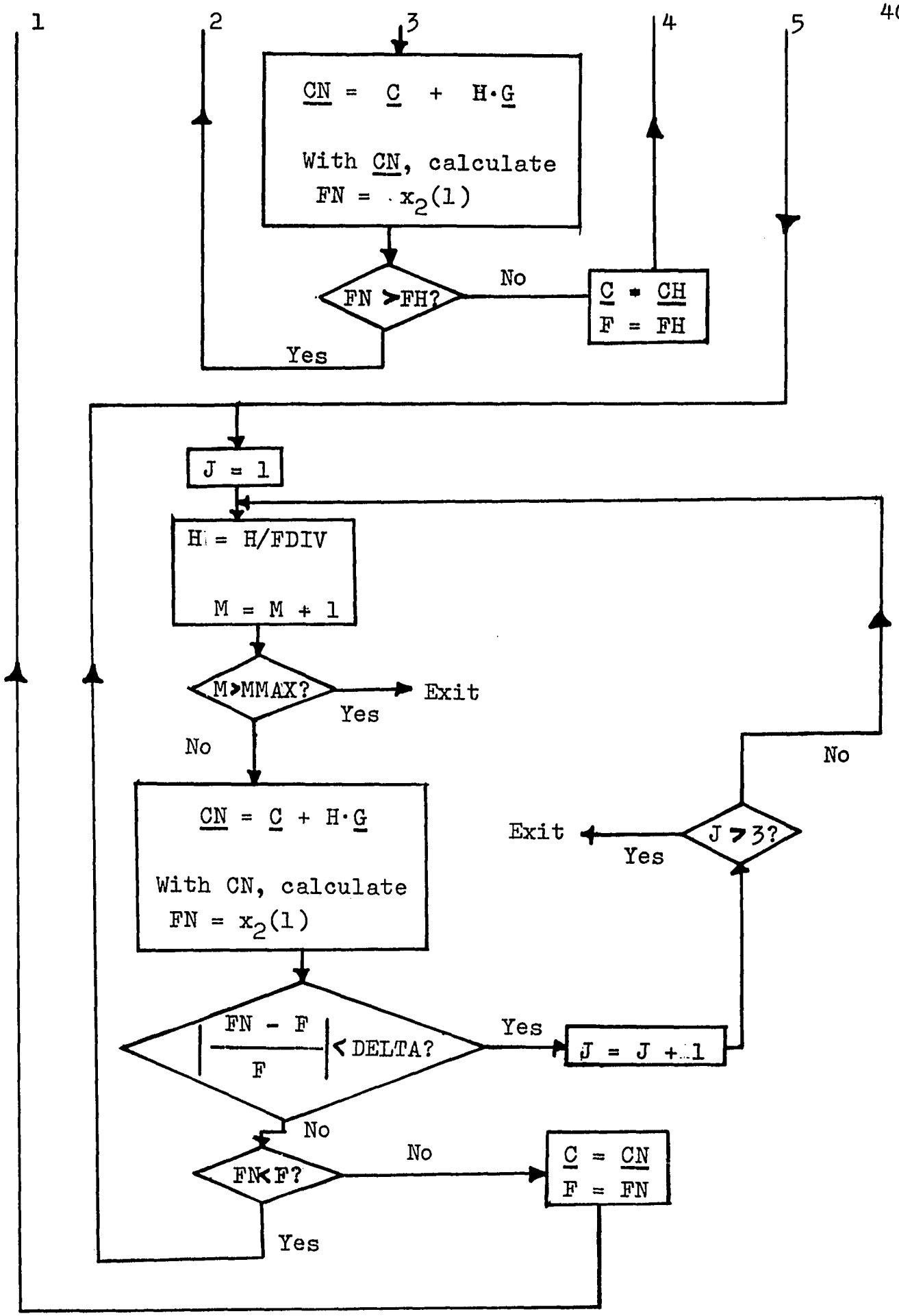
which are used to calculate (through the same Runge-Kutta-Gill scheme employed before) FN, a new value of product yield. If FN is an improvement over the basepoint value F, it is held along with its associated controller settings while H is multiplied by FMULT (always greater than unity) to give a higher value of H, which is used to calculate a new vector of settings, with which a new product yield can be obtained. The new yield is compared with the held value and H is again increased if the new value is greater than the held value of yield while the new value becomes the held value. When the new value is worse than the held value, the held settings become the new basepoint and the process is repeated, starting with a new calculation of the gradients.

If the new value for product yield is less than the basepoint value, the H is made smaller by dividing it by FDIV (always greater than unity), the smaller H value is used to calculate a new vector of settings, with which a new product yield can be obtained. If the absolute value of the difference between the new yield and the basepoint yield divided by the basepoint yield is less than DELTA, it can be assumed that the basepoint might be near a stationary point. But to be sure of this, the H is made smaller three times before an exit is made. When the absolute value is greater than DELTA, if the new

product yield is smaller than the basepoint yield, the H is continually made smaller until either an improvement is found which is then used as a new basepoint, or an exit is made either through the absolute value tester or by M exceeding MMAX.

FIGURE 3.1 STEEPEST ASCENT COMPUTATION SCHEME




1
2
3
4
5
CN = C + H·G
With CN, calculate
FN = x2(1)
FN >FH?
No
Yes
C = CH
F = FH
J = 1
H = H/FDIV
M = M + 1
M>MMAX?
Exit
Yes
No
CN = C + H·G
With CN, calculate
FN = x2(1)
J >3?
Exit
Yes
No
|FN - F / F| < DELTA?
Yes
J = J + 1
No
FN<F?
No
Yes
C = CN
F = FN

3.5 SOME DIFFICULTIES ENCOUNTERED IN OBTAINING RESULTS

The numerical scheme does not automatically yield perfect answers. It is an organized trial and error scheme and as such must be used with great care, particularly when the surfaces being climbed are difficult, as is the case with the controller settings. The input data must be chosen carefully, particularly the initial guesses of the controller settings. Also it is very useful and sometimes necessary to start climbing from different places and compare the results obtained to be sure that the true maximum has been reached. Some case histories of the course of particular calculations, chosen so as to illustrate these difficulties, are set down below.

The first studies of this work were made on the parallel reaction scheme with the kinetic parameters set at $\alpha = 2$, $\beta = \frac{1}{2}$. From the work of Evangelista and Katz (1968) it was known that the best isothermal temperature was 1.25 with an associated yield of 0.5347, and the yield obtained following the best temperature schedule was 0.5750. To test the numerical routine, DELTA was made very small (10^{-7}) and the best isothermal temperature was sought by starting with $c_1 = 1.0$ and searching for its best value while holding all the other controller settings zero throughout the run. After M had reached MMAX (set at 100)

an exit was made and at that point the value of c_1 was 1.3040 (equivalent to the best isothermal temperature) with an associated product yield of 0.5351. Thus by forcing a hill climbing routine, it was seen that slight increases can usually always be made, but at a great expense as the maximum is closely approached. For the purposes of this work the best isothermal yield of 0.535 is sufficiently accurate and therefore no further studies on the best isothermal yield were done for the consecutive and parallel reaction schemes.

Next, again for the parallel scheme with $\alpha = 2$, $\beta = \frac{1}{2}$, the optimal settings for the proportional controller were sought, starting with $c_1 = 1.3$ (the best isothermal temperature) and $c_2 = 0.0$. After a considerable number of iterations (over 200), the optimal settings were determined to be $c_1 = 0.617$, $c_2 = 2.419$ with an associated yield of 0.5586. In order to gain familiarity with the method, other trial settings were used. For example, starting with $c_1 = 0.3$, $c_2 = 4.0$ (which has an associated yield of only 0.3508) gave the optimal settings as $c_1 = 0.434$, $c_2 = 3.29$ with an associated yield of 0.5585. Thus the same maximum value can be obtained with quite different optimal controller settings. Choosing $c_1 = 0.5$, $c_2 = 3.0$ (with an associated yield of 0.5591) as the starting values led to an immediate exit from the climbing routine. Just guessing these last settings gave a yield higher than

that of any of the steepest ascent results, even though the others had taken 200 and even 300 iterations to finish. It was seen that good starting values of the controller settings are extremely important and that the optimum settings are not unique points. It was realized that an organized method of choosing good starting values of the controller settings would be extremely important to the work, as would a method for studying the shape of the contours of constant yield in the region of the optimum, in the space of the controller settings. The latter results would explain why it took so many iterations to optimize a function depending on only two parameters.

For the proportional-integral controller, starting with $c_1 = 1.3$, $c_2 = c_3 = c_4 = 0.0$, gave after 200 iterations $c_1 = 0.5876$, $c_2 = 1.33$, $c_3 = 1.21$, $c_4 = 1.45$ with an associated yield of 0.5633. These same values were taken, with c_1 truncated to 0.58, and put in for another 200 iterations to give $c_1 = 0.599$, $c_2 = -1.25$, $c_3 = 15.84$ and $c_4 = 15.438$ with an associated yield of 0.5639. Now using the original starting values (the best isothermal temperature of 1.3) and simply changing the initial value of H from 1.0 to 0.1 gave after 200 iterations $c_1 = 0.587$, $c_2 = -.0739$, $c_3 = 1.51$, $c_4 = 1.519$ with an associated yield of 0.5638. At this point, one might ask why the yield is not simply taken as being 0.564 with the understanding that there is a range of controller settings

which give this yield. The answer is that by other means, the optimal yield using a proportional-integral controller was found to be 0.572, much closer to the yield of .575 attained by following the optimal temperature schedule. Thus, in this type of work, one must beware of false optima.

All of the final results for the consecutive and parallel reaction schemes will be presented and discussed in Chapter 4. The remainder of this chapter will be devoted to describing the method of obtaining very useful starting values of controller settings, and some modifications which were made on the steepest ascent scheme to obtain certain results.

3.6 OBTAINING STARTING ESTIMATES OF THE CONTROLLER SETTINGS

In order to obtain good starting estimates of the controller settings, the optimal temperature trajectory along with the associated concentration trajectories are employed. The controller of Equation 2.13 can be written in the form

$$u = c_1' + c_2x_2 + c_3\int_0^t x_2dt + c_4t + c_5(dx_2/dt) \qquad (3.21)$$

$$\text{where } c_1' = c_1 + c_5\left(\frac{dx_2}{dt}\right)_{t=0}$$

With the information available, it is a straightfoward task to obtain least squares values of the constants for the various controller types described in Chapter 2. The u, x_2 and t are immediately available, while $\int_0^t x_2dt$ is obtained by a Simpson's rule integration and (dx_2/dt) is replaced by $f_2(\underline{x},u)$ and is easily evaluated. Thus a table is created where u is the dependent variable and x_2, $\int_0^t x_2dt$, (dx_2/dt) and t are the independent variables. With this table, a least squares calculation can be applied to each controller type. These least squares values provide excellent starting values for the steepest ascent calculation.

In developing the program to do this calculation, it was decided that it would be better to have it generate its own trajectories rather than to require them as data. This is because it would have been very time consuming to feed in hundreds of data points for each case. Also, the cases to be studied would have been limited to the optimal temperature schedule work available from others.

The main program for obtaining optimal controller settings was modified, essentially by changing from a vector of controller settings to a vector of temperature values to be optimized. All of the temperature components are originally set to the same value, usually the best isothermal temperature. After the steepest ascent section is exited from, the least squares fitting section is entered. As with the optimal controller setting program, all equations are contained in subroutines so that the main program is independent of kinetics, and it is relatively easy to change to a new reaction scheme.

Since the purpose of this program for the consecutive and parallel reaction schemes was simply to obtain least squares values, not extremely accurate optimal yield values, which were already available (Evangelista and Katz, 1968), only forty temperature points were taken. Starting with the best isothermal temperature values, it took only approximately 25 iterations per case to exit

from the steepest ascent section. Executing these programs was extremely simple and straightfoward compared to obtaining optimal controller settings for even the two term proportional controller.

3.7 MODIFICATIONS OF THE METHOD

Even with the least squares starting values available, the attempts made to find optimal settings for the three, four and five parameter controllers proved to be very difficult. The original program had the variables H, FMULT, FDIV, DELTA and MMAX (all shown in Figure 3.1) permanently built in as 1.0, 2.0, 2.0, 10^{-6} and 150, respectively. These five parameters were soon made variables for greater flexibility in controlling the program. After a great deal of trial and error, enough experience was gained so that most of the desired answers could be obtained.

To obtain the remaining results, a method was sought which would both work well and easily fit into the existing steepest ascent programs. The conjugate gradient method of Fletcher and Reeves (1964), which has quadratic convergence, was employed. As it was adapted for this work, the method follows essentially the same numerical scheme as shown in Figure 3.1, expect that the vector along which the ascent takes place is not the gradient calculated at the basepoint, but a modified direction which takes into account the gradients of previous basepoints.

The method is simply

$$\underline{P}_1 = \underline{G}_1$$

$$\underline{P}_{i+1} = \underline{G}_{i+1} + b_i \cdot \underline{P}_i \qquad (3.22)$$

where $b_i = (\underline{G}^2_{i+1}/\underline{G}^2_i)$ and $i > 1$

where $\underline{G}$ is the gradient vector and $\underline{P}$ is the vector along which the ascending is done. The initial direction is simply the gradient.

The conjugate gradient method finds the maximum for an ellipse in two iterations whereas a standard steepest ascent method, which follows the gradient, takes an infinite number of iterations. Thus it was not surprising, since it was suspected that the proportional controllers had very ridgy contours, to find that it took only approximately 20 iterations to optimize any of the proportional controllers. For the higher order controllers, the executions of the programs were still difficult, but with the experience gained previously and the more powerful conjugate gradient technique, the remaining results were obtained.

CHAPTER 4 BEST CONTROLLER SETTING VALUES

4.1 INTRODUCTION

The conclusion which may be drawn from the results is that a three-term controller can give essentially the maximum theoretical yield attained following the best temperature schedule. In the cases where proportional temperature control alone represents some improvement over the best isothermal operation but still falls noticeably short of the theoretical highest yield, it has been found that proportional plus derivative or proportional plus integral control comes very close to the theoretical maximum, and it is hardly necessary to utilize the full strength of the three-term controller. In the cases where proportional control alone comes close to the theoretical maximum, it is usually difficult and in some cases impossible to get much closer to the optimal yield by adding derivative or integral action.

The detailed results are presented in the following sections of this chapter and various studies of the results are presented in the following chapters.

4.2 CONSECUTIVE REACTIONS

If the ratio of the activation energies, $\alpha < 1$, the best schedule of temperature in the consecutive reaction schemes is to run at the maximum allowable temperature. In the cases where it is best to run at the maximum allowable temperature, there is no feedback control question to be answered. Thus all of the cases studied are for $\alpha > 1$. Table 4.1 contains the best isothermal yield, the associated best temperature and the optimal yield which results from following the best temperature schedule. Tables 4.2, 4.3 and 4.4 present both the least squares starting values and the best values of controller settings together with the associated yields for proportional, proportional-derivative and proportional-integral control. The final results are summarized in Table 4.5. Figure 4.1 compares the temperature histories for best proportional control and for the theoretical optimum; the temperature histories for the higher modes of control lie essentially on top of the theoretical optimum. Figure 4.2 presents some approximate contour plots of the control constants c_1, c_2 for the best proportional controller. It can be seen that while the set point constant, c_1, depends on both kinetic parameters, the proportionality constant, c_2, depends over most of its range only on α, the ratio of the activation energies.

Although the least squares values serve as excellent starting values for a numerical search, slight changes in controller settings can sometimes mean an enormous change in yield. This was found to be so in all of the proportional integral cases. In these cases, without good starting values a numerical search gets nowhere, and even with them the computation proves very difficult and sensitive.

TABLE 4.1 ISOTHERMAL AND OPTIMAL OPERATIONS

CONSECUTIVE REACTIONS

α	β	Best Isothermal Temperature	Best Isothermal Yield	Best Schedule Yield
2	1/2	1.003	.4773	.4905
2	1	.778	.3901	.4039
2	2	.598	.3103	.3238
5	1/2	.857	.5055	.5171
5	1	.750	.4628	.4744
5	2	.664	.4216	.4331
10	1/2	.863	.5418	.5489
10	1	.807	.5185	.5257
10	2	.756	.4956	.5027

TABLE 4.2 BEST CONTROLLER VALUES FOR CONSECUTIVE REACTIONS - PROPORTIONAL CONTROL

γ	β	c_1	c_2	Proportional Yield
2	1/2	1.97	-2.68	.4887
		(2.24)*	(-3.41)*	(.4880)**
2	1	1.57	-2.63	.4019
		(1.76)	(-3.32)	(.4013)
2	2	1.31	-3.02	.3217
		(1.40)	(-3.39)	(.3212)
5	1/2	1.15	-0.87	.5149
		(1.20)	(-0.98)	(.5146)
5	1	1.02	-0.85	.4722
		(1.03)	(-0.90)	(.4720)
5	2	0.899	-0.816	.4308
		(0.904)	(-0.845)	(.4308)
10	1/2	1.00	-0.395	.5474
		(1.01)	(-0.428)	(.5473)
10	1	0.938	-0.382	.5242
		(0.932)	(-0.379)	(.5241)
10	2	0.880	-0.381	.5012
		(0.846)	(-0.312)	(.5007)

* Values in ( ) are least squares results

** Yields in ( ) result from using least squares values

TABLE 4.3 BEST CONTROLLER VALUES FOR CONSECUTIVE REACTIONS - PROPORTIONAL - DERIVATIVE CONTROL

α	β	c_1	c_2	c_5	Prop.-Deriv. Yield
2	1/2	+0.689	-.197	+.861	.4907
		(+1.140)*	(-1.13)*	(+.56)*	(.4896)**
2	1	+.723	-.610	+.769	.4038
		(+.733)	(-.704)	(+.67)	(.4032)
2	2	+.630	-.923	+.726	.3236
		(+.722)	(-1.23)	(+.578)	(.3232)
5	1/2	+.342	+.424	+.769	.5166
		(+.342)	(+.424)	(+.769)	(.5166)
5	1	+.239	+.495	+.812	.4732
		(+.233)	(+.487)	(+.822)	(.4731)
5	2	+.206	+.437	+.821	.4316
		(+.206)	(+.437)	(+.821)	(.4316)
10	1/2	+.236	.605	.795	.5483
		(+.236)	(.605)	(.795)	(.5483)
10	1	+.156	+.663	+.857	.5244
		(+.156)	(+.663)	(+.857)	(.5244)

* Values in ( ) are least squares results.

** Yields in ( ) result from using least squares values.

TABLE 4.4 BEST CONTROL VALUES FOR CONSECUTIVE REACTIONS

PROPORTIONAL - INTEGRAL CONTROL

α	β	c_1	c_2	c_3	c_4	Prop.-Intg. Yield
2	1/2	2.59	-7.08	-3.37	+2.86	.4898
		(2.51)*	(-6.90)*	(-3.12)*	(+3.43)*	(.4407)**
2	1	2.34	-10.3	-7.70	+4.62	.4032
		(2.11)	(-9.99)	(-7.11)	(+5.86)	(.0849)
2	2	1.71	-8.38	-4.32	+2.42	.3233
		(1.62)	(-8.24)	(-4.08)	(+2.98)	(.1862)
5	1/2	1.42	-8.61	-8.48	6.45	.5168
		(1.42)	(-9.75)	(-6.48)	(7.85)	(.0305)
5	2	1.36	-11.9	-8.50	+6.67	.4314
		(1.12)	(12.3)	(-7.27)	(+7.95)	(.0331)
10	1/2	1.15	-8.78	-8.58	+7.29	.5485
		(1.13)	(-9.79)	(-5.87)	(+8.23)	(.2651)

* Values in ( ) are least squares results.

** Yields in ( ) result from using least squares values.

TABLE 4.5 SUMMARY - CONSECUTIVE REACTIONS

α	β	Best Iso.	Best Prop.	Best Prop. Deriv.	Best Prop. Integ.	Optimal
2	1/2	.477	.489	.491	.490	.491
2	1	.390	.402	.404	.403	.404
2	2	.310	.322	.324	.323	.324
5	1/2	.506	.515	.517	.517	.517
5	1	.463	.472	.473	(.472)	.474
5	2	.422	.431	.432	.431	.433
10	1/2	.542	.547	.548	.549	.549
10	1	.519	.524	.524	(.524)	.526
10	2	.496	.501	(.501)	(.501)	.503

Figures in parenthesis ( ) are estimated values.

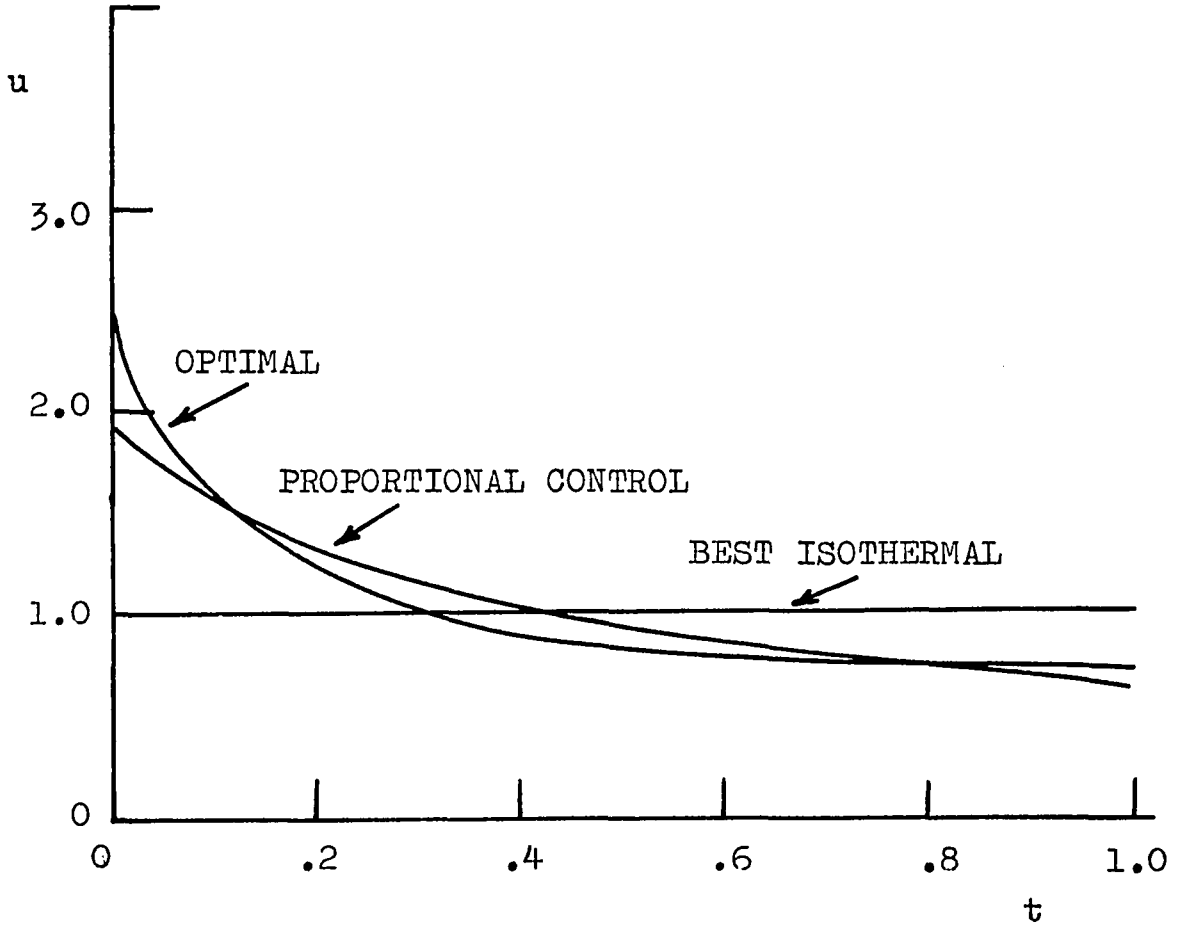


FIGURE 4.1 TEMPERATURE HISTORIES FOR CONSECUTIVE REACTIONS ($\alpha = 2, \beta = \frac{1}{2}$)

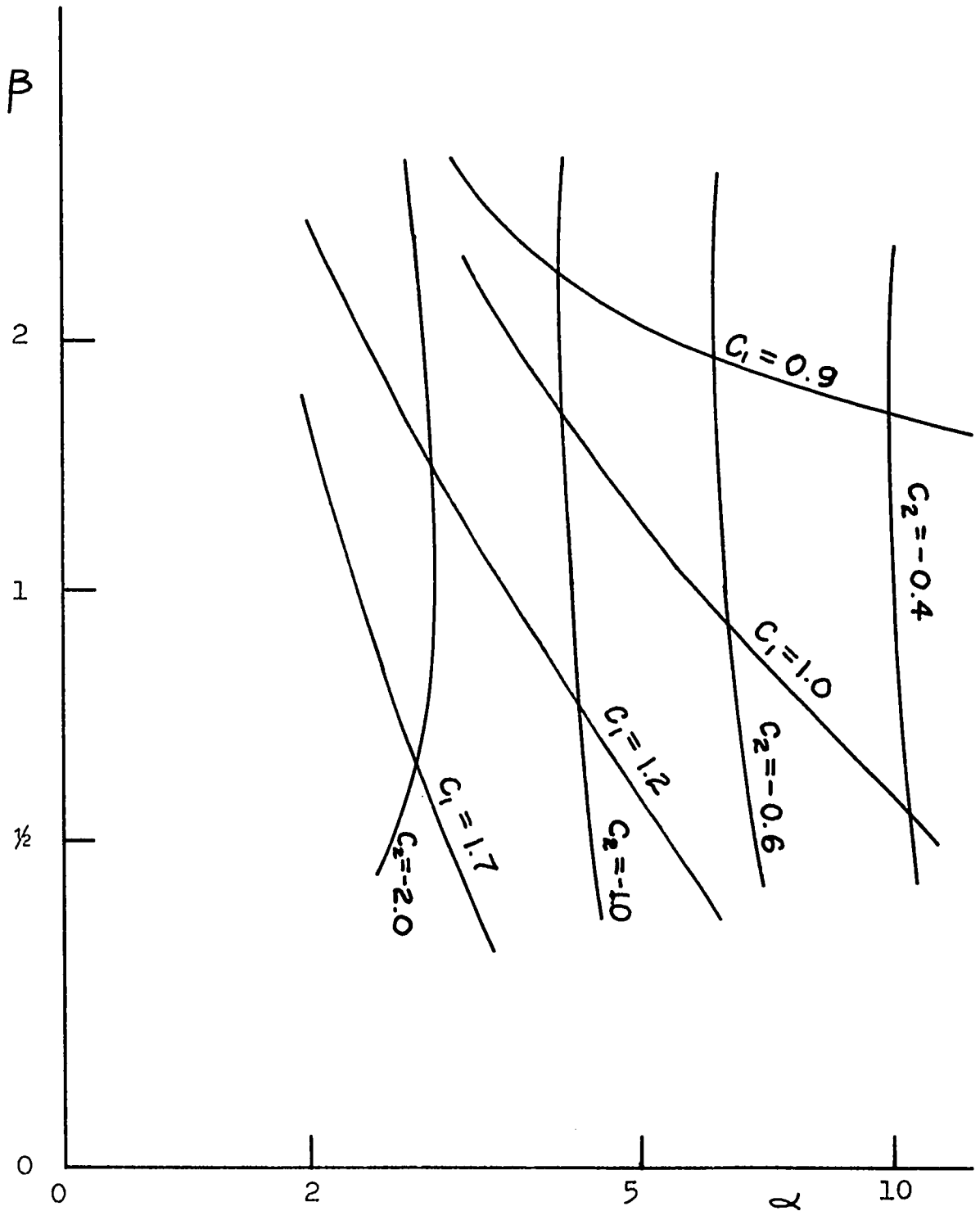


FIGURE 4.2 APPROXIMATE CONTOUR PLOTS OF BEST CONTROLLER SETTINGS FOR CONSECUTIVE REACTIONS

4.3 PARALLEL REACTIONS

Just as for the consecutive reactions, when the variable $\alpha < 1$, the best schedule of temperature in the parallel reaction schemes is to run at the maximum allowable temperature. Again there is no feedback control question to be answered. All the cases studied are for $\alpha > 1$. Table 4.6 contains the best isothermal yield, the associated best temperature, and the optimal yield which results from following the best temperature schedule. Tables 4.7, 4.8 and 4.9 present both the least squares starting values and the best values of controller settings together with the associated yields for proportional, proportional-derivative and proportional-integral control. The results are summarized in Table 4.10. Figure 4.3 compares the temperature histories for best proportional control and for the theoretical optimum; the temperature histories for the higher modes of control lie essentially on top of the theoretical optimum just as they do for the consecutive reactions. Figure 4.4 presents some approximate contour plots of the control constants c_1, c_2 for the best proportional controller. Just as for the consecutive reactions, it can be seen that while the set point constant, c_1, depends on both kinetic parameters, the proportionality constant c_2 depends over most of its range only on α, the ratio of the activation energies.

TABLE 4.6 ISOTHERMAL AND OPTIMAL OPERATIONS

PARALLEL REACTIONS

α	β	Best Isothermal Temperature	Best Isothermal Yield	Best Schedule Yield
2	1/2	1.25	.5347	.5750
2	1	0.97	.4325	.4681
2	2	0.74	.3389	.3691
5	1/2	0.91	.5253	.5479
5	1	0.83	.4787	.5010
5	2	0.71	.4358	.4558
10	1/2	0.90	.5513	.5646
10	1	0.83	.5279	.5402
10	2	0.79	.5037	.5162

TABLE 4.7 BEST CONTROLLER VALUES FOR PARALLEL REACTIONS ∴ PROPORTIONAL CONTROL

γ	β	c_1	c_2	Proportional Yield
2	1/2	.596	2.56	.5589
		(.618)*	(1.88)*	(.5516)**
2	1	.468	2.30	.4539
		(.479)	(1.75)	(.4484)
2	2	.341	2.24	.3574
		(.368)	(1.56)	(.3519)
5	1/2	.684	0.895	.5407
		(.729)	(0.616)	(.5380)
5	1	.604	0.856	.4944
		(.630)	(0.628)	(.4920)
5	2	.538	0.804	.4496
		(.564)	(0.581)	(.4475)
10	1/2	.764	0.464	.5610
		(.780)	(0.361)	(.5601)
10	1	.722	0.430	.5367
		(.731)	(0.351)	(.5359)
10	2	.668	0.405	.5128
		(.690)	(0.320)	(.5118)

* Values in ( ) are least squares results.

** Yields in ( ) result from using least squares values.

TABLE 4.8 BEST CONTROLLER VALUES FOR PARALLEL REACTIONS - PROPORTIONAL - DERIVATIVE CONTROL

α	β	c_1	c_2	c_5	Prop.-Deriv. Yield
2	1/2	+1.122	-.377	-4.61	.5691
		(+1.072)*	(-.404)*	(-4.56)*	(.5660)**
2	1	+14.66	-7.05	-24.8	.4665
		(+14.80)	(-7.05)	(-24.8)	(.4661)
5	1/2	+1.819	-.269	-1.41	.5435
		(+1.767)	(-.327)	(-1.35)	(.5415)
5	1	+1.163	.232	-.775	.4959
		(+1.127)	(.206)	(-.748)	(.4944)
5	2	+4.145	-2.55	-6.05	.4548
		(+4.145)	(-2.55)	(-6.05)	(.4548)
10	1/2	+1.269	-.0590	-.594	.5620
		(+1.251)	(-.0646)	(-.588)	(.5614)
10	1	+1.369	-.220	-.847	.5381
		(+1.366)	(-.221)	(-.846)	(.5376)
10	2	+0.984	.0731	-.405	.5133
		(+0.972)	(.0637)	(-.396)	(.5128)

* Values in ( ) are least squares results.

** Yields in ( ) result from using least squares values.

TABLE 4.9 BEST CONTROL VALUES FOR PARALLEL REACTIONS

PROPORTIONAL - INTEGRAL CONTROL

α	β	c_1	c_2	c_3	c_4	Prop. Intg. Yield
2	1/2	.517	-51.9	-12.6	38.7	.5720
		(.787)*	(-51.9)*	(-12.6)*	(38.7)*	(.5716)**
2	1	.100	-77.0	-14.4	43.5	.4656
		(.574)	(-76.8)	(-14.4)	(43.9)	(.4651)
2	2	.458	-79.8	-10.9	34.0	.3676
		(.457)	(-79.8)	(-10.9)	(34.0)	(.3676)
5	1	.556	-10.1	-2.65	+6.70	.4972
		(.665)	(-10.1)	(-2.65)	(+6.71)	(.4954)

* Values in ( ) are least squares results.

** Yields in ( ) result from using least squares values.

TABLE 4.10 SUMMARY - PARALLEL REACTIONS

γ	β	Best Iso.	Best Prop.	Best Prop. Deriv.	Best Prop. Integ.	Optimal
2	1/2	.535	.559	.569	.572	.575
2	1	.433	.454	.467	.466	.468
2	2	.339	.357	(.357)	.368	.369
5	1/2	.525	.541	.544	(.541)	.548
5	1	.479	.494	.496	.497	.501
5	2	.436	.450	.455	(.455)	.456
10	1/2	.551	.561	.562	(.561)	.565
10	1	.528	.537	.538	(.537)	.540
10	2	.504	.513	.513	(.513)	.516

Figures in parenthesis ( ) are estimated values.

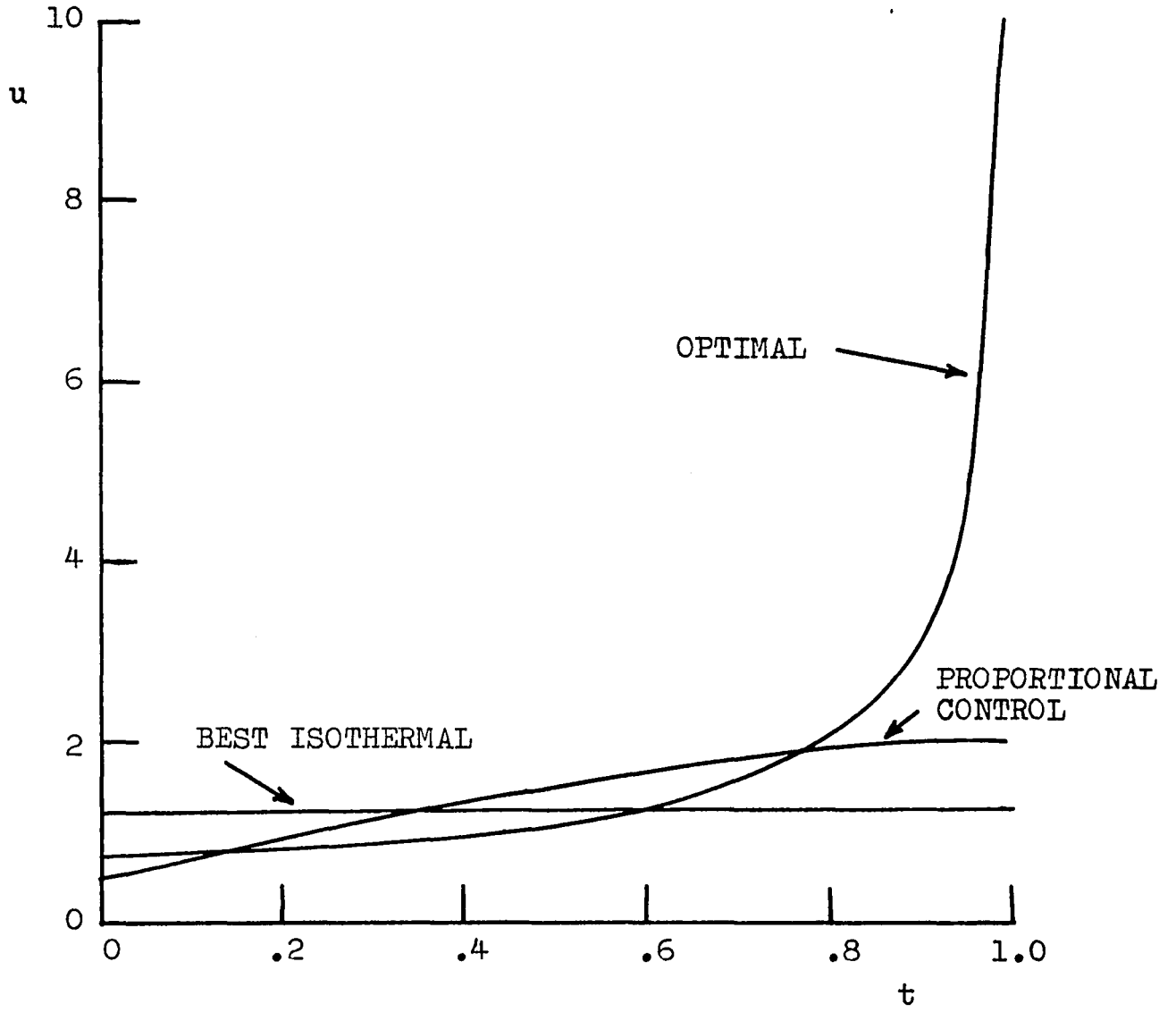


FIGURE 4.3 TEMPERATURE HISTORIES FOR PARALLEL REACTIONS ($\alpha = 2$, $\beta = \frac{1}{2}$)

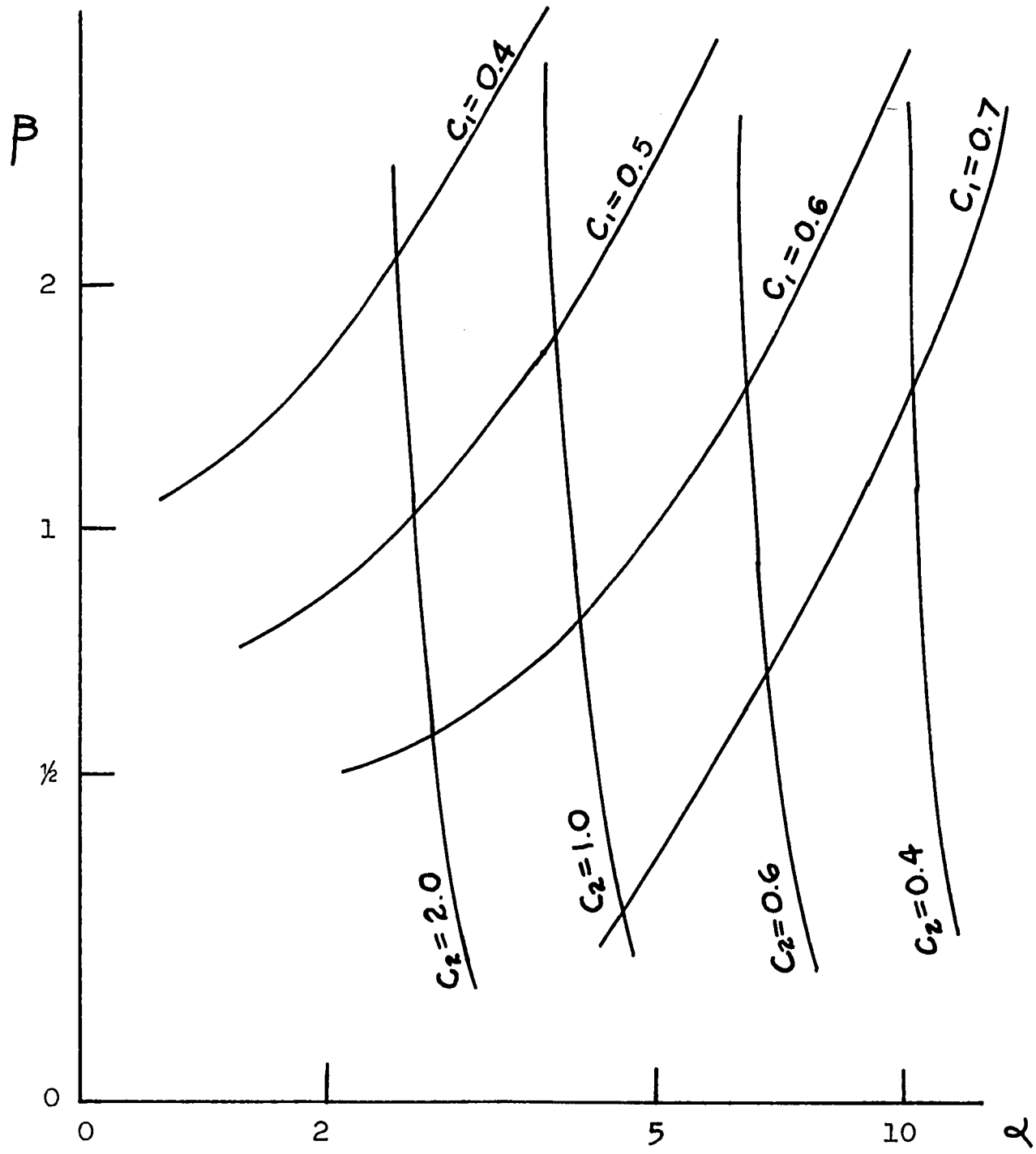


FIGURE 4.4 APPROXIMATE CONTOUR PLOTS OF BEST CONTROLLER SETTINGS FOR PARALLEL REACTIONS

CHAPTER 5 SHARPNESS OF THE OPTIMUM

5.1 INTRODUCTION

With the values of the best controller settings now calculated, it is useful to study the sensitivity of product yield to changes in these controller settings. It is important to know how much the product yield is damaged when the controller settings are not optimal. It is also useful to know the shape of the contours of constant yield, in the space of the controller settings, near the optimal point. This latter information indicates over what region the controller settings can be varied without greatly damaging the yield. Since these controller settings cannot be made more accurately than 10% or thereabouts, this information as a whole has a strong bearing on the practical utility of this kind of linear control.

An organized method has been developed to obtain this information. It is presented here along with a detailed study of the proportional controller cases for the consecutive and parallel reaction schemes.

5.2 METHOD OF SOLUTION

The equations to be studied are of the form

$$d\underline{x}/dt = \underline{f}(\underline{x},\underline{c}); \quad \underline{x}(0) = \underline{a} \tag{5.1}$$

where

$$\underline{x}(t) = [x_1(t), x_2(t), \cdots\cdots x_n(t)]$$

is the state vector, and

$$c = [c_1, c_2, \cdots\cdots c_m]$$

is the vector of controller settings, $\underline{a}$ is the vector of initial conditions and t is time. With the control parameters specified, the equations can be solved for the complete history of the state variables. To examine behavior in the neighborhood of this solution, the controller settings are perturbed

$$\begin{aligned} c_q &= c_q^* + \epsilon b_q \\ q &= 1,2,\cdots\cdots m \end{aligned} \tag{5.2}$$

and correspondingly

$$x_i = x_i^* + \epsilon \sum_q b_q y_{iq} + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r z_{iqr} + \cdots\cdots ; \quad i = 1,2,\cdots n \tag{5.3}$$

where the x_i's are expanded in a Taylor series about the original solution, which is denoted by $\underline{x}^*$ and corresponds to $\underline{c}^*$. Substituting Equations 5.2 and 5.3 into 5.1 and expanding $\underline{f}(\underline{x},\underline{c})$ in a Taylor series about $\underline{f}(\underline{x}^*,\underline{c}^*)$ gives

$$\begin{aligned}
&dx_i^*/dt + \epsilon \sum_q b_q (dy_{iq}/dt) + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r (dz_{iqr}/dt) + \cdots\cdots \\
&= f_i(\underline{x}^*,\underline{c}^*) + \\
&\sum_j \left[\left(\epsilon \sum_q b_q y_{jq} + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r z_{jqr} \cdots \right) (\partial f_i/\partial x_j) \right] \\
&\quad + \sum_q (\epsilon b_q)(\partial f_i/\partial c_q) + \\
&\quad \tfrac{1}{2} \sum_j \sum_k \left[\left(\epsilon \sum_q b_q y_{jq} + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r z_{jqr} + \cdots \right) \cdot \right. \\
&\quad \left. \left(\epsilon \sum_q b_q y_{kq} + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r z_{kqr} + \cdots \right) \cdot (\partial^2 f_i/\partial x_j \partial x_k) \right] \\
&\quad + \tfrac{1}{2} \sum_q \sum_r (\epsilon b_q)(\epsilon b_r)(\partial^2 f_i/\partial c_q \partial c_r) + \\
&\sum_j \sum_q \left[\left(\epsilon \sum_q b_q y_{jq} + \frac{\epsilon^2}{2} \sum_q \sum_r b_q b_r z_{jqr} + \cdots \right) \cdot \right. \\
&\quad \left. (\epsilon b_q)(\partial^2 f_i/\partial x_j \partial c_q) \right];
\end{aligned} \tag{5.4}$$

$$i = 1,2,\cdots n$$

where all derivatives are evaluated at the basepoint. When the terms containing equal powers of ϵ are equated, one obtains

$$
\begin{aligned}
dx_i^*/dt &= f_i(\underline{x}^*, \underline{c}^*) ; \\
dy_{iq}/dt &= \sum_j y_{jq}(\partial f_i/\partial x_j) + (\partial f_i/\partial c_q); \\
dz_{iqr}/dt &= \sum_j z_{jqr}(\partial f_i/\partial x_j) + (\partial^2 f_i/\partial c_q \partial c_r) \\
&\quad + \sum_j \sum_k y_{jq} y_{kr}(\partial^2 f_i/\partial x_j \partial x_k) \\
&\quad + 2 \sum_j y_{jq}(\partial^2 f_i/\partial x_j \partial c_q);
\end{aligned}
\tag{5.5}
$$

$$
\begin{aligned}
i &= 1,2,\cdots\cdots n \\
q &= 1,2,\cdots\cdots m \\
r &= 1,2,\cdots\cdots m
\end{aligned}
$$

where all terms up to ϵ^2 are considered. These are sufficient if the contours of constant yield are nearly quadratic. Otherwise more terms must be taken.

The set of differential equations is integrated with the initial conditions

$$
\begin{aligned}
x_i^*(0) &= a_i; \quad && i = 1,2,\cdots n \\
y_{iq}(0) &= 0; && i = 1,2,\cdots n \\
& && q = 1,2,\cdots m \\
z_{iqr}(0) &= 0; && i = 1,2,\cdots n \\
& && q = 1,2,\cdots m \\
& && r = 1,2,\cdots m
\end{aligned}
\tag{5.6}
$$

The partial derivatives are evaluated at the basepoint values of $\underline{c}^*$ and $\underline{x}^*$. Now at the final time, from Equation 5.3, for proportional control where m = 2,

$$
\begin{aligned}
x_2(1) = x_2^*(1) &+ \epsilon (b_1 y_{21} + b_2 y_{22}) \\
&+ \frac{\epsilon^2}{2}(b_1^2 z_{211} + b_1 b_2 z_{212} + b_2 b_1 z_{221} \\
&+ b_2^2 z_{222})
\end{aligned}
\tag{5.7}
$$

where $x_2(1)$ is the final product yield as a function of (ϵb_1) and (ϵb_2) which, from Equation 5.2, are

$$
\begin{aligned}
\epsilon b_1 &= (c_1 - c_1^*) \\
\epsilon b_2 &= (c_2 - c_2^*)
\end{aligned}
\tag{5.8}
$$

Thus Equation 5.7 is a quadratic relationship between product yield and the two controller settings about the basepoint.

First the quadratic must be tested, using standard techniques (Yefimov, 1964), to verify that it is an ellipse. Then the center of the ellipse and its associated yield are found. The slopes of the major and minor axes and the ratio of the length of the major to minor axis are calculated. If the basepoint values are near the optimal, then the center of the ellipse should be close to the basepoint and represent the "true" optimal. The ratio of the length of the major to minor axis gives an indication of the shape of the contours about the basepoint and indicates the amount of difficulty to be expected in following a steepest ascent scheme. If the ratio is large, not only is there difficulty in finding the optimum, but any movement in the direction of the minor axis away from the center of the ellipse will greatly damage the yield.

5.3 COMPUTATION FOR THE CONSECUTIVE AND PARALLEL REACTION SCHEMES

A main computer program was set up to integrate Equations 5.5 and then evaluate the quadratic shown in Equation 5.7. Although the consecutive and parallel reaction schemes require only two state variables (n = 2) and the proportional controller contains just two settings (m = 2), there are still fourteen differential equations to be solved. As was done in all the previous work discussed, the differential equations are programmed in subroutines so that the main program will be general with respect to reaction schemes.

Equations 5.5 are the same for both the parallel and consecutive reaction schemes, except for the partial derivatives. Therefore the equations are programmed with the partial derivatives left as unspecified variables (such as F2X2C1 for $(\partial^2 f_2/\partial x_2 \partial c_1)$). This allows the identical equations to be used for both the parallel and consecutive reaction schemes. On entering the subroutines, all partial derivatives are first evaluated (this is where the specific reaction scheme is first identified), then the fourteen differential equations are evaluated.

This method of setting up the subroutines greatly decreased coding errors and added very little to program execution time. All thirty-six results presented in this

chapter were executed in under six minutes on the I.B.M. 7040 computer.

For the consecutive reaction scheme,

$$u = c_1 + c_2 x_2$$

$$g_1 = \beta u^{\gamma}$$

$$g_2 = \gamma \beta u^{\gamma-1}$$

$$g_3 = \gamma(\gamma-1) \beta u^{\gamma-2}$$

$$f_1 = -ux_1$$

$$f_2 = ux_1 - g_1 x_2$$

$$\partial f_1/\partial x_1 = -u$$

$$\partial f_1/\partial x_2 = -c_2 x_1$$

$$\partial f_1/\partial c_1 = -x_1$$

$$\partial f_1/\partial c_2 = -x_1 x_2$$

$$\partial f_2/\partial x_1 = +u$$

$$\partial f_2/\partial x_2 = c_2 x_1 - g_1 - g_2 x_2 c_2$$

$$\partial f_2/\partial c_1 = x_1 - g_2 x_2$$

$$\partial f_2/\partial c_2 = x_1 x_2 - g_2 x_2^2$$

$$\partial^2 f_1/\partial x_1 \partial x_1 = 0$$

$$\partial^2 f_1/\partial x_1 \partial x_2 = - c_2$$

$$\partial^2 f_1/\partial x_1 \partial c_1 = - 1$$

$$\partial^2 f_1/\partial x_1 \partial c_2 = - x_2$$

$$\partial^2 f_1/\partial x_2 \partial x_2 = 0$$

$$\partial^2 f_1/\partial x_2 \partial c_1 = 0$$

$$\partial^2 f_1/\partial x_2 \partial c_2 = - x_1$$

$$\partial^2 f_1/\partial c_1 \partial c_1 = 0$$

$$\partial^2 f_1/\partial c_1 \partial c_2 = 0$$

$$\partial^2 f_1/\partial c_2 \partial c_2 = 0$$

$$\partial^2 f_2/\partial x_1 \partial x_1 = 0$$

$$\partial^2 f_2/\partial x_1 \partial x_2 = + c_2$$

$$\partial^2 f_2/\partial x_1 \partial c_1 = + 1$$

$$\partial^2 f_2/\partial x_1 \partial c_2 = + x_2$$

$$\partial^2 f_2/\partial x_2 \partial x_2 = - g_2 c_2 - g_3 x_2 c_2^2$$

$$\partial^2 f_2/\partial x_2 \partial c_1 = - g_2 - g_3 x_2 c_2$$

$$\partial^2 f_2/\partial x_2 \partial c_2 = x_1 - 2 g_2 x_2 - g_3 x_2^2 c_2$$

$$\partial^2 f_2/\partial c_1 \partial c_1 = - g_3 x_2$$

$$\partial^2 f_2/\partial c_1 \partial c_2 = - g_3 x_2^2$$

$$\partial^2 f_2/\partial c_2 \partial c_2 = - g_3 x_2^3$$

For the parallel reaction scheme,

$$u = c_1 + c_2 x_2$$

$$g_1 = - (u + \beta u^{\alpha})$$

$$g_2 = - (1 + \alpha \beta u^{\alpha-1})$$

$$g_3 = -(\alpha (\alpha - 1) \beta x_1 u^{\alpha-2})$$

$$f_1 = g_1 x_1$$

$$f_2 = u x_1$$

$$\partial f_1/\partial x_1 = g_1$$

$$\partial f_1/\partial x_2 = c_2 x_1 g_2$$

$$\partial f_1/\partial c_1 = x_1 g_2$$

$$\partial f_1/\partial c_2 = x_1 x_2 g_2$$

$$\partial f_2/\partial x_1 = u$$

$$\partial f_2/\partial x_2 = c_2 x_1$$

$$\partial f_2/\partial c_1 = x_1$$

$$\partial f_2/\partial c_2 = x_1 x_2$$

$$\partial f_1/\partial x_1\partial x_1 = 0$$

$$\partial f_1/\partial x_1\partial x_2 = c_2 g_2$$

$$\partial f_1/\partial x_1\partial c_1 = g_2$$

$$\partial f_1/\partial x_1\partial c_2 = x_2 g_2$$

$$\partial f_1/\partial x_2\partial x_2 = c_2^2 g_3$$

$$\partial f_1/\partial x_2\partial c_1 = c_2 g_3$$

$$\partial f_1/\partial x_2\partial c_2 = -x_1 + c_2 x_2 g_3$$

$$\partial f_1/\partial c_1\partial c_1 = g_3$$

$$\partial f_1/\partial c_1\partial c_2 = x_2 g_3$$

$$\partial f_1/\partial c_2\partial c_2 = x_2^2 g_3$$

$$\partial f_2/\partial x_1\partial x_1 = 0$$

$$\partial f_2/\partial x_1\partial x_2 = c_2$$

$$\partial f_2/\partial x_1\partial c_1 = 1$$

$$\partial f_2/\partial x_1\partial c_2 = x_2$$

$$\partial f_2/\partial x_2\partial x_2 = 0$$

$$\partial f_2/\partial x_2\partial c_1 = 0$$

$$\partial f_2/\partial x_2\partial c_2 = x_1$$

$$\partial f_2/\partial c_1\partial c_1 = 0$$

$$\partial f_2/\partial c_1 \partial c_2 = 0$$

$$\partial f_2/\partial c_2 \partial c_2 = 0$$

The results for the consecutive and parallel reaction schemes are summarized in Tables 5.1 and 5.2. For each set of kinetic parameters studied, the programs were run with both the least squares controller settings and the optimal controller settings. In all cases the quadratic turned out to be an ellipse. For the consecutive reaction scheme with $\alpha = 2$, $\beta = \frac{1}{2}$, the least squares estimates are $c_1 = 2.24$, $c_2 = -3.41$ with an associated yield of 0.4880. The center of the elliptical contour was found to be at $c_1 = 1.97$, $c_2 = -2.69$ and an associated yield was calculated (from the ellipse) as 0.4886. The optimal values obtained from the steepest ascent calculations are $c_1 = 1.97$, $c_2 = -2.68$ with an associated yield of 0.4887, and when these values were programmed, the center of the ellipse turned out to be identical to the optimal. Thus, running this program with the least squares estimates brings us much closer to the optimal values, and running with the optimal values gives no further improvement. This procedure could, in principle, be used in an iterative scheme to obtain optimal parameter values, but it would be extremely time consuming and costly for more than two parameters, and might be extremely sensitive to initial

guesses of the parameters.

The slopes presented in Tables 5.1 and 5.2 are of the major axis and the ratios are of the lengths of the major to minor axes. Since the ratios are large, it is expected that a steepest ascent calculation will take many iterations to reach the optimum, and movement away from the optimum along the minor axis will greatly damage the yield, whereas movement along the major axis can take place without greatly damaging the yield. Figures 5.1 and 5.2 present typical contours for the consecutive and parallel reaction schemes at $\alpha = 2$, $\beta = \frac{1}{2}$. It can be seen that, near the optimum, the quadratic gives a good estimate of the true yield. The yield may be seen to be quite insensitive to changes in the controller settings along the major axis. The fall off from the maximum is of course much sharper for changes along the minor axis, and in this direction a change of 10% or thereabouts might well decrease the yield by one or two percent.

TABLE 5.1 CONTOUR CALCULATIONS - CONSECUTIVE REACTIONS

		Starting Values			Estimated Optimum				
γ	β	c_1	c_2	Yield	c_1	c_2	Yield	Major Axis Slope (c_2/c_1)	Axis Length Ratio
2	$\frac{1}{2}$	2.24[a]	-3.41	.4880	1.97	-2.69	.4886	-2.6	16.8
		1.97[b]	-2.68	.4887	1.97	-2.67	.4887	-2.6	15.8
2	1	1.76	-3.32	.4013	1.56	-2.65	.4018	-3.1	18.6
		1.57	-2.63	.4018	1.56	-2.66	.4018	-3.1	18.7
2	2	1.40	-3.39	.3212	1.21	-2.63	.3218	-3.9	22.4
		1.31	-3.02	.3217	1.22	-2.66	.3218	-3.9	21.2
5	$\frac{1}{2}$	1.20	-0.98	.5146	1.14	-0.84	.5148	-2.9	10.0
		1.15	-0.87	.5148	1.14	-0.84	.5148	-2.9	9.9
5	1	1.03	-0.90	.4720	1.01	-0.82	.4722	-3.2	10.4
		1.02	-0.85	.4722	1.01	-0.82	.4722	-3.2	10.5
5	2	0.90	-0.85	.4308	0.90	-0.80	.4308	-3.5	11.2
		0.90	-0.82	.4308	0.90	-0.80	.4308	-3.5	11.2
10	$\frac{1}{2}$	1.01	-0.43	.5473	1.00	-0.39	.5474	-2.9	8.5
		1.00	-0.40	.5474	1.00	-0.39	.5474	-2.9	8.5
10	1	0.93	-0.38	.5241	0.94	-0.39	.5242	-3.0	8.8
		0.94	-0.38	.5242	0.94	-0.39	.5242	-3.0	8.8
10	2	0.85	-0.31	.5007	0.88	-0.39	.5012	-3.1	9.0
		0.88	-0.38	.5012	0.88	-0.38	.5012	-3.2	9.1

a First values are least squares estimates.

b Second values are optimum settings.

TABLE 5.2 CONTOUR CALCULATIONS - PARALLEL REACTIONS

		Starting Values			Estimated Optimum				
γ	β	c_1	c_2	Yield	c_1	c_2	Yield	Major Axis Slope (c_2/c_1)	Axis Length Ratio
2	$\frac{1}{2}$	.618[a]	1.88	.5516	.592	2.68	.5606	-6.4	7.1
		.596[b]	2.56	.5589	.568	2.65	.5590	-7.9	6.5
2	1	.479	1.75	.4484	.460	2.48	.4555	-8.3	8.7
		.468	2.30	.4539	.446	2.42	.4540	-9.9	8.1
2	2	.368	1.56	.3519	.351	2.33	.3590	-10.9	11.2
		.341	2.24	.3574	.330	2.29	.3574	-13.7	10.5
5	$\frac{1}{2}$	.729	.616	.5380	.687	.924	.5409	-4.4	7.3
		.684	.895	.5407	.675	.933	.5408	-4.3	7.1
5	1	.630	.628	.4920	.604	.894	.4947	-4.9	7.9
		.604	.856	.4944	.596	.889	.4944	-4.8	7.7
5	2	.564	.581	.4475	.535	.841	.4498	-5.4	8.7
		.538	.804	.4496	.529	.842	.4496	-5.3	8.4
10	$\frac{1}{2}$	.780	.361	.5601	.767	.465	.5610	-3.7	7.3
		.764	.464	.5610	.765	.461	.5609	-3.5	7.3
10	1	.731	.351	.5359	.719	.451	.5368	-3.9	7.6
		.722	.430	.5367	.718	.444	.5367	-3.7	7.5
10	2	.690	.320	.5118	.674	.438	.5129	-4.2	7.9
		.668	.405	.5124	.671	.443	.5128	-4.0	7.9

a First values are least squares estimates.

b Second values are optimum settings.

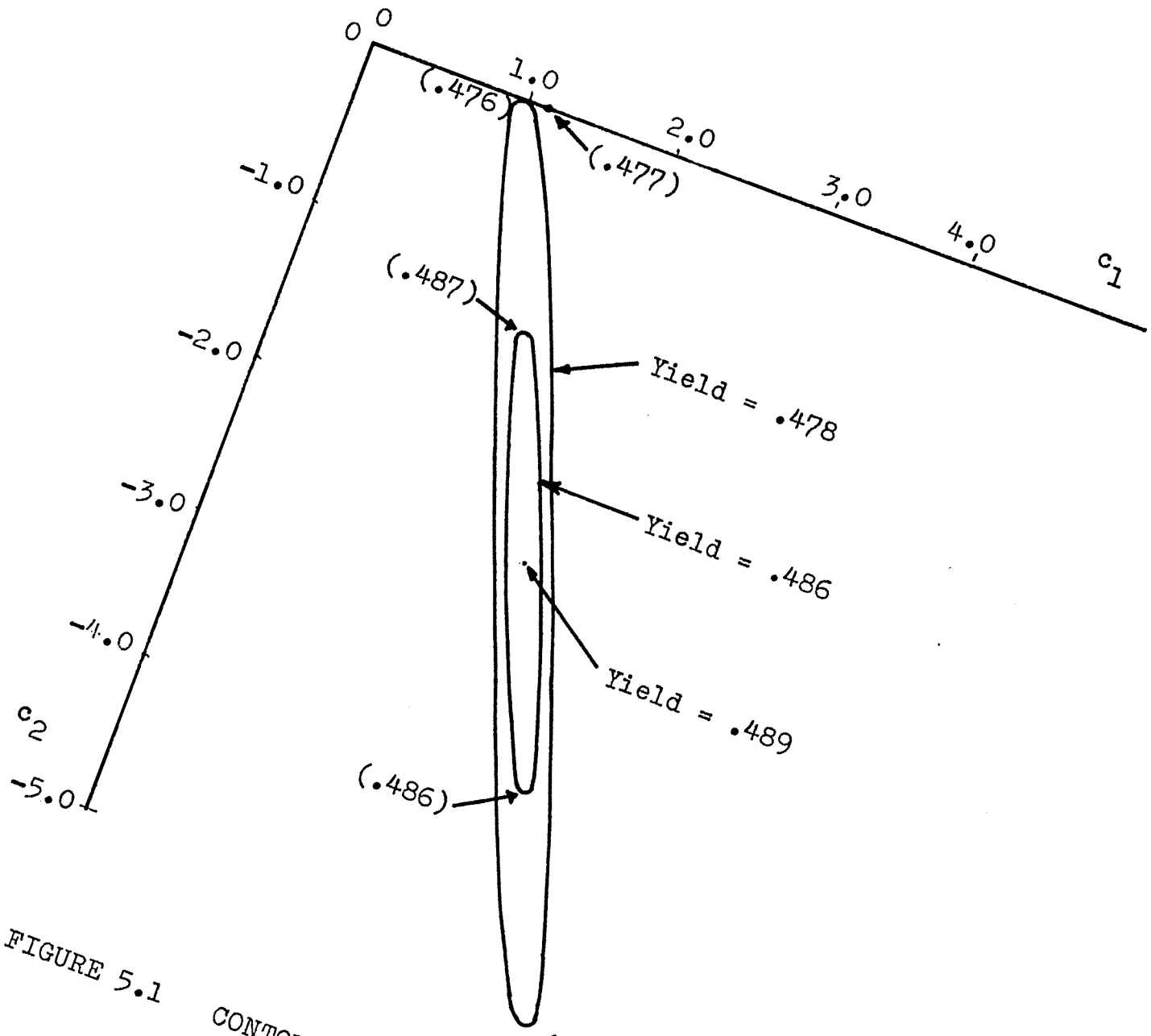


FIGURE 5.1 CONTOURS OF CONSTANT YIELD FOR THE CONSECUTIVE REACTION SCHEME ($\alpha = 2, \beta = \frac{1}{2}$)

* All values in () are found by integration

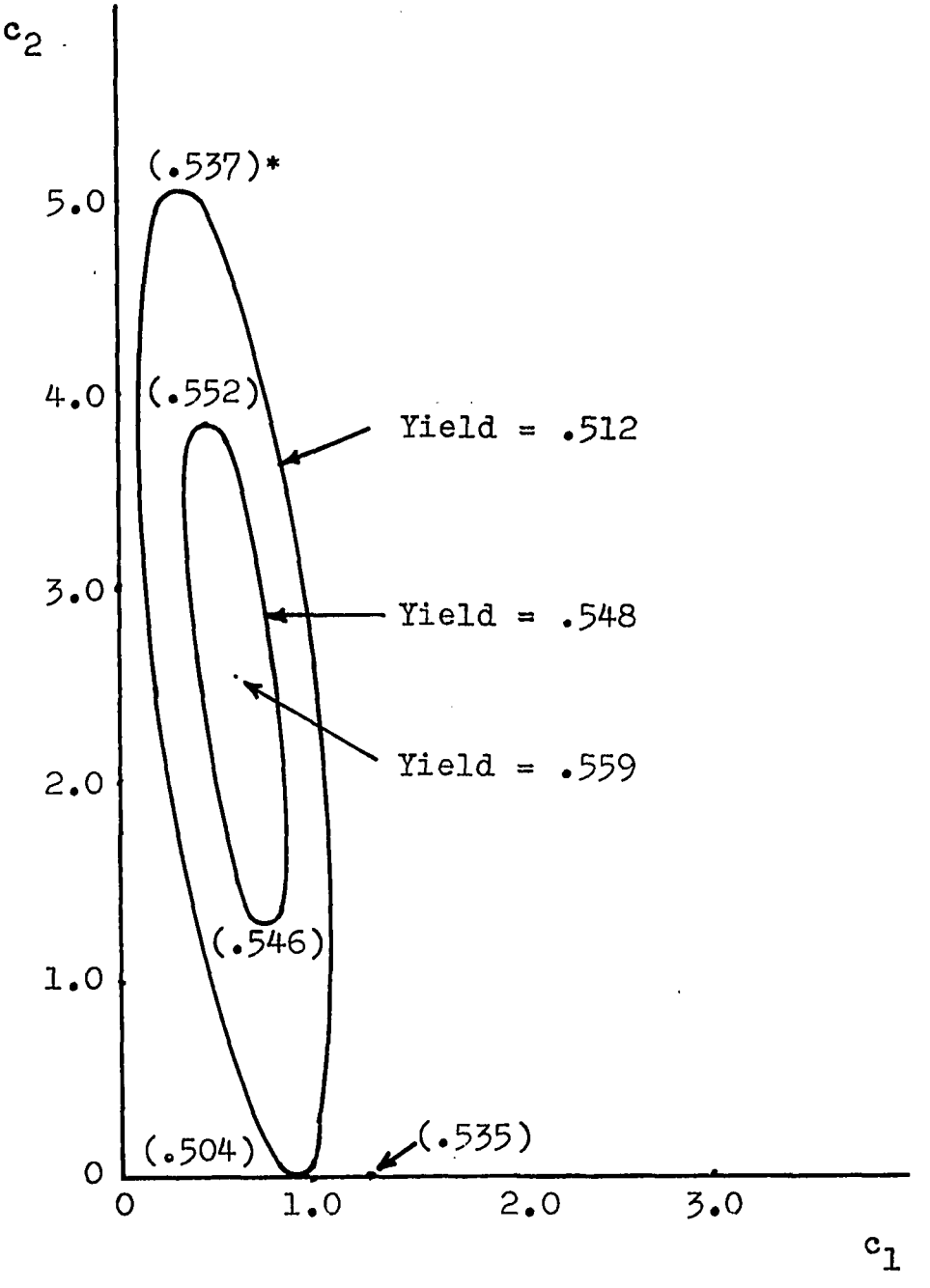


FIGURE 5.2 CONTOURS OF CONSTANT YIELD FOR THE PARALLEL REACTION SCHEME ($\alpha = 2, \beta = \frac{1}{2}$)

* All values in () are found by integration

CHAPTER 6 STUDIES OF THE EFFECTS OF DISTURBANCES ON REACTOR PERFORMANCE

6.1 INTRODUCTION

With the values of the best controller settings calculated and studies made on the sensitivity of product yield to changes in these controller settings, it is now useful to study the effects of disturbances on reactor performance. Since the state of the batch reactor is time varying, and a particular charge is reacted only for a specified fixed time interval (the total batch time), the standard stability analysis, of the kind used for continous stirred tank reactors, does not apply. Firstly, there is no steady state about which to study the effects of perturbations. Secondly, a finite disturbance may be increasing or decreasing as the batch progresses in time, yet not either grow beyond bounds or go to zero, because the reactor is stopped after the total batch time is reached. Thus for this work, the basepoint to be considered is the disturbance free state of the reactor as it progresses in time, and there is to be a measure of relative stability of various types of operation rather than a specific criterion of stability.

The mathematical methods applied here are essentially those of linearized stability analysis. That is, the differential equations describing the evolution of the system in time are linearized about the values of the variables attained at successive times. At these successive times, the eigenvalues of the resulting linear equations are examined. These eigenvalues change with time, and the relative local stability of the system is analyzed in terms of the signs and magnitudes of their real parts. This mode of analysis does not of course carry the same mathematical weight in studying the stability of a trajectory as it does in studying the stability of an equilibrium point. It is offered here simply as a means of attaining valuable intuitive insight into the behavior of the system trajectories under outside disturbances. In the study that follows, all comparisons of stability and instability between two systems, or between two operating times of the same system, are simply comparisons of the signs and magnitudes of the real parts of the appropriate eigenvalues.

6.2 METHOD OF SOLUTION

The equations to be studied are of the form

$$dx_i/dt = f_i(\underline{x}); \quad i = 1,2,\cdots n \qquad (6.1)$$

$$0 \leq t \leq 1$$

where temperature is considered to be one of the state variables. If $\underline{x}(t)$ is perturbed about some basepoint $\underline{x}^*(t)$,

$$x_i(t) = x_i^*(t) + \delta x_i; \qquad (6.2)$$

$$i = 1,2,\cdots n$$

and Equation 6.2 is substituted into Equation 6.1, with the f_i's being expanded in a Taylor series, then

$$dx_i^*/dt + d\delta x_i/dt = f_i(\underline{x}^*) + \sum_{j=1}^{n} (\partial f_i/\partial x_j)\delta x_j + \cdots\cdots \qquad (6.3)$$

with the partial derivatives evaluated at the basepoint. Thus the set of linearized differential equations about the basepoint $\underline{x}^*$ is

$$d\delta x_i/dt = \sum_{j=1}^{n} (\partial f_i/\partial x_j)\delta x_j \qquad (6.4)$$

For the consecutive and parallel reaction schemes being studied, there are three state variables, including temperature. It is convenient to define a matrix, A, as

$$\begin{bmatrix} a_1 & a_2 & a_3 \\ a_4 & a_5 & a_6 \\ a_7 & a_8 & a_9 \end{bmatrix} = \begin{bmatrix} \partial f_1/\partial x_1 & \partial f_1/\partial x_2 & \partial f_1/\partial x_3 \\ \partial f_2/\partial x_1 & \partial f_2/\partial x_2 & \partial f_2/\partial x_3 \\ \partial f_3/\partial x_1 & \partial f_3/\partial x_2 & \partial f_3/\partial x_3 \end{bmatrix} \qquad (6.5)$$

where it is seen that the matrix coefficients are time varying. Now the linearized equations can be written as

$$d\delta \underline{x}/dt = A(t)\delta \underline{x} \qquad (6.6)$$

It is the eigenvalues of matrix A(t) which indicate the relative stability of the various modes of operation. The eigenvalues are time varying, and must be studied as the batch progresses in time. Because of the fixed batch time, even if all the eigenvalues have negative real parts, a disturbance imposed during the reaction time interval cannot be reduced to zero. If a positive real part appears in one of the eigenvalues, a disturbance may be enlarged, but will not grow beyond bounds. Thus the

value of the largest real part of the set of eigenvalues, and the time at which it occurs, give a measure of stability rather than a yes-or-no criterion, because the disturbances remain bounded. If a disturbance is decreased to 90% of its original value or increased to 110%, there is perhaps no problem. But if a disturbance can be amplified to 500% of its original value, there is clearly cause for practical concern.

The eigenvalues of matrix A can be determined by solving the cubic equation

$$\lambda^3 + b_1\lambda^2 + b_2\lambda + b_3 = 0 \qquad (6.7)$$

where

$$\begin{aligned} b_1 &= -(a_1 + a_5 + a_9) \\ b_2 &= (a_1a_5 + a_1a_9 + a_5a_9 - a_6a_8 - a_2a_4 - a_3a_7) \\ b_3 &= (a_1a_6a_8 + a_2a_4a_9 + a_3a_5a_7 - a_1a_5a_9 - a_2a_6a_7 - a_3a_4a_9) \end{aligned} \qquad (6.8)$$

Before solving the cubic, the Routh-Hurwitz criterion is applied. This test (Wylie, 1960) is for the existence of roots with non-negative real parts. If

$$b_1 > 0, \quad b_2 > 0, \quad \text{and} \quad b_1b_2 > b_3$$

then all eigenvalues have negative real parts. It is unneces-

sary to solve the cubic unless a positive real part is indicated by applying the Routh-Hurwitz criterion. In that case, the cubic is solved using a standard set of formulas (Perry, 1962) to study the size of the positive real part.

For the consecutive reactions, the following factors are defined

$$
\begin{aligned}
g_1 &= \beta x_3^{\alpha} \\
g_2 &= \beta x_3^{\alpha+1} \\
g_3 &= \beta x_3^{\alpha-1} \\
g_4 &= 1 - c_5 x_1 + \alpha g_3 c_5 x_2 \\
g_5 &= x_1(x_3 c_2 - g_2 c_5 - c_5 x_3^2) \\
&\quad + x_2(g_1(g_1 c_5 - c_2) + c_3) + c_4
\end{aligned}
\tag{6.9}
$$

and the components of the matrix, A, are

$$
\begin{aligned}
a_1 &= -x_3 \\
a_2 &= 0 \\
a_3 &= -x_1 \\
a_4 &= x_3
\end{aligned}
$$

$$
\begin{aligned}
a_5 &= -g_1 \\
a_6 &= x_1 - \alpha g_3 x_2 \\
a_7 &= (c_2 x_3 - g_2 c_5 - c_5 x_3^2)/g_4 \\
&\quad + c_5 g_5 / g_4^2 \\
a_8 &= (g_1 (g_1 c_5 - c_2) + c_3)/g_4 \\
&\quad - \alpha g_3 c_5 g_5 / g_4^2 \\
a_9 &= (x_1 (c_2 - (\alpha + 1) g_1 c_5 - 2 c_5 x_3) \\
&\quad + x_2 (2 g_1 g_3 c_5 - c_2 \alpha g_3))/g_4 \\
&\quad - (\alpha (\alpha - 1) g_3 c_5 x_2 g_5)/(g_4^2 x_3)
\end{aligned}
\tag{6.10}
$$

For the parallel reactions, the following factors are defined.

$$
\begin{aligned}
g_1 &= x_3 + \beta x_3^{\alpha} \\
g_2 &= 1 + \alpha \beta x_3^{\alpha - 1} \\
g_3 &= (1 - x_1 c_5)^{-1} \\
g_4 &= x_1 (c_2 x_3 - c_5 x_3 g_1) + x_2 c_3 \\
&\quad + c_4
\end{aligned}
\tag{6.11}
$$

and the components of matrix, A, are

$$
\begin{aligned}
a_1 &= -g_1 \\
a_2 &= 0 \\
a_3 &= -x_1 g_2 \\
a_4 &= x_3 \\
a_5 &= 0 \\
a_6 &= x_1 \\
a_7 &= g_3(c_2 x_3 - c_5 x_3 g_1) + g_3^2 g_4 c_5 \\
a_8 &= g_3 c_3 \\
a_9 &= g_3 x_1(c_2 - c_5(x_3 g_2 + g_1))
\end{aligned}
\qquad (6.12)
$$

The state differential equations for the consecutive and parallel reaction schemes have already been presented in Section 3.3.

For the cases with proportional control, temperature is replaced by $(c_1 + c_2 x_2)$ so that there are only two state variables. The matrix A is redefined as

$$
\begin{bmatrix} a_1 & a_2 \\ a_3 & a_4 \end{bmatrix} = \begin{bmatrix} \partial f_1/\partial x_1 & \partial f_1/\partial x_2 \\ \partial f_2/\partial x_1 & \partial f_2/\partial x_2 \end{bmatrix} \qquad (6.13)
$$

The eigenvalues of this matrix can be determined by solving the quadratic equation

$$\lambda^2 + b_1 \lambda + b_2 = 0 \qquad (6.14)$$

where

$$\begin{aligned} b_1 &= (- a_1 - a_4) \\ b_2 &= a_1 a_4 - a_2 a_3 \end{aligned} \qquad (6.15)$$

The Routh-Hurwitz criterion for a quadratic is simply that $b_1 > 0$ and $b_2 > 0$ for both eigenvalues to have negative real parts. If a positive real part is indicated, then the quadratic is solved for the eigenvalues so that the magnitude of the positive real part can be studied.

For the consecutive reaction scheme with proportional control,

$$\begin{aligned} f_1 &= -x_1(c_1 + c_2 x_2) \\ f_2 &= x_1(c_1 + c_2 x_2) - \beta(c_1 + c_2 x_2)^{\alpha} x_2 \\ a_1 &= -(c_1 + c_2 x_2) \\ a_2 &= -c_2 x_1 \\ a_3 &= (c_1 + c_2 x_2) \end{aligned} \qquad (6.16)$$

$$a_4 = c_2x_1 - \beta(c_1 + c_2x_2)^{\alpha} - \gamma\beta(c_1 + c_2x_2)^{(\alpha-1)} c_2x_2$$

For the parallel reaction scheme with proportional control,

$$\begin{aligned}
f_1 &= -\left((c_1 + c_2x_2) + \beta(c_1 + c_2x_2)^{\alpha} \right)x_1 \\
f_2 &= + x_1(c_1 + c_2x_2) \\
a_1 &= -\left((c_1 + c_2x_2) + \beta (c_1 + c_2x_2)^{\alpha} \right) \\
a_2 &= -\left(1 + \gamma\beta(c_1 + c_2x_2)^{(\alpha-1)} x_1c_2 \right) \\
a_3 &= c_1 + c_2x_2 \\
a_4 &= x_1c_2
\end{aligned} \tag{6.17}$$

6.3 COMPUTATION FOR THE CONSECUTIVE AND PARALLEL REACTION SCHEMES

The computer programs were set up in a manner similar to the previous work. A main program controls the flow and calls to one subroutine, which contains the equations, whenever it requires these for the integration. A second subroutine contains the matrix, A, just discussed, and this subroutine is entered at certain time intervals to evaluate the matrix and study its eigenvalues. All of the optimal controller values, presented in Tables 4.2, 4.3, 4.4, 4.7, 4.8 and 4.9, were studied. A summary of the results is presented in Tables 6.1, 6.2 and 6.3 for the consecutive reactions, and Tables 6.4, 6.5 and 6.6 for the parallel reactions.

For the consecutive reactions with proportional control, the results are presented in Table 6.1. They show that although an eigenvalue usually contains a positive real part, its size is small and it would not be expected that a disturbance would be enormously amplified. The worst case seems to be at $\alpha = 2$, and $\beta = 1$. The effect of approximately a 0.35 positive real part over about 60% of the batch time gives $e^{.35(.6)} = 1.23$, which indicates that a disturbance could be enlarged to about 125% of its original value. The results for proportional-integral control, presented in Table 6.2, indicate that these controllers have a much greater tendency to enlarge a disturb-

TABLE 6.1 STABILITY STUDY FOR CONSECUTIVE REACTIONS WITH PROPORTIONAL CONTROL

		Largest Real Part of Eigenvalues at Various Times					
α	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	n*	n	.20	.28	.31	.33
2	1	n	.06	.29	.36	.37	.35
2	2	n	.03	.27	.35	.35	.31
5	$\frac{1}{2}$	n	n	.08	.13	.13	.13
5	1	n	n	.08	.13	.14	.13
5	2	n	n	.08	.13	.14	.14
10	$\frac{1}{2}$	n	n	.04	.06	.07	.06
10	1	n	n	.04	.06	.07	.07
10	2	n	n	.04	.07	.07	.07

* n means that all eigenvalues have negative real parts.

TABLE 6.2 STABILITY STUDY FOR CONSECUTIVE REACTIONS WITH PROPORTIONAL INTEGRAL CONTROL

		Largest Real Part of Eigenvalues at Various Times					
γ	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	4.4	2.2	1.7	1.5	1.5	1.6
2	1	6.0	2.6	2.0	1.8	1.8	2.0
2	2	n*	2.2	1.7	1.5	1.5	1.6
5	$\frac{1}{2}$	n	2.3	2.0	1.8	1.7	1.6
5	2	n	2.6	2.2	2.0	2.0	3.3
10	$\frac{1}{2}$	n	2.3	2.0	1.9	1.9	1.9

* n means all eigenvalues have negative real parts.

TABLE 6.3 STABILITY STUDY FOR CONSECUTIVE REACTIONS WITH PROPORTIONAL DERIVATIVE CONTROL

		Largest Real Part of Eigenvalues at Various Times					
γ	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	150	n*	n	n	n	n
2	1	61	n	n	n	n	n
2	2	50	n	n	n	n	n
5	$\frac{1}{2}$	35	n	n	n	n	n
5	1	42	n	n	n	n	n
5	2	53	n	n	n	n	n
10	$\frac{1}{2}$	36	n	n	n	n	n
10	1	66	n	n	n	n	n

* n means all eigenvalues have negative real parts.

ance than the proportional controllers, and from the point of view of stability, addition of integral action makes the situation worse. The proportional-derivative action controller studies, presented in Table 6.3, show that a positive real part appears only at the very beginning of the batch, that it is enormous, and rapidly decreases with time. An explanation for this behavior is that the consecutive reaction scheme has an optimal temperature profile which decreases with time. At $t = 0$, temperature is at its maximum value, and decreasing rapidly. The reaction rates are extremely fast and changing rapidly. Thus the rate term, in the proportional-derivative controller, is changing rapidly, causing a great sensitivity to disturbances only at the very beginning of the batch. Except at the beginning of the batch, proportional-derivative action is better than proportional action alone.

The results for the parallel reaction schemes with proportional control are presented in Table 6.4. Although some positive real parts appear early in the batch, they quickly decrease, and it is expected that the parallel reactions with proportional control will be well behaved. The proportional-integral and proportional-derivative cases are presented in Tables 6.5 and 6.6. It can be seen that the positive real parts are relatively large and appear throughout the batch. Thus the addition of

TABLE 6.4 STABILITY STUDY FOR PARALLEL REACTIONS WITH PROPORTIONAL CONTROL

		Largest Real Part of Eigenvalues at Various Times					
α	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	1.5	.33	n*	n	n	n
2	1	1.2	.34	n	n	n	n
2	2	1.2	.43	n	n	n	n
5	$\frac{1}{2}$	.07	n	n	n	n	n
5	1	.09	n	n	n	n	n
5	2	.09	n	n	n	n	n
10	$\frac{1}{2}$	n	n	n	n	n	n
10	1	n	n	n	n	n	n
10	2	n	n	n	n	n	n

* n means that all eigenvalues have negative real parts.

TABLE 6.5 STABILITY STUDY FOR PARALLEL REACTIONS WITH PROPORTIONAL INTEGRAL CONTROL

		Largest Real Part of Eigenvalues at Various Times					
α	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	18.	15.	12.	8.8	7.0	8.2
2	1	26.	22.	18.	14.	10.	10.
2	2	27.	23.	19.	15.	11.	10.
5	1	3.8	3.7	3.7	3.9	4.9	7.0

TABLE 6.6 STABILITY STUDY FOR PARALLEL REACTIONS WITH PROPORTIONAL **DERIVATIVE** CONTROL

		Largest Real Part of Eigenvalues at Various Times					
γ	β	t = 0.0	0.2	0.4	0.6	0.8	1.0
2	$\frac{1}{2}$	1.6	1.9	2.5	3.3	4.3	4.2
2	1	1.5	1.8	2.4	3.5	6.7	2.5
5	$\frac{1}{2}$	1.0	1.1	1.3	1.5	1.8	2.0
5	1	.73	.80	.88	.97	1.05	1.08
5	2	1.1	1.3	1.7	2.3	3.9	1.8
10	$\frac{1}{2}$	.64	.68	.73	.80	.89	.96
10	1	.70	.76	.85	.97	1.2	1.4
10	2	.47	.48	.50	.53	.55	.57

integral or derivative action in the parallel reaction cases harms the stability.

For an illustrative study of the effect of different controller settings on the stability characteristics of the system, it was decided to try some of the false optimum values obtained (and presented in Chapter 4) for the proportional-integral controllers with parallel reactions ($\alpha = 2$, $\beta = \frac{1}{2}$). The first set of values in Table 6.7 is for the best proportional-integral controller. The next two sets of values are the false optimum ones, which give smaller yields than the true optimum, but also give smaller positive real parts. They are only slightly more unstable than the best proportional controllers, but also give only slightly better yields than the proportional controllers. Thus, at the expense of decreased yield, the positive real parts of the eigenvalues can be decreased in size.

Since isothermal operation gives all negative real parts for the eigenvalues, it was decided to try moving along the major axes of the ellipses presented in Figures 5.1 and 5.2, where it was expected that movement in either direction away from the optimal would not greatly damage yield, but that movement towards isothermal ($c_2 = 0$) would lead to greater stability, whereas movement away from isothermal would lead to a less stable condition. This proved to be so, as can be seen from the results presented in Table 6.7 for the parallel reaction scheme and Table

TABLE 6.7 SPECIAL STABILITY STUDY FOR PARALLEL REACTIONS ($\alpha = 2$, $\beta = \frac{1}{2}$)

					Largest Real Part of Eigenvalues at Various Times					
c_1	c_2	c_3	c_4	Yield	t =0	.2	.4	.6	.8	1.0
.517	-51.9	-12.6	38.7	.5720	18.0	15.0	12.0	8.8	7.0	8.2
.588	.133	1.21	1.46	.5633	.88	.72	.86	1.09	1.44	1.9
.567	.568	.597	1.31	.5623	.53	.47	.65	.96	n*	n
.569	2.66	-	-	.5591	1.5	.33	n	n	n	n
.738	1.32	-	-	.5457	.28	n	n	n	n	n
.399	3.98	-	-	.5524	2.97	1.18	n	n	n	n
.907	0.0	-	-	.5040	n	n	n	n	n	n
.231	5.31	-	-	.5374	4.32	3.70	.44	n	n	n

* n means that all eigenvalues have negative real parts.

TABLE 6.8 SPECIAL STABILITY STUDY FOR CONSECUTIVE REACTIONS ($\alpha = 2$, $\beta = \frac{1}{2}$)

			Largest Real Part of Eigenvalues at Various Times					
c_1	c_2	Yield	t = 0	0.2	0.4	0.6	0.8	1.0
1.97	-2.67	.4887	n*	n	.20	.28	.31	.33
2.48	-4.00	.4866	n	.18	.38	.41	.38	.34
1.45	-1.34	.4860	n	n	n	.04	.09	.13
3.00	-5.34	.4813	n	.38	.54	.48	.38	.28
.933	0.0	.4756	n	n	n	n	n	n

* n means that all eigenvalues have negative real parts.

6.8 for the consecutive reaction scheme, where two points on either side of the optimal were studied. As is to be expected, the further one gets from the optimal, the poorer the ellipse is at estimating the contours. Thus, the isothermal values obtained when the major axis intersects $c_2 = 0$ are not exactly the best isothermal values, but are fairly close, as can be seen in Figures 5.1 and 5.2. Thus the ellipses are useful tools in studying stability.

As a final illustrative example, it was decided, for the consecutive and parallel schemes ($\gamma = 2$, $\beta = \frac{1}{2}$) with proportional control, to integrate Equation 6.6, putting in a disturbance at some time t and following it in time. There are two state variables, x_1 and x_2, for the proportional case, and if at time, t

$$\begin{aligned} \delta x_1(t) &= a \\ \delta x_2(t) &= b \end{aligned} \tag{6.18}$$

it is desired to know $\delta x_2(1)$. Since Equation 6.6 is linear, if it is integrated twice with the two sets of boundary conditions

$$\begin{aligned} \delta x_1^{(1)}(0) &= 1, \quad \delta x_2^{(1)}(0) = 0 \\ \delta x_1^{(2)}(0) &= 0, \quad \delta x_2^{(2)}(0) = 1 \end{aligned} \tag{6.19}$$

TABLE 6.9 ADDITIONAL STUDY FOR CONSECUTIVE REACTIONS

($\gamma = 2, \beta = \frac{1}{2}$)

t	x_1	x_2	$\delta x_1^{(1)}$	$\delta x_2^{(1)}$	$\delta x_1^{(2)}$	$\delta x_2^{(2)}$
0	1.000	0.000	1.000	0.000	0.000	1.000
.2	.727	.241	.776	.212	.287	.594
.4	.577	.351	.696	.305	.395	.512
.6	.479	.416	.665	.373	.457	.504
.8	.408	.459	.655	.432	.502	.522
1.0	.355	.489	.657	.489	.538	.554

TABLE 6.10 ADDITIONAL STUDY FOR PARALLEL REACTIONS

($\alpha = 2$, $\beta = \frac{1}{2}$)

t	x_1	x_2	$\delta x_1^{(1)}$	$\delta x_2^{(1)}$	$\delta x_1^{(2)}$	$\delta x_2^{(2)}$
0	1.000	0.000	1.000	0.000	0.000	1.000
.2	.816	.134	.758	.166	-.966	1.54
.4	.572	.290	.348	.390	-1.80	1.85
.6	.333	.426	-.012	.547	-1.85	1.69
.8	.165	.513	-.148	.584	-1.30	1.31
1.0	.0743	.559	-.135	.559	-.717	.9871

then

$$\delta x_1(t) = a = K_1 \delta x_1^{(1)}(t) + K_2 \delta x_1^{(2)}(t)$$
$$\delta x_2(t) = b = K_1 \delta x_1^{(1)}(t) + K_2 \delta x_2^{(2)}(t) \qquad (6.20)$$

Thus if at time, t, disturbances a and b are specified, K_1 and K_2 can be solved for and

$$\delta x_2(1) = K_1 \delta x_2^{(1)}(1) + K_2 \delta x_2^{(2)}(1) \qquad (6.21)$$

The results are presented in Table 6.9 for the consecutive reaction scheme and Table 6.10 contains the results for the parallel reaction scheme. It is interesting to note that $x_2^{(1)}(1)$ in both cases is identical to $x_2(1)$. This means that a tiny increase in reagent will behave in almost the same manner as the rest of the batch. For the parallel reactions, $x_2^{(2)}(1) = .99$, which indicates that if a small amount of product is added at the beginning, it will be present at the end essentially untouched. For the consecutive reactions, $x_2^{(2)}(1) = .55$, which means that if a small amount of product is placed in the reactor at the beginning, 45% of it will have reacted to waste by the end of the batch.

6.4 EFFECT OF A DISTURBANCE IMPOSED ON TEMPERATURE

It is useful to study the effect of imposing a noise on the batch reactor, to see how successfully the control system may be expected to react with real disturbances. For the proportional controller cases, the temperature has been determined by calculating $(c_1 + c_2x_2)$, and using this value as the temperature, u(t). If instead,

$$u(t) = c_1 + c_2x_2(t) + h(t) \qquad (6.22)$$

where h(t) is some random function of time, then trials can be made to determine the behavior of the reactor towards disturbances in temperature.

The particular h(t) to be employed here is a random signal which may with equal probability assume, at any instant of time, either of the values $+\epsilon$ or $-\epsilon$, where $|\epsilon|$ is the temperature disturbance. The signal makes independent random traversals from one value to the other, with λ being the average number of traversals during the batch. In setting up h(t), another random variable, r, is called upon, which can with equal probability take on any value between zero and one. At the start of the batch if $r_o < \frac{1}{2}$, $h(0) = -\epsilon$ and if $r_o > \frac{1}{2}$, $h(0) = +\epsilon$. Then the time intervals between traversals are calculated by the standard relations for such Poisson processes (Davenport and Root, 1958)

$$t_i = -(\ln r_i)/\lambda \quad ; \; i = 1,2,\cdots m \qquad (6.23)$$

where

$$\sum_{i=1}^{m} t_i \geq 1 \qquad (6.24)$$

This signal, h(t), has a mean of zero, a variance of ϵ^2, and a covariance function $\epsilon^2 \exp(-2\lambda|\tau|)$.

In performing the computation, first h(t) is determined, then the integration of the batch equations is performed with the temperature values calculated using Equation 6.22. In order to obtain meaningful results, the computation must be repeated many times and the results evaluated statistically. For a particular case being studied, the data required are ϵ, λ, and N, the number of trials to be made. The important results for a case are the sample mean and sample standard deviation of the N values of product yield.

Some illustrative cases for both the consecutive and parallel reaction schemes with $\gamma = 2$, $\beta = \frac{1}{2}$, were studied using both the best isothermal temperature and the best proportional controller values. Some studies for various values of ϵ, and a moderately large λ of 20, so that the signal could be considered nearly white noise, were performed. The results are presented in Tables 6.11 and 6.12 along with ranges on the mean and standard deviation, calculated using Student's t test for the mean

TABLE 6.11 EFFECT OF TEMPERATURE DISTURBANCE ON REACTOR YIELD - CONSECUTIVE REACTIONS ($\alpha = 2$, $\beta = \frac{1}{2}$, $\lambda = 20$, N = 50)

	MEAN		STANDARD DEVIATION		
ϵ	Sample	Range*	Lower* Limit	Sample	Upper* Limit
		(Isothermal)			
0	.4773	-	-	0	-
.05	.4770	± .0002	.0007	.0008	.0009
.10	.4762	± .0004	.0012	.0015	.0018
.25	.4686	± .0010	.0029	.0035	.0044
		(Proportional Control)			
0	.4887	-	-	0	-
.05	.4883	± .0001	.0003	.0003	.0004
.10	.4870	± .0001	.0004	.0005	.0006
.25	.4788	± .0004	.0011	.0013	.0016

* 95% Confidence Interval

TABLE 6.12 EFFECT OF TEMPERATURE DISTURBANCE ON REACTOR YIELD - PARALLEL REACTIONS

($\alpha = 2$, $\beta = \frac{1}{2}$, $\lambda = 20$, N = 50)

	MEAN		STANDARD DEVIATION		
ϵ	Sample	Range*	Lower* Limit	Sample	Upper* Limit
		(Isothermal)			
0	.5347	-	-	0	-
.05	.5346	± .0003	.0010	.0012	.0015
.10	.5339	± .0007	.0021	.0025	.0031
.25	.5296	± .0014	.0040	.0048	.0060
		(Proportional Control)			
0	.5589	-	-	0	-
.05	.5587	± .0002	.0005	.0006	.0007
.10	.5579	± .0004	.0010	.0012	.0015
.25	.5503	± .0008	.0022	.0026	.0032

* 95% Confidence Interval

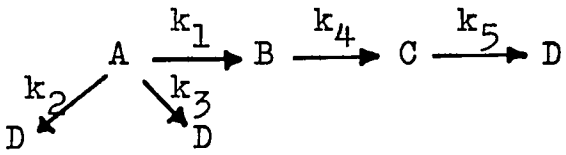
and the chi-square test for the standard deviation, for a 95% confidence interval.

It can be seen that the mean product yield decreases with increasing ϵ, and the standard deviation increases with increasing ϵ. As the temperature disturbance is increased, it is to be expected that it will decrease the average yield obtained from a number of runs, and increase the spread of the individual yield values about the average value. The results in general show that the behavior of the system is not unduly sensitive to such random disturbances as may be expected to occur in practice. And it is especially interesting to note that the proportional control operation is slightly less sensitive to disturbance than the isothermal operation. Thus the nominal instabilities evidenced by the small positive real parts of the eigenvalues in the A matrix do not necessarily indicate that the system is going to behave poorly.

CHAPTER 7 STUDY OF A COMPLEX REACTION SCHEME

7.1 INTRODUCTION

The methods discussed in Chapters 2, 3, 4, 5 and 6 are now applied to a more complex reaction scheme which was originally studied by Rosenbrock and Storey (1966). The reaction scheme is



(7.1)

where A is the feed material, B is the transient intermediate, C is the desired product and D is an unwanted side product. The rate constants are given in the form

$$k_i = (p_i T_f)\exp\left[-(E_i/R)(1/\theta - 1/658)\right] \quad (7.2)$$

where p_i is a frequency factor, E_i is an activation energy, T_f is the total batch time, R is the gas constant and θ is the reactor temperature. The values of the kinetic parameters are shown in Table 7.1.

Since the optimization computer programs are set up to maximize $x_2(1)$, the state variables are defined with x_1 as the concentration of A, x_3 as the concentration of B, and x_2 as the concentration of C. The differential

TABLE 7.1 KINETIC PARAMETERS FOR THE COMPLEX REACTION SCHEME

i	$p_i T_f$	$E_i/R(^oK)^{-1}$
1	.077	8,000
2	.070	7,000
3	.029	7,500
4	.248	5,000
5	.0064	7,500

equations governing the state of the reactor are

$$\begin{aligned} dx_1/dt &= -(k_1 + k_2 + k_3)x_1 = f_1 \\ dx_2/dt &= k_4x_3 - k_5x_2 = f_2 \\ dx_3/dt &= k_1x_1 - k_4x_3 = f_3 \end{aligned} \tag{7.3}$$

with the initial conditions

$$x_1(0) = 1, \quad x_2(0) = 0, \quad x_3(0) = 0 \tag{7.4}$$

The optimal temperature schedule, the best isothermal temperature and the best proportional controller are found along with their associated yields. Then the sharpness of the optimum is studied for the proportional controller case and studies are made on the effects of disturbances on reactor performance. All computations are made with the programs previously described and only the subroutines are changed to incorporate the new reaction scheme.

7.2 METHOD OF SOLUTION

The solution is straightfoward and follows directly from the previous work. As was mentioned in Section 5.3, it is convenient to set up the equations requiring partial derivatives with the derivatives left as unspecified variables (such as DFJDXI(J,I) for $\partial f_j/\partial x_i$). When the subroutine is entered, the partial derivatives are first evaluated (this is where the reactor scheme is identified), then the equations are evaluated. This procedure requires only slightly more computation time and greatly decreases chances for algebraic and programming errors.

In seeking the best proportional controller settings ($\theta = c_1 + c_2 x_2$), the state equations

$$dx_4/dt = c_2(dx_2/dt) = f_4;\ x_4(0) = c_1$$
$$dx_5/dt = 0 = f_5;\ x_5(0) = c_2 \qquad (7.5)$$

are required, where $x_4 = \theta$ and $x_5 = c_2$.

The rest of this section will be devoted to presenting, for completeness, all of the partial derivatives required for the calculations.

$$\partial k_i/\partial\theta = (E_i/R\theta^2)k_i$$

$$\partial k_i/\partial c_1 = \partial k_i/\partial\theta$$

$$\partial k_i/\partial c_2 = (\partial k_i/\partial\theta)x_2$$

$$\partial k_i/\partial x_2 = (\partial k_i/\partial\theta)c_2$$

$$\partial^2 k_i/\partial\theta^2 = (k_i E_i/R\theta^3)(E_i/R\theta - 2)$$

$$\partial^2 k_i/\partial\theta\partial c_1 = \partial^2 k_i/\partial\theta^2$$

$$\partial^2 k_i/\partial\theta\partial c_2 = (\partial^2 k_i/\partial\theta^2)x_2$$

$$\partial^2 k_i/\partial\theta\partial x_2 = (\partial^2 k_i/\partial\theta^2)c_2$$

$$\partial f_1/\partial\theta = -(\partial k_1/\partial\theta + \partial k_2/\partial\theta + \partial k_3/\partial\theta)x_1$$

$$\partial f_1/\partial x_1 = -(k_1 + k_2 + k_3)$$

$$\partial f_1/\partial x_2 = (\partial f_1/\partial\theta)c_2$$

(= 0 for temperature schedule calculation)

$$\partial f_1/\partial x_3 = 0$$

$$\partial f_1/\partial c_1 = \partial f_1/\partial\theta$$

$$\partial f_1/\partial c_2 = (\partial f_1/\partial\theta)x_2$$

$$\partial f_2/\partial\theta = x_3(\partial k_4/\partial\theta) - x_2(\partial k_5/\partial\theta)$$

$$\partial f_2/\partial x_1 = 0$$

$$\partial f_2/\partial x_2 = -k_5 + c_2(\partial f_2/\partial \theta)$$

(= $-k_5$ for temperature schedule calculation)

$$\partial f_2/\partial x_3 = k_4$$

$$\partial f_2/\partial c_1 = \partial f_2/\partial \theta$$

$$\partial f_2/\partial c_2 = (\partial f_2/\partial \theta)x_2$$

$$\partial f_3/\partial \theta = x_1(\partial k_1/\partial \theta) - x_3(\partial k_4/\partial \theta)$$

$$\partial f_3/\partial x_1 = k_1$$

$$\partial f_3/\partial x_2 = (\partial f_3/\partial \theta)c_2$$

(= 0 for temperature schedule calculation)

$$\partial f_3/\partial x_3 = -k_4$$

$$\partial f_3/\partial c_1 = \partial f_3/\partial \theta$$

$$\partial f_3/\partial c_2 = (\partial f_3/\partial \theta)x_2$$

$$\partial f_4/\partial \theta = c_2(\partial f_2/\partial \theta)$$

$$\partial f_4/\partial x_1 = c_2(\partial f_2/\partial x_1)$$

$$\partial f_4/\partial x_2 = c_2(\partial f_2/\partial x_2)$$

$$\partial f_4/\partial x_3 = c_2(\partial f_2/\partial x_3)$$

$$\partial f_4/\partial c_1 = c_2(\partial f_2/\partial c_1)$$

$$\partial f_4/\partial c_2 = f_2$$

$$\partial^2 f_1/\partial\theta^2 = -(\partial^2 k_1/\partial\theta^2 + \partial^2 k_2/\partial\theta^2 + \partial^2 k_3/\partial\theta^2)x_1$$

$$\partial^2 f_1/\partial\theta\partial x_1 = -(\partial k_1/\partial\theta + \partial k_2/\partial\theta + \partial k_3/\partial\theta)$$

$$\partial^2 f_1/\partial x_1\partial x_1 = 0$$

$$\partial^2 f_1/\partial x_1\partial x_2 = (\partial^2 f_1/\partial\theta\partial x_1)c_2$$

$$\partial^2 f_1/\partial x_1\partial x_3 = 0$$

$$\partial^2 f_1/\partial x_1\partial c_1 = \partial^2 f_1/\partial\theta\partial x_1$$

$$\partial^2 f_1/\partial x_1\partial c_2 = (\partial^2 f_1/\partial\theta\partial x_1)x_2$$

$$\partial^2 f_1/\partial x_2\partial x_2 = (\partial^2 f_1/\partial\theta^2)c_2^2$$

$$\partial^2 f_1/\partial x_2\partial x_3 = 0$$

$$\partial^2 f_1/\partial x_2\partial c_1 = (\partial^2 f_1/\partial\theta^2)c_2$$

$$\partial^2 f_1/\partial x_2\partial c_2 = (\partial^2 f_1/\partial\theta^2)x_2c_2 + (\partial f_1/\partial\theta)$$

$$\partial^2 f_1/\partial x_3\partial x_3 = 0$$

$$\partial^2 f_1/\partial x_3\partial c_1 = 0$$

$$\partial^2 f_1/\partial x_3\partial c_2 = 0$$

$$\partial^2 f_1/\partial c_1\partial c_1 = \partial^2 f_1/\partial\theta^2$$

$$\partial^2 f_1/\partial c_1\partial c_2 = (\partial^2 f_1/\partial\theta^2)x_2$$

$$\partial^2 f_1/\partial c_2\partial c_2 = (\partial^2 f_1/\partial\theta^2)x_2^2$$

$$\partial^2 f_2/\partial\theta^2 = x_3(\partial^2 k_4/\partial\theta^2) - x_2(\partial^2 k_5/\partial\theta^2)$$

$$\partial^2 f_2/\partial\theta\partial x_2 = c_2(\partial^2 f_2/\partial\theta^2) - (\partial k_5/\partial\theta)$$

$$\partial^2 f_2/\partial\theta\partial x_3 = \partial k_4/\partial\theta$$

$$\partial^2 f_2/\partial x_1\partial x_1 = 0$$

$$\partial^2 f_2/\partial x_1\partial x_2 = 0$$

$$\partial^2 f_2/\partial x_1\partial x_3 = 0$$

$$\partial^2 f_2/\partial x_1\partial c_1 = 0$$

$$\partial^2 f_2/\partial x_1\partial c_2 = 0$$

$$\partial^2 f_2/\partial x_2\partial x_2 = -(\partial k_5/\partial\theta)c_2 + (\partial^2 f_2/\partial\theta\partial x_2)c_2$$

$$\partial^2 f_2/\partial x_2\partial x_3 = (\partial^2 f_2/\partial\theta\partial x_3)c_2$$

$$\partial^2 f_2/\partial x_2\partial c_1 = (\partial^2 f_2/\partial\theta\partial x_2)$$

$$\partial^2 f_2/\partial x_2\partial c_2 = \partial f_2/\partial\theta + (\partial^2 f_2/\partial\theta\partial x_2)x_2$$

$$\partial^2 f_2/\partial x_3\partial x_3 = 0$$

$$\partial^2 f_2/\partial x_3\partial c_1 = \partial^2 f_2/\partial x_3\partial\theta$$

$$\partial^2 f_2/\partial x_3\partial c_2 = (\partial^2 f_2/\partial x_3\partial\theta)x_2$$

$$\partial^2 f_2/\partial c_1\partial c_1 = \partial^2 f_2/\partial\theta^2$$

$$\partial^2 f_2/\partial c_1\partial c_2 = (\partial^2 f_2/\partial\theta^2)x_2$$

$$\partial^2 f_2/\partial c_2\partial c_2 = (\partial^2 f_2/\partial\theta^2)x_2^2$$

$$\partial^2 f_3/\partial\theta^2 = x_1(\partial^2 k_1/\partial\theta^2) - x_3(\partial^2 k_4/\partial\theta^2)$$

$$\partial^2 f_3/\partial\theta\partial x_1 = \partial k_1/\partial\theta$$

$$\partial^2 f_3/\partial\theta\partial x_2 = c_2(\partial^2 f_3/\partial\theta^2)$$

$$\partial^2 f_3/\partial\theta\partial x_3 = (- \partial k_4/\partial\theta)$$

$$\partial^2 f_3/\partial x_1\partial x_1 = 0$$

$$\partial^2 f_3/\partial x_1\partial x_2 = c_2(\partial^2 f_3/\partial\theta\partial x_1)$$

$$\partial^2 f_3/\partial x_1\partial x_3 = 0$$

$$\partial^2 f_3/\partial x_1\partial c_1 = \partial^2 f_3/\partial\theta\partial x_1$$

$$\partial^2 f_3/\partial x_1\partial c_2 = x_2(\partial^2 f_3/\partial\theta\partial x_1)$$

$$\partial^2 f_3/\partial x_2\partial x_2 = (\partial^2 f_3/\partial\theta^2)c_2^2$$

$$\partial^2 f_3/\partial x_2\partial x_3 = (\partial^2 f_3/\partial\theta\partial x_3)c_2$$

$$\partial^2 f_3/\partial x_2\partial c_1 = (\partial^2 f_3/\partial\theta\partial x_2)$$

$$\partial^2 f_3/\partial x_2\partial c_2 = \partial f_3/\partial\theta + (\partial^2 f_3/\partial\theta^2)x_2c_2$$

$$\partial^2 f_3/\partial x_3\partial x_3 = 0$$

$$\partial^2 f_3/\partial x_3\partial c_1 = \partial^2 f_3/\partial\theta\partial x_3$$

$$\partial^2 f_3/\partial x_3\partial c_2 = (\partial^2 f_3/\partial\theta\partial x_3)x_2$$

$$\partial^2 f_3/\partial c_1\partial c_1 = \partial^2 f_3/\partial\theta^2$$

$$\partial^2 f_3/\partial c_1\partial c_2 = (\partial^2 f_3/\partial\theta^2)x_2$$

$$\partial^2 f_3/\partial c_2\partial c_2 = (\partial^2 f_3/\partial\theta^2)x_2^2$$

7.3 RESULTS FOR COMPLEX REACTION SCHEME STUDY

The best isothermal temperature is 983°K with an associated product yield of 0.4231. The optimal temperature schedule and associated product yield history are shown in Table 7.2. It can be seen that the maximum allowable temperature has been set at 1120°K and the product yield is 0.4479. The least squares constants for the proportional controller ($\theta = c_1 + c_2 x_2$) are $c_1 = 1209$, $c_2 = -713$. But when operating with proportional control, $\theta(0) = c_1$ and therefore c_1 cannot be greater than the maximum allowable temperature. Starting the search for the best proportional controller with $c_1 = 1120$, $c_2 = -713$ and an associated product yield of 0.4309 gives the optimal values of $c_1 = 1120$, $c_2 = -513$, with an associated product yield of 0.4451. The temperature and associated product yield histories are shown in Table 7.2.

In studying the sharpness of the optimum with respect to the controller settings, the quadratic approximations of the contours of constant yield were again found to be elongated ellipses, with the major axes being 8.9 times as long as the minor axis, and the slope (c_2/c_1) of the major axis being -3.71. But the center of the ellipse turned out to be at $c_1 = 1240$, $c_2 = -701$ with a predicted yield of 0.4602. This tells us again what was

TABLE 7.2 TEMPERATURE AND PRODUCT YIELD HISTORIES FOR COMPLEX REACTION SCHEME

	Best Temperature Schedule		Best Proportional Controller	
t	Temp. (°K)	Conc.	Temp. (°K)	Conc.
0	1120	0.0	1120	0.0
.1	1120	.1525	1053	.1295
.2	1085	.3054	996	.2419
.3	994	.3720	961	.3102
.4	941	.4021	938	.3544
.5	909	.4189	923	.3845
.6	888	.4296	911	.4058
.7	873	.4368	904	.4210
.8	864	.4419	898	.4320
.9	857	.4455	895	.4397
1.0	852	.4479	891	.4451

Best Isothermal Temperature = 983

Best Isothermal Yield = .4231

already known from the optimization work - that to obtain an increase in final yield, the maximum allowable temperature must be raised. The temperature wants to be at infinity at the beginning of the batch. This is not practical and a realistic upper temperature bound has been set.

The results of a linearized stability study of the type described in Section 6.2 are presented in Table 7.3. Although the A matrix contains positive real parts, they are small and it is expected that the system will be well behaved.

The results of a study made to determine the effect of a random temperature disturbance on reactor performance, using the techniques described in Section 6.4, are presented in Table 7.4. It can be seen that even with a 20 K^{o} error, there is very little damage done to the yield for either the isothermal or proportional control case.

TABLE 7.3 STABILITY STUDY FOR COMPLEX REACTION SCHEME WITH PROPORTIONAL CONTROL

t	Largest Real Part of Eigenvalues
0	all negative
.2	.717
.4	.398
.6	.302
.8	.263
1.0	.274

TABLE 7.4 EFFECT OF TEMPERATURE DISTURBANCE ON REACTOR YIELD FOR COMPLEX REACTION SCHEME (λ = 20, N = 50)

	MEAN		STANDARD DEVIATION		
ϵ	Sample	Range*	Lower* Limit	Sample	Upper* Limit
		(Isothermal)			
0	.4231	-	-	0	-
5	.4231	± .0001	.0003	.0004	.0005
10	.4232	± .0002	.0007	.0008	.0010
20	.4231	± .0005	.0015	.0018	.0022
		(Proportional Control)			
0	.4451	-	-	0	-
5	.4450	± .0001	.0004	.0005	.0006
10	.4447	± .0002	.0007	.0008	.0010
20	.4448	± .0004	.0012	.0014	.0017

* 95% Confidence Interval

CHAPTER 8 FOLLOWING A TEMPERATURE SCHEDULE

8.1 INTRODUCTION

Until now all of the work presented has assumed that the temperature of a batch is exactly at the value which is specified by the optimal schedule or the sub-optimal controller. But the contours of reactor performance in the space of the controller settings show that the yield may be uncomfortably sensitive to inaccuracies in these settings (in certain directions). Besides, the studies up to this point have assumed that an arbitrarily large heat duty was available to implement the desired control in the face of heats of reaction and the thermal lag of the reacting mass. Accordingly, in this chapter, a study is presented to see how closely the actual yield approaches the schedule yield and how stable the system is when it is enlarged to include heat exchange equipment.

For this study, the reversible reaction scheme

$$A \underset{k_2}{\overset{k_1}{\rightleftharpoons}} B \qquad (8.1)$$

is employed, where A is the feed and B is the desired product. This scheme is chosen because it can be described mathematically with one differential equation and

its optimal schedule can be solved for analytically. This allows a great deal of the work to be carried out analytically, with digital computation required only for a numerical example.

8.2 THE REVERSIBLE REACTION SCHEME

The differential equation describing the state of the reversible reaction scheme is

$$\begin{aligned} dy/dt &= k_1(1 - y) - k_2 y = f_1 \\ y(0) &= 0, \quad 0 \leqslant t \leqslant 1 \end{aligned} \tag{8.2}$$

where y is the concentration of B, $y(0) = 0$ because the material charged to the reactor is pure A and

$$k_i = (p_i T_f)\exp(- E_i/R\theta) \tag{8.3}$$

where p_i is a frequency factor, E_i is an activation energy, T_f is the total batch time, R is the gas constant and θ is the reactor temperature. For convenience,

$$\begin{aligned} \gamma &= E_2/E_1 \\ \beta' &= p_2/p_1 \end{aligned} \tag{8.4}$$

are defined.

In order to obtain the maximum final product yield, the rate of reaction must be maximized at every point in time. Taking the derivative of dy/dt with respect to θ gives

$$df_1/d\theta = (E_1k_1(1 - y) - E_2k_2y)/(R\theta^2)$$

$$= p_1E_1T_f(\exp(-E_1/R\theta)(1 - y) - \exp(-E_2/R\theta)y)/R\theta^2 \qquad (8.5)$$

If $\theta > 0$ and remains finite, then the stationary point is given by

$$(1 - y)\exp(-E_1/R\theta) = \alpha\beta' \exp(-E_2/R\theta)$$

which gives a temperature of

$$\theta_s = (\alpha - 1)(E_1/R)/ \ln(\alpha\beta' y/(1 - y)) \qquad (8.6)$$

The second derivative has been tested to determine if the temperature calculated from Equation 8.6 gives a maximum reaction rate. If $\alpha \leq 1$, the stationary point gives a minimum, and the temperature boundaries must be explored. If θ_{max} is the maximum allowable temperature, then the optimal policy for $\alpha \leq 1$ is to run at the maximum allowable temperature. When $\alpha > 1$, the stationary point gives a maximum and the optimal policy is to follow θ_s given by Equation 8.6, unless $\theta_s > \theta_{max}$, in which case θ_{max} is used instead.

The Arrhenius expressions for rate constants are

based on the fact that

$$-\Delta H_r = (E_2 - E_1) \qquad (8.7)$$

where $-\Delta H_r$ is the heat of reaction. When $\gamma<1$, the reaction is endothermic and the best policy is to operate at the maximum allowable temperature. Since the reaction absorbs heat and it is best to operate at a very high temperature, heat is simply added to the reactor as fast as is possible, within the limits of the equipment and reaction material. Thus there is no interesting optimization question to be asked. For the cases where $\gamma>1$, the optimal policy is a temperature profile which starts at the maximum allowable temperature and decreases as the reaction progresses. At the same time, the reaction is exothermic and tends to heat the reactor. Here is an interesting optimization problem because the reactor wants to be cooling down for maximum yield, while the heat of reaction would tend to raise the temperature. Thus controlled cooling is required to optimize the reactor behavior. It is this latter case which is studied in detail.

The course of temperature in the reactor is governed by the differential equation

$$A_o C_r (d\theta/T_f dt) = (-\Delta H_r) A_o (dy/dt) - Q \qquad (8.8)$$

where A_o is the total number of moles in the reactor, C_r is the heat capacity of the material in the reactor and Q is the heat removed from the reactor by the heat exchange equipment. It is assumed that C_r is constant and that $(-\Delta H_r)$ is replaced by $(E_2 - E_1)$.

8.3 THE SENSIBLE HEAT EXCHANGER

A coolant with an initial temperature of θ_i is passed through a heat exchanger with an overall heat transfer coefficient, U (which is assumed to be constant) and a total heat transfer area, S. The coolant leaves the exchanger at a temperature θ_e and the average heat transfer temperature, θ_A, is assumed to be $\frac{1}{2}(\theta_i + \theta_e)$. The coolant is assumed to have a constant density ρ_H and a constant heat capacity, C_H. The total volume of the exchanger is V_H. If w is the flow rate of coolant, then the heat removed from the reactor is

$$Q = US(\theta - \theta_A) \tag{8.9}$$

and the dynamics of the heat exchanger are represented by the differential equation

$$\rho_H C_H V_H (d\theta_A/dt) = US(\theta - \theta_A) - 2wC_H(\theta_A - \theta_i) \tag{8.10}$$

The tracking controller, which measures the reactor temperature, compares it to the desired schedule temperature and sets the coolant flow rate, may be represented by

$$w = w_o + K(\theta - \theta_s) \tag{8.11}$$

where w_o is the set point and K is the proportional sensitivity. It is convenient to define the variable

$$g_3 = (E_2 - E_1)/C_r \tag{8.12}$$

which has the units of temperature. Now the dimensionless temperatures

$$\begin{aligned} x &= \theta/g_3 \\ x_A &= \theta_A/g_3 \\ x_i &= \theta_i/g_3 \\ x_s &= \theta_s/g_3 \end{aligned} \tag{8.13}$$

are defined. Also defined are the dimensionless quantities

$$\begin{aligned} g_1 &= UST_f/A_oC_r \\ g_2 &= UST_f/\rho_H C_H V_H \\ w^* &= 2wC_H/US \\ K^* &= 2K(E_2 - E_1)C_H/USC_r \end{aligned} \tag{8.14}$$

and in terms of the variables just defined, the state

equations for the total system are

$$\begin{aligned} dy/dt &= k_1(1 - y) - k_2 y = f_1 \\ dx/dt &= dy/dt - g_1(x - x_A) = f_2 \\ dx_A/dt &= g_2\left((x - x_A) - w^*(x_A - x_i) \right) \\ &= f_3 \end{aligned} \tag{8.15}$$

$$w^* = w_o^* + K^*(x - x_s)$$

with the initial conditions

$$y(0) = 0, \quad x(0) = x_o, \quad x_A(0) = x_i \tag{8.16}$$

As $g_2 \longrightarrow \infty$, the dynamics of the heat exchanger can be neglected and

$$g_1(x - x_A) = g_1 w^*(x_A - x_i) \tag{8.17}$$

But this can be solved for x_A,

$$x_A = (x + x_i w^*)/(1 + w^*) \tag{8.18}$$

so that

$$g_1(x - x_A) = g_1 w^*(x - x_i)/(1 + w^*) \tag{8.19}$$

and the state equations for the system when heat exchanger dynamics may be neglected are

$$\begin{aligned} dy/dt &= k_1(1 - y) - k_2 y = f_1 \\ dx/dt &= dy/dt - g_1(x - x_i)w^*/(1 + w^*) \\ &= f_2 \\ w^* &= w_o^* + K^*(x - x_s) \end{aligned} \tag{8.20}$$

with the initial conditions

$$y(0) = 0, \quad x(0) = x_o \tag{8.21}$$

The adiabatic case is obtained as $g_1 \rightarrow 0$.

A stability analysis, of the kind described in Chapter 6, is performed on Equations 8.15. The A matrix, as described with Equation 6.5, is set up as follows

$$\begin{aligned} a_1 &= -(k_1 + k_2) \\ a_2 &= E_1 C_r(k_1(1 - y) - k_2 y)/R(E_2 - E_1)x^2 \\ a_3 &= 0 \\ a_4 &= a_1 \\ a_5 &= a_2 - g_1 \\ a_6 &= g_1 \end{aligned} \tag{8.22}$$

$$a_7 = 0$$

$$a_8 = g_2(1 - K^*(x_A - x_i))$$

$$a_9 = -g_2(1 + w^*)$$

Then the variables b_1, b_2 and b_3 are calculated with Equations 6.8, and the Routh-Hurwitz criterion, that $b_1 > 0$, $b_2 > 0$ and $b_1 b_2 > b_3$ are the three conditions which must be satisfied if all the eigenvalues of the A matrix are to have negative real parts, is applied. Condition 1 is that

$$g_2(1 + w^*) + k_1 + k_2 + g_1 > a_2 \qquad (8.23)$$

Condition 2 is that

$$K^*(x_A - x_i) + w^* > 0 \qquad (8.24)$$

Condition 3 is that

$$\begin{aligned} &\Big[g_1 + g_2(1 + w^*)\Big]\cdot\Big[(k_1 + k_2)^2 + (k_1 + k_2)(g_1 + g_2(1 + w^*)) \\ &\quad + g_1 g_2(K^*(x_A - x_i) + w^*)\Big] > \\ &\quad a_2\Big[g_1(g_2 K^*(x_A - x_i) + g_2 w^* + k_1 + k_2) \\ &\quad g_2(1 + w^*)(2(k_1 + k_2) + g_2(1 + w^*) + g_1 - a_2)\Big] \end{aligned} \qquad (8.25)$$

Condition 2 is always satisifed and conditions 1 and 3 are satisfied when $a_2 \leq 0$ and for small positive values of a_2. But $a_2 = \partial f_1/\partial x$ is equivalent to Equation 8.5, which is set equal to zero to obtain the optimal temperature schedule. Thus when the optimal temperature schedule is followed, $a_2 = 0$, and all conditions for negative real parts are satisfied. The variable a_2 is simply the derivative of the rate of reaction with respect to temperature. Since the temperature at which $a_2 = 0$ is the optimal temperature, then $a_2 < 0$ for temperatures greater than optimal and $a_2 > 0$ for temperatures less than optimal. Thus it is when the reactor temperature is below the optimal value that there is a possibility that a positive real part may appear in one of the eigenvalues.

At temperatures below optimal, a small positive temperature disturbance away from the basepoint would tend to increase the rate of reaction which would further increase the temperature disturbance. But this would only speed the temperature toward the optimal, and as the optimal was approached, the instability would disappear.

The stability study for the case where heat exchange dynamics are neglected can be made by setting up the A matrix for Equations 8.20 or, more easily, by taking the conditions just set down, studying them as $g_2 \to \infty$ and eliminating the extra condition which arises. The two conditions for negative real parts are

$$K^*(x_A - x_i) + w^* > 0 \tag{8.26}$$

and

$$(k_1 + k_2) + (K^*(x_A - x_i) + w^*)g_1/(1 + w^*) > a_2 \tag{8.27}$$

For the adiabatic case, $g_1 \to 0$, and the single condition is

$$(k_1 + k_2) > a_2 \tag{8.28}$$

Thus even the simple adiabatic case shows a positive real part when the reactor temperature is sufficiently below the optimal. But the reactor quickly heats up, and the positive real part disappears.

A numerical example is presented in Section 8.5 to support the work presented here. A block diagram of the scheme is shown in Figure 8.1.

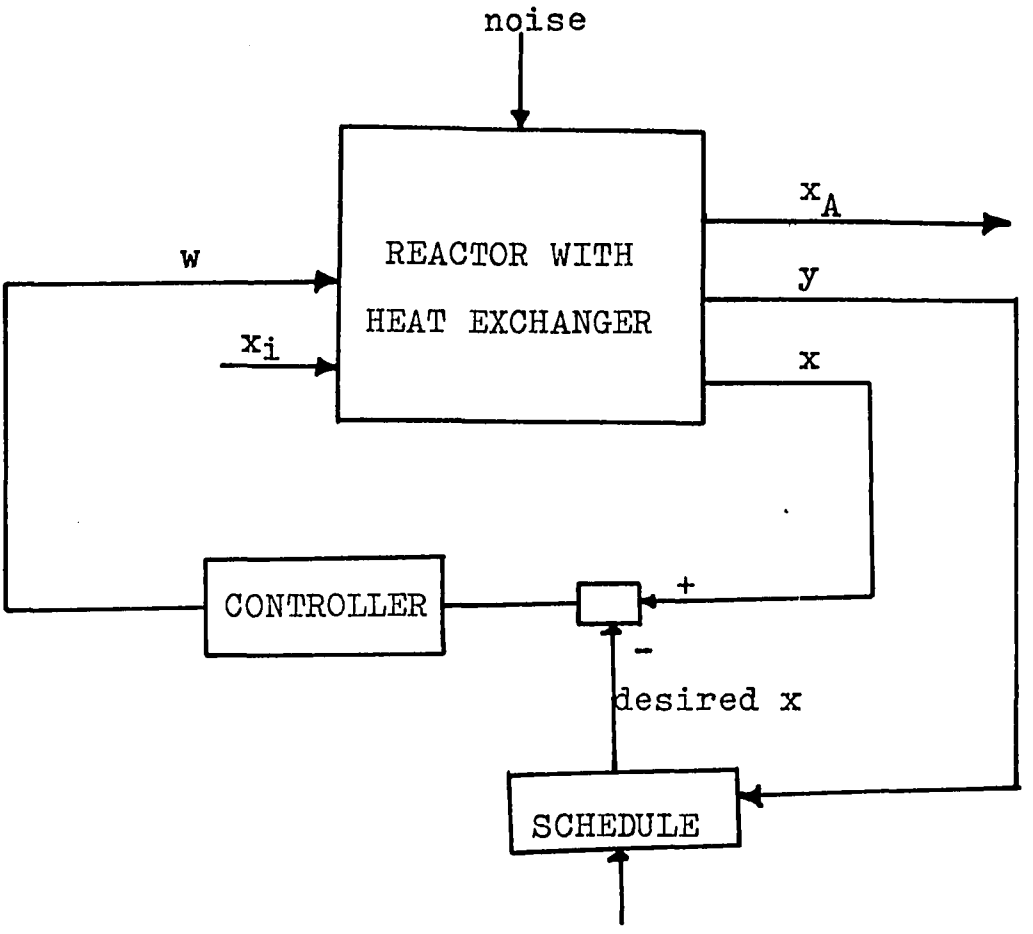


FIGURE 8.1 TRACKING CONTROLLER

8.4 THE SIMPLE BOILER

In order to see if the type of heat exchanger model would have any effect on the results, it was decided to try a second model, this time a boiler. The boiler model studied has a fixed volume, V, and an equilibrium mixture of vapor and liquid at temperature θ_A and pressure P. It is assumed that flow rate of liquid into the boiler equals the flow rate of vapor from the boiler so that the material inventory in the boiler remains constant, although the fraction of vapor as well as pressure and temperature may change. Assuming that the ideal gas law applies, that the heat of vaporization, λ, remains constant, and that the temperature of the liquid input is close to the boiler temperature, then if f is defined as the fraction of vapor, m as the total mass in the boiler, w as the flow rate of liquid input, and Q as the heat input to the boiler, the boiler may be represented by the differential equation

$$\lambda m(df/dt) = Q - \lambda w \qquad (8.29)$$

The relationship between temperature and vapor fraction is obtained from the equilibrium relation and the volume restraint. If it is assumed that the liquid density is much greater than the vapor density, then the volume

restraint is simply

$$(1/f)(df/dt) = (1/P)(dP/dt) - (1/\theta_A)(d\theta_A/dt) \tag{8.30}$$

The equilibrium restraint is given by the Clausius-Clapeyron relation (assuming the ideal gas law and that liquid density is much greater than vapor density)

$$dP/d\theta_A = P\lambda/R\theta_A^2 \tag{8.31}$$

Equations 8.30 and 8.31 are combined to give

$$d\theta_A/dt = (\theta_A/f)(df/dt)/(\lambda/R\theta_A - 1) \tag{8.32}$$

which can be integrated to give

$$f\theta_A = c\cdot\exp(-\lambda/R\theta_A) \tag{8.33}$$

where c is a constant of integration. In carrying out numerical calculations, it was found to be more convenient to use Equation 8.32 than to solve for θ_A in a trial and error calculation, using Equation 8.33, but it must be remembered that this differential equation appears only for convenience, is not one of the state equations, and

does not belongin the stability analysis.

The tracking controller is again assumed to be of the form shown in Equation 8.11 and Q is governed by Equation 8.9. The following factors are defined

$$\begin{aligned} g_1 &= UST_f/A_oC_r \\ g_2 &= US(E_2 - E_1)T_f/C_r\lambda m \\ g_3 &= (E_2 - E_1)/C_r \\ g_4 &= \lambda/Rg_3 \\ w^* &= w\lambda C_r/US(E_2 - E_1) \\ K^* &= K\lambda/US \end{aligned} \tag{8.34}$$

and the dimensionless temperatures are defined in Equations 8.13. The state equations for the system are

$$\begin{aligned} dy/dt &= k_1(1 - y) - k_2y = f_1 \\ dx/dt &= dy/dt - g_1(x - x_A) = f_2 \\ df/dt &= g_2((x - x_A) - w^*) = f_3 \\ w^* &= w_o^* + K^*(x - x_s) \end{aligned} \tag{8.35}$$

and

$$dx_A/dt = g_5(df/dt) \tag{8.36}$$

where

$$g_5 = (x_A/f)/(g_4/x_A - 1) \tag{8.37}$$

The initial conditions are

$$y(0) = 0, \quad x(0) = x_o, \quad f(0) = f_o, \quad x_A(0) = x_i \tag{8.38}$$

As $g_2 \to \infty$, the dynamics of the heat exchanger can be neglected and

$$w^* = x - x_A \tag{8.39}$$

The state equations for the system when heat exchange dynamics may be neglected are

$$\begin{aligned} dy/dt &= k_1(1 - y) - k_2 y = f_1 \\ dx/dt &= dy/dt - g_1 w^* = f_2 \end{aligned} \tag{8.40}$$

with the initial conditions

$$y(0) = 0, \quad x(0) = x_o \tag{8.41}$$

A stability analysis of the type just performed on the sensible heat exchanger is performed on Equations 8.35. The A matrix components are

$$\begin{aligned} a_1 &= -(k_1 + k_2) \\ a_2 &= E_1 C_r (k_1(1 - y) - \alpha k_2 y)/R(E_2 - E_1)x^2 \\ a_3 &= 0 \\ a_4 &= a_1 \\ a_5 &= a_2 - g_1 \\ a_6 &= g_1 g_5 \\ a_7 &= 0 \\ a_8 &= g_2(1 - K^*) \\ a_9 &= -g_2 g_5 \end{aligned} \tag{8.42}$$

and the three conditions for the matrix, A, to have eigenvalues with negative real parts, are as follows. Condition 1 is that

$$k_1 + k_2 + g_1 + g_2 g_5 > a_2 \tag{8.43}$$

Condition 2 is that

$$K^* > 0 \tag{8.44}$$

Condition 3 is that

$$(g_1 + g_2g_5)\left[(k_1 + k_2)(g_1 + g_2g_5) + g_1g_2g_5K^* + (k_1 + k_2)^2\right] > a_2\left[g_2g_5(g_1K^* - a_2 + 2(k_1 + k_2) + g_1 + g_2g_5) + (k_1 + k_2)g_1\right] \tag{8.45}$$

For the case where the dynamics of the heat exchanger may be neglected (Equation 8.40), the conditions are that

$$k_1 + k_2 + K^*g_1 > a_2 \tag{8.46}$$

and

$$K^* > 0 \tag{8.47}$$

Thus for the boiler model, it can be seen from a similar analysis to that used for the sensible heat exchanger stability, that only when the reactor temperature is sufficiently below the optimal value will a positive real part appear, but it will disappear as the reactor heats up and approaches the optimal.

8.5 SOME NUMERICAL EXAMPLES

A reversible reaction scheme, presented by Fan (1966), is used for a numerical example. The kinetic data are

$$\begin{aligned} p_1 T_f &= 2.51 \cdot 10^3 \\ p_2 T_f &= 1.99 \cdot 10^5 \\ E_1/R &= 5{,}000^\circ K \\ E_2/R &= 10{,}000^\circ K \end{aligned} \tag{8.48}$$

The computer programs used are very similar to those described in Chapter 6, with the state equations contained in one subroutine and the stability analysis in another. A test is built in whereby the equations for the situations where heat exchange dynamics are neglected are used when g_2 is greater than some specified value. There are two variations of each equation subroutine - one employs a pre-calculated temperature-time schedule for the tracking controller's desired temperature values; the other calculates the desired temperature at each point by using the product concentration value at that point in time with Equation 8.6, which is equivalent to following the best temperature-product concentration schedule, instead of

the best temperature-time schedule.

A summary of the results is presented in Tables 8.1 and 8.2 for the sensible heat exchanger and the boiler. Table 8.3 presents temperature and product concentration histories for the optimal and adiabatic situations. Table 8.4 presents temperature and product concentration histories for typical situations using the sensible heat exchanger and the boiler. The reactor temperature is not bounded, but the schedule temperature is kept below 900°K.

It can be seen that running in an optimal manner (yield of 0.908) is a vast improvement over adiabatic operation (yield of 0.605). As can be seen from Tables 8.1 and 8.2, whether the sensible heat exchanger or boiler is used, whether heat exchange dynamics are included or neglected, whether temperature-time schedules or temperature-product concentration schedules are followed, the yield is 0.908. It seems that for any reasonable values, the optimal yield is essentially obtained.

As is to be expected, the stability calculations show all negative real parts for the eigenvalues of the A matrix, except at $t = 0$ for the sensible exchanger and adiabatic cases. But these positive real parts are gone by the time $t = 0.05$ (which is the next time that the stability is studied).

Thus the optimum tracking controllers are stable, they attain essentially the optimum value of product

TABLE 8.1 RESULTS FOR SENSIBLE HEAT EXCHANGER STUDY

Case No.	1	2	3	4	5
g_1	10	10	10	10	10
g_2	10	10	1	∞	∞
g_3 (^{o}K)	400	400	400	400	400
w_o*	.2	.1	.2	.2	.2
K*	100	1000	100	100	100
θ (0) (^{o}K)	800	800	800	800	840
θ_A(0) (^{o}K)	500	500	500	500	500
θ_{max} (^{o}K)	900	900	900	900	900
Yield (Time Schedule)	.9075	.9075	.9075	.9077	.9084
Yield (Conc. Schedule)	.9076	.9076	.9076	.9077	.9084

TABLE 8.2 RESULTS FOR BOILER STUDY

Case No.	1	2	3	4
g_1	10	10	10	10
g_2	1.0	1.0	0.1	∞
g_3 (^oK)	400	400	400	400
g_4	20	20	20	20
w_o*	0.0	0.0	0.0	0.0
K*	1.0	10.0	1.0	10.0
θ (0) (^oK)	800	800	800	800
θ_A(0) (^oK)	500	500	500	500
θ_{max} (^oK)	900	900	900	900
f(0)	.2	.2	.2	.2
Yield (Time Schedule)	.9076	.9076	.9075	.9080
Yield (Conc. Schedule)	.9076	.9076	.9076	.9079

TABLE 8.3 RESULTS FOR OPTIMAL AND ADIABATIC PATHS

	Optimal		Adiabatic	
t	θ (^{o}K)	Prod. Conc.	θ (^{o}K)	Prod. Conc.
0	900	0.0000	800	0.0000
.1	900	0.5439	1017	0.5429
.2	839	0.7079	1042	0.6043
.3	791	0.7770	1042	0.6049
.4	761	0.8174	1042	0.6049
.5	739	0.8444	1042	0.6049
.6	723	0.8640	1042	0.6049
.7	709	0.8790	1042	0.6049
.8	698	0.8908	1042	0.6049
.9	688	0.9004	1042	0.6049
1.0	679	0.9084	1042	0.6049

TABLE 8.4 ILLUSTRATIVE TEMPERATURE AND PRODUCT CONCENTRATION PATHS

	Sensible Exchanger (Case 1 with Time Schedule)		Boiler (Case 1 with Yield Schedule)	
t	θ (°K)	Prod. Conc.	θ (°K)	Prod. Conc.
0	800	0.0000	800	0.0000
.1	894	0.5137	900	0.5154
.2	838	0.6973	841	0.6982
.3	791	0.7713	794	0.7719
.4	761	0.8137	763	0.8141
.5	739	0.8419	741	0.8421
.6	723	0.8621	724	0.8623
.7	709	0.8775	711	0.8776
.8	697	0.8896	698	0.8897
.9	687	0.8994	687	0.8995
1.0	679	0.9075	678	0.9076

yield and are insensitive to heat exchanger and tracking controller parameters.

These tracking controllers may be used in cascade with the suboptimal controllers discussed in the previous chapters, the suboptimal controllers sensing product concentration and setting desired temperature which the tracking controllers follow. It is felt that such systems would behave well, although no detailed analysis of such cascades has been made.

NOMENCLATURE

a_i	ith initial condition in Equation 3.1
a_i	ith component of the A matrix, defined by Equation 6.5
A	matrix defined by Equation 6.5
A_i	frequency factor
A_o	total number of moles in the reactor
b_i	variation of controller setting, defined in Equation 5.2
c_i	ith controller constant
C_r	heat capacity of the material in the reactor
e	deviation of the measured variable from its set point
E_i	activation energy
f	fraction of vapor in boiler
f_i	ith component of the function vector which contains the reaction mechanism and kinetic parameters
g_i	ith component of a vector of factors defined in various places to simplify the presentation
$\underline{G}_i$	gradient vector, defined in Equation 3.22
h(t)	random function of time, defined in Equation 6.22
H	positive constant
k_i	Arrhenius rate constant
K, K_c	proportional sensitivity

K^*	dimensionless proportional sensitivity
m	manipulated variable in three-mode controller
M	set point of three-mode controller
M_i	concentration of charged raw material
M,P,W	chemical symbols, as well as concentrations, of raw material, product and waste, respectively
p_i	frequency factor
P	pressure
$\underline{P}_i$	vector defined by Equation 3.22
Q	heat removed from the reactor
r	random number
R	gas constant
S	total heat transfer area
t	reaction time
T_d	derivative time
T_f	total batch time
T_i	integral time
U	overall heat transfer coefficient
u	dimensionless temperature, defined by Equation 2.26
u_o	constant defined in Equation 2.6
w	coolant flow rate
w^*	dimensionless flowrate
x	dimensionless temperature, defined in Chapter 8
x_2^d	constant defined in Equation 2.6
x_i	ith component of the state vector

y	product composition, defined in Equation 8.2
y_{iq}	variable defined in Equation 5.3
z_i	ith component of the adjoint vector, defined by Equation 3.2
z_{iqr}	variable defined by Equation 5.3
α	dimensionless parameter defined by Equation 2.27
β	dimensionless parameter defined by Equation 2.28
β'	dimensionless quantity defined in Equation 8.4
γ	upper dimensionless temperature bound, defined by Equation 2.31
ΔH_r	heat of reaction, Equation 8.7
λ	average number of traversals, Equation 6.23
λ	heat of vaporization, Equation 8.29
λ_i	eigenvalue of matrix A
ρ	density
θ	absolute temperature
θ_m	maximum allowable maximum temperature

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AUTOBIOGRAPHICAL STATEMENT

Martin C. Millman was born on July 20, 1940 in New York City. He attended the New York City Public School System, graduating from William H. Taft High School(Bronx) in January, 1958, at which time he received the English, Chemistry and Music Awards. He then attended the City College of New York, where he was elected to Tau Beta Pi and Omega Chi Epsilon, and received his Bachelor of Chemical Engineering degree in June, 1962 (Cum Laude), at which time he was given the Steven Heller Unit Operations Award. Afterwards he attended Yale University and was awarded the degree of Master of Engineering in June, 1966. He started in the Ph.D. program at the City University of New York in February, 1964, successfully defending his Ph.D. dissertation on May 23, 1968. He was a part-time lecturer at City College in thermodynamics and transport phenomena.

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