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OPTIMUM CONSTRAINED CONTROL OF AN  
UNSTEADY-STATE TUBULAR REACTOR

by

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## I. INTRODUCTION

Intensive international competition and concurrent higher quality standards have forced the chemical industry to seek more efficient methods of operation. This search for efficient operation is manifested by growing interest in the mathematical study of optimal control of both simple parts of chemical processes and entire plants. Recent development of computer technology not only has increased greatly the speed of computation but also has led to a sharp reduction in the cost of computation.<sup>1</sup> Also, essential quantitative knowledge about chemical processes has become available. The combination of this availability of process information and cheaper computation cost have given rise to considerable interest in developing and applying mathematical techniques to determine optimum control of chemical processes.

There is a parallel interest today in direct digital control of chemical processes. The rational design of an on-line computer control system requires very careful financial and technical analysis. In the technical arena, one needs to have accurate knowledge of process dynamics plus a clear idea of

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<sup>1</sup>Wise, T. A. (1) reported that the IBM System/360 model 75 was designed to perform 375,000 multiplications per second. The first generation IBM electronic computer could perform only 2,500 multiplications per second. The cost of executing 100,000 computations would be 3-1/2¢ on the 360, compared to \$1.38 on the first generation machine.

optimum control strategy or philosophy. The present study is directed to the general question of optimum control strategy in physically distributed systems. These systems are important elements of chemical plants. Thus, the present work may be regarded as helping to provide a basis for the rational design and operation of computer control systems for chemical plants.

Chemical processes can be classified according to the number of independent variables present in the simplest mathematical model which adequately describes the dynamic behavior of a process. If there is only one independent variable - time, the system is called a lumped-parameter system. If there are two or more independent variables - such as time and space coordinates, the system is called a distributed-parameter system. The typical example of a lumped-parameter system is a perfectly mixed tank reactor; the typical example of a distributed-parameter system is a tubular reactor. The concentrations and temperature in a perfectly mixed tank reactor at any time can be represented by a finite set of quantities, while the concentrations and temperature of a tubular reactor at any time are function of distance along the reactor. The dynamic behavior of a perfectly mixed tank reactor and a tubular reactor are described by ordinary and partial differential equations respectively.

The analysis of lumped-parameter systems is obviously



simpler and the majority of previous optimization studies have accordingly been concerned with optimal lumped systems. An important exception is the formulation of a general distributed parameter problem by Butkovskii and Lerner (2) in 1960. Since then, a number of rather abstract theorems on various optimum distributed-parameter systems have been reported.

Being one of the important processes in a chemical plant, tubular reactor systems have been analyzed by several authors. Aris (3), Roberts (4) and Katz (5) have studied optimum control of steady-state tubular reactors.

The present study deals with constrained optimal controls of an unsteady-state tubular reactor system. It differs from previous studies in two important respects. First, an unsteady-state behavior of the system is taken into consideration. This is a more realistic viewpoint, since actual tubular reactors in a chemical plant never operate in a condition of steady-state but are always disturbed and continuously corrected by control actions. Second, and the key feature of this work, practical constraints on the realizability of control action have been incorporated directly into the analysis. In addition to idealistic control action which is an unconstrained function of time and space, we have incorporated a constrained form of control which is a function of time only. As one example, if temperature is considered as a manipulatable control parameter of an unsteady-state tubular reactor, the

idealistic control action is a changing temperature profile along the length of reactor. Unfortunately, even a stationary temperature profile is not practical to carry out in general, not to mention the ever-changing profile. As an approach to a more practical control, the tubular reactor is divided into one, two or three control zones. In each zone, there is a single zone temperature at any one time, uniform throughout the zone. Clearly, this is a sub-class of the "ideal" control. As a sub-class, the performance of the constrained control might be as good as that of the ideal control but can never be better.

The specific question to which this work has been directed is "what is the optimum manner in which to manipulate the constrained control variables in an unsteady-state tubular reactor system?" The control variables are constrained not only by the usual magnitude constraints but also by the requirement that the control variables be independent of position over finite region of space. The specific systems studied are idealized tubular reactors in which either reactor temperature or heating medium temperature is the constrained control variable.

## II. REVIEW OF LITERATURE

The two mathematical devices which are most widely applied to the study of optimal control of processes are Bellman's Dynamic Programming and Pontryagin's Maximum Principle.

The foundation of dynamic programming, the Principle of Optimality, was initially formulated for the multistage decision process. Later on, it was applied to the study of optimal trajectories, feedback control and certain problems associated with the calculus of variations.

In its original form, Pontryagin's Maximum Principle is tailored to treat the optimal problems of systems which are governed by a set of first order ordinary differential equations. The equations which describe a lumped system may not consist of first order equations but can always be reduced to a collection of first order ordinary differential equations by the introduction of suitable new variables. Thus, the Maximum Principle is found to be applicable to lumped systems in general, as is Dynamic Programming.

Distributed parameter systems are frequently approximated as multi-variable lumped systems. When such an approximation is no longer tolerable, it is then necessary to develop appropriate theorems applicable to the distributed system as in the present work.

The present literature review is concerned with Dynamic

Programming, the Maximum Principle and recent studies of distributed systems.

#### A. Dynamic Programming

Bellman's dynamic programming (6,7) is an efficient general method of obtaining an optimal policy for a multi-stage decision process. There is a considerable saving of calculation effort compared to the brute force or combinatorial search. However, it is not practical to apply dynamic programming directly to a process with a substantial number of state variables. For there are sharp increases not only in calculation effort but also in the required memory capacity of the computing machine with increase of number of state variables. This difficulty makes dynamic programming an unlikely candidate for the analysis of any distributed system which requires more than one or two lumped variables to approximate its dynamic behavior.

#### B. Maximum Principle

Pontryagin (8) derived the Maximum Principle (9,10,11) for a system characterized by a set of first order ordinary differential equations.

$$\frac{dx_i}{dt} = f_i(x_1, x_2, \dots, x_n, u_1, u_2, \dots, u_r)$$

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

$$(IIB. 1)$$

with initial condition  $x_i(0)$ . In equations (IIB. 1),  $x_i$  are state variables and  $u_j$  are control variables.

The function to be minimized is:

$$I = \sum_{k=1}^n c_k x_k(T) \quad (\text{IIB. 2})$$

where  $T$  is the terminal time.

The details of Pontryagin's result are not presented here. The essential character of his result is a strong necessary condition, which must be satisfied by an optimal control  $u(t)$ . He considers the optimal control  $u(t)$  and a perturbed control  $u(t) + \Delta u$ , a pattern analogous to the classical calculus of variations except that  $\Delta u$  need not be small in magnitude.

A function  $H$ , called the Hamiltonian, is introduced which is in effect a function of time  $t$ , control  $u$ , and perturbation  $\Delta u$ . In order that  $u(t)$  be the optimal control  $u$  at any fixed time  $t$ , the function  $H$  must achieve an absolute maximum with respect to  $\Delta u$  at the value  $\Delta u=0$ . In general, this is only a necessary but not a sufficient condition of the optimal control  $u(t)$ . Since the function  $H$  depends not only explicitly on  $u$  but also implicitly on the complete control function  $u(t)$  over the period of time  $0 \leq t \leq T$ , an iterative search is required to obtain an optimal control if one tries to apply the necessary condition for this purpose.

Butkovskii and Lerner (2) applied the Maximum Principle to a rather special distributed system by transforming the governing equations of the system to the form given in (IIB. 1). Similar transformation is not always possible for a general tubular reactor system.

In a later paper, Butkovskii (12) discretized space variables and approximated a distributed system with  $N$  lumped points. Pontryagin's Maximum Principle was used for its optimal solution. The approximate system approaches the distributed system with increase of number of lumped points.

### C. Contributions on Distributed-Parameter System

Butkovskii (13,14,15,16) developed maximum theorems for distributed systems which can be described by a set of nonlinear integral equations.

Wang and Tung (17) formulated various control problems. A maximum principle for the so-called terminal control problem has been derived by application of dynamic programming. The result obtained is applicable to general distributed parameter systems. Nevertheless, it is of limited use before the development of an efficient method for solving the functional equation.

Katz (18) used functional analysis to obtain a minimum principle for the end point control of a heat exchanger system.

In these papers, either weak or strong necessary condi-

tions of optimal control were obtained for various distributed-parameter systems. The system studied by Katz can be considered as a special case of the problem studied in this work. He obtained a weak minimum principle for a heat exchanger system. His result on initial control is not in a very useful form.

#### D. Contribution by Sirazetdinov

Sirazetdinov (19) derived a maximum principle for a system which is governed by a single quasilinear first order partial differential equation with many independent variables.

The variable  $x(t,y)$  is the state of the system at time  $t$  and at  $n$ -dimensional coordinate  $y$ . The variable  $u(t,y)$  is an  $r$ -dimensional controlling function with constraints:

$$\phi_k(u) \leq 0 \quad k=1,2,\dots,k_r < r \quad (\text{IID. 1})$$

The partial differential equation governing the system is:

$$\frac{\partial x}{\partial t} = f(t,y,x,x_y,u) \quad (\text{IID. 2})$$

where

$$f = f_0(t,y,x,u) + \sum_{k=1}^n f_k(t,y,x,u)x_{y_k} \quad (\text{IID. 3})$$

and

$$x_{y_k} \equiv \frac{\partial x}{\partial y_k} \quad k=1,2,\dots,n \quad (\text{IID. 4})$$

The initial condition is given by

$$x(0,y) = x_0(y) \quad (\text{IID. 5})$$

The boundary condition at the hypersurface  $S_1$  is:

$$x(t,y)|_{S_1} = x_1(t,y) \quad (\text{IID. 6})$$

If the system is one-dimensional, the system occupies an interval  $0 \leq y \leq Y$ . The boundary condition at the end of interval  $y=0$  is given, while the condition at the other end  $y=Y$  is not given. Similarly, if the system is  $n$ -dimensional, the system occupies an  $n$ -dimensional space  $S$ . The region occupied is bounded by a surface composed of parts  $S_1$  and  $S_2$ . The boundary condition at one hypersurface  $S_1$  is given, while that at the complementary surface  $S_2$  is not given.

The performance index is expressed as

$$I_1 = \int_0^T \int_S g \, ds \, dt \quad (\text{IID. 7})$$

where

$$g = g_0(t,y,x,u) + \sum_{k=1}^n g_k(t,y,x,u) x_{y_k} \quad (\text{IID. 8})$$

Sirazetdinov formulates two optimal problems with the performance index  $I_1$ .

Problem 1: Find the optimal controlling function  $u(t,y)$  which minimizes the functional  $I_1$ .

Problem 2: Find the space independent optimal controlling



function  $u(t)$  which minimizes the functional  $I_1$ .

He introduces a function  $H$ :

$$H = H(t, y, x, x_y, p, u) = -g + pf \quad (\text{IID. 9})$$

where  $p$  satisfies the equation

$$\frac{\partial p}{\partial t} = - \frac{\partial H}{\partial x} + \sum_{k=1}^n \frac{\partial}{\partial y_k} \left( \frac{\partial H}{\partial x_{y_k}} \right) \quad (\text{IID. 10})$$

and initial and boundary conditions

$$p(T, y) = 0 \quad (\text{IID. 11})$$

$$\left\{ \sum_{k=1}^n \frac{\partial H}{\partial x_{y_k}} \cos(\widehat{ny_k}) \right\}_{S_2} = 0 \quad (\text{IID. 12})$$

Condition (IID. 12) is equivalent to

$$p(t, y) |_{S_2} = \left( \frac{\sum_{k=1}^n g_k \cos(\widehat{ny_k})}{\sum_{k=1}^n f_k \cos(\widehat{ny_k})} \right)_{S_2} \quad (\text{IID. 13})$$

$\cos(\widehat{ny_k})$  are the direction cosines of the outer normal  $\bar{n}$  to the hypersurface  $S_2$ .

**Theorem 1:** If the controlling function  $u(t, y)$  yields the minimum of the functional  $I_1$ , then at any  $t(0 \leq t \leq T)$  and  $y(y \in S)$   $H$  attains its absolute maximum with respect to  $u$  at  $u = u(t, y)$ . i.e.,

$$\Delta H = H(t, y, x, x_y, p, u + \Delta u) - H(t, y, x, x_y, p, u) \leq 0 \quad (\text{IID. 14})$$

where  $u$  represents  $u(t, y)$  and  $\Delta u$ , any variation.

Theorem 2: If the controlling function  $u$  is a function of  $t$  only, and if  $u(t)$  yields the minimum of the functional  $I_1$ , then  $H^*$  attains its absolute maximum with respect to  $u$  at  $u=u(t)$ , at any instant  $t(0 \leq t \leq T)$ .  $H^*$  is defined by

$$H^* = \int_S H(t, s, x, x_y, p, u) \, ds \quad (\text{IID. 15})$$

Theorem 2 can be stated as:

$$\Delta H^* = H^*(u + \Delta u) - H^*(u) \leq 0 \quad (\text{IID. 16})$$

If the controlling function is a function of  $y$  alone, and if the performance index is given by  $I_2$ , another problem is formulated:

Problem 3: Find the time independent optimal controlling function  $u(y)$  which minimizes the functional  $I_2$ .

$$I_2 = \int_S g'(s, x(T, s), u(s)) \, ds \quad (\text{IID. 17})$$

Let

$$H_1 = pf \quad (\text{IID. 18})$$

and

$$\frac{\partial p}{\partial t} = \sum_{k=1}^n \frac{\partial}{\partial y_k} \left( \frac{\partial H_1}{\partial x_{y_k}} \right) - \frac{\partial H_1}{\partial x} \quad (\text{IID. 19})$$

$$p(T, y) = - \frac{\partial g'}{\partial x} \quad (\text{IID. 20})$$

$$p(t, y) |_{S_2} = 0 \quad (\text{IID. 21})$$

The function  $H^{**}$  which must attain an absolute maximum is defined as:

$$H^{**} \equiv -g' + \int_0^T H_1 dt \quad (\text{IID. 22})$$

Theorem 3: If a time independent control function  $u(y)$  yields the minimum of the functional  $I_2$ , then  $H^{**}$  attains its absolute maximum with respect to  $u$  at  $u=u(y)$  at any point  $y$  in the region  $S$ .

Linear system: The system is considered as linear, if it is governed by the linear partial differential equation:

$$\frac{\partial x}{\partial t} = a_0(t, y)x + \sum_{k=1}^n a_k(t, y)x_{y_k} + a_{n+1}(t, y, u) \quad (\text{IID. 23})$$

and the integrands  $g$  and  $g'$  are given by

$$g = b_0(t, y)x + \sum_{k=1}^n b_k(t, y)x_{y_k} + b_{n+1}(t, y, u) \quad (\text{IID. 24})$$

and

$$g' = b'_0(y)x(T, y) + b'_1(y, u) \quad (\text{IID. 25})$$

Theorem 4: For the linear system, the maximum principles given in Theorems 1, 2, and 3 are both necessary and sufficient conditions.

Sirazetdinov obtained strong necessary conditions for the

particular system he studied. The theorems developed by Sirazetdinov can be used as a local test of a control to determine whether it could be part of the optimal control of the system. If these necessary conditions are utilized for finding an optimal control, an iterative search has to be carried out.

Sirazetdinov's method of obtaining an increment  $\Delta I$  corresponding to change of control variable by  $\Delta u$ , is applied to the tubular reactor system in this work.

The pattern of proof in Sirazetdinov's work was originally given by Rozonoër (9) for proving Pontryagin's Maximum Principle. The same pattern of proof is also adopted here for proving certain theorems applicable to the problem of determining optimum constrained control of a tubular reactor.

### III. THEORETICAL DEVELOPMENT

#### A. Introduction

In this chapter, an optimal control problem of an unsteady-state tubular reactor system is formulated. Then, theorems concerning different forms of control variables as well as some examples of possible applications are given.

The question raised in this chapter is "what is the 'best' way of operating an unsteady-state tubular reactor system, if the control variables are constrained?" The approach taken to answering this question has involved transposition of the question into mathematical form.

The reduction of the above question to mathematical form involves three major parts. First, an adequate mathematical model of the unsteady-state tubular reactor is established in terms of appropriate partial differential equations. Second, physical limitations upon this system are recognized and expressed by appropriate constraint statements. Third, the 'best' operation is defined by giving a functional rule which may be used to compute a single performance index for any candidate mode of operation.

In developing the mathematical model for the dynamic tubular reactor system, the standard simplifying assumptions such as plug flow, constant fluid properties etc. are employed. With these assumptions, a set of linear first order partial

differential equations may be used to describe the dynamic behavior of a tubular reactor system.

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### B. Mathematical Model

A material balance and energy balance of a tubular reactor give a set of linear first order partial differential equations. Rather standard assumptions are used to obtain this result. These assumptions are itemized as follows:

1. Plug flow is assumed ;
2. Radial gradients of concentration and temperature are assumed to be absent;
3. Longitudinal mass diffusion and thermal conduction are assumed to be negligible compared to the convective transport of mass and thermal energy;
4. The change in kinetic energy and potential energy are also assumed to be negligible;
5. Fluid densities, average specific heats of the fluids, heats of reactions and overall heat transfer coefficient are assumed to be constant.

Mass balances yield equations:

$$\frac{\partial x_i}{\partial t} + v(t) \frac{\partial x_i}{\partial y} = r_i(x, T_r, P)$$

$$i=1,2,\dots,n-2 \quad \text{(IIIB. 1)}$$

where  $t$  is time;  $y$ , distance from tubular reactor inlet;

$x_i(t,y)$ , mass concentration of component  $i$  per unit volume of solution;  $v(t)$  is the axial velocity of the fluid;  $r_i$ , the kinetic expression for the production of component  $i$  per unit time;  $x$  is a vector with  $n-2$  elements,  $x_1, x_2, \dots, x_{n-2}$ ;  $T_r$  is the temperature of the reactant fluid;  $P$  is the pressure of the fluid.

An energy balance leads to the equation:

$$\frac{\partial T_r}{\partial t} + v(t) \frac{\partial T_r}{\partial y} = \frac{2U_T}{R_t \rho C_p} (T_h - T_r) + \frac{1}{\rho C_p} \sum_{j=1}^m R_j(x, T_r, P) (-\Delta H_j) \quad (\text{IIIB. 2})$$

where  $T_h$  is the temperature of the heating fluid or temperature of the furnace;  $U_T$ , the overall heat transfer coefficient;  $R_t$ , radius of the tube;  $\rho$ , density of the reactant fluid;  $C_p$ , average heat capacity of the reactant fluid;  $R_j$ , rate of  $j$ th reaction in relation to a unit stoichiometric coefficient;  $(-\Delta H_j)$ , heat generated by the  $j$ th reaction per unit of the rate  $R_j$ .

The kinetic expression in the mass balance can be related to the rate of  $j$ th reaction  $R_j$  in the following form:

$$r_i(x, T_r, P) = \sum_{j=1}^m s_{ij} R_j(x, T_r, P) \quad (\text{IIIB. 3})$$

where  $s_{ij}$  is the stoichiometric coefficient of component  $i$  in the  $j$ th reaction. For example, two simultaneous reactions



can be rearranged as

$$3C - A - 2B = 0 \quad (\text{Reaction 1}) \quad (\text{IIIB. 6})$$

$$D - B - C = 0 \quad (\text{Reaction 2}) \quad (\text{IIIB. 7})$$

The stoichiometric coefficients for various components are

$$\text{Component A (or component 1): } s_{11} = -1 \quad s_{12} = 0$$

$$\text{Component B (or component 2): } s_{21} = -2 \quad s_{22} = -1$$

$$\text{Component C (or component 3): } s_{31} = 3 \quad s_{32} = -1$$

$$\text{Component D (or component 4): } s_{41} = 0 \quad s_{42} = 1$$

If the heating is done by co-current or countercurrent flow of a heating fluid, an equation is required to describe the dynamics of the temperature of heating medium.

$$\frac{\partial T_h}{\partial t} + v_h(t) \frac{\partial T_h}{\partial y} = \frac{2\pi R_t U_T}{A' \rho' C'_p} (T_r - T_h) \quad (\text{IIIB. 8})$$

where  $v_h(t)$  is the axial velocity of the heating fluid;  $A'$  is the cross sectional area for the flow of heating fluid;  $\rho'$  and  $C'_p$  are the average density and heat capacity for the heating fluid.

Thus, the dynamics of tubular reactors are governed by a set of linear first order partial differential equations.



$$\frac{\partial x_i}{\partial t} + v_i(t) \frac{\partial x_i}{\partial y} = r_i(x, u) \quad i=1, 2, \dots, n \quad (\text{IIIB. 9})$$

where the state vector  $x$  is an  $n$ -dimensional vector. The state vector is determined from the solution of this set of equations with known initial and boundary conditions. The first  $n-2$  elements of the state vector  $x$ , represent the concentration of the components in the reactor; the  $(n-1)$ th element,  $x_{n-1}$ , represents the temperature of the reactants; the  $n$ th element,  $x_n$ , represents the temperature of the heating medium (or coolant). The control vector  $u$  is a variable that can be chosen as is required. The elements of control vector  $u$  may assume one of three forms,  $u_j^t(t)$ ,  $u_k''(t, y)$  and  $u_m^y(y)$ . The control  $u_j^t(t)$  is a function of time only and exerts its control action over the space interval  $[Y_{ja}, Y_{jb}]$  at any time  $t$ . The control  $u_k''(t, y)$  is a function of both time and space, and exerts its action only at a point  $y$  at any time  $t$ . The control  $u_m^y(y)$  is a function of space only and exerts its control over the time interval  $[T_{ma}, T_{mb}]$ , at any point  $y$  on the space.

If the flow rates of both reactant fluid and heating medium are control variables or either of them is the control variable, the whole set of  $n$  equations are required to describe the system adequately.

In case the temperature of the reactant is the control variable  $u$ , only the first  $n-2$  equations are necessary for

describing the dynamics of the system. The state vector  $x$  is then a vector with  $n-2$  elements.

If the temperature of the heating medium is the control variable  $u$ , then the first  $n-1$  equations are sufficient to describe the dynamics of the process. The state vector  $x$  is an  $n-1$  dimensional vector.

Generally, the functions  $r_i$  are complicated expressions of  $x$  and  $u$ . Except for very simplified cases, a numerical integration is required for the solution of the set of equations. The characteristic equations for the system are a set of first order ordinary differential equations. The Runge-Kutta-Gill (20) integration procedure has been used for solution of the characteristic equations. This method is abstracted in Appendix E.

### C. Constraints

Physical limitations of quality and design requirements are expressed in the constraints. All of the constraints given below have been considered in the theoretical development and some in the numerical examples.

In some cases, it might appear that the natural way to express a constraint would be in terms of a stochastic function. For example, if an integral constraint is a certain function of an outlet concentration  $x_1(t, Y)$ , we might reasonably require that  $x_1$  have certain stochastic properties. However, in the

present work, only deterministic functions have been considered.

Various constraints are discussed as follows:

(a). Constraints on the form of control function may be imposed by design requirements. For example, if the temperature of the reactant is the control variable in a tubular reactor and if the reactor is subdivided into three control zones such that in each zone there is a single uniform temperature at any time, then the control variable  $T_r(t,y)$  can be represented by a set of three space independent functions  $T_i^t(t)$ .

$$T_r(t,y) = \begin{cases} T_1^t(t) & 0 \leq y \leq Y_1 \\ T_2^t(t) & Y_1 < y \leq Y_2 \\ T_3^t(t) & Y_2 < y \leq Y \end{cases}$$

where  $Y_1$  and  $Y_2$  are boundaries between zone 1 and zone 2 and between zone 2 and zone 3 respectively;  $Y$  is the outlet of the reactor.

Similarly, several elements of the control vector may be constrained to a form which may be represented by a set of time independent functions  $u_j^y(y)$ .

(b). Constraints on the magnitude of control are always present in physical systems, and may be expressed by relations of the form:

$$\phi_k(u) \leq 0$$

For example, upper and lower limits of temperature may exist for a system either due to the energy source or for the preservation of catalyst activity.

$$T_a \leq T_r \leq T_b$$

Because the control variable may assume a value on the boundary of the admissible range, it is important to consider the closed and bounded control region. This makes the problem different from the one usually considered in the classical calculus of variations where a control can only take values from an open control region.

$$(c) . \quad \int_0^T \int_0^Y f_i(t, y, x, u) \, dy dt \leq L_i$$

where  $L_i$  is a constant.

If  $f_i$  represents rate of energy transfer at point  $y$ , this form of integral constraint expresses the limited amount of total energy available to the system.

$$(d) . \quad \int_0^T g_i(t, x(t, 0), x(t, Y), u(t)) \, dt \leq M_i$$

If the average amount of impurities in the end products has to be less than a certain value, it is expressed in this form of constraint. For example

$$\int_0^T x_1(t, Y) \, dt < 0.03$$

$$(e) . \quad \int_0^Y h_1(y, x(0, y), x(T, y), u(y)) \, dy \leq M_1^i$$

In a start-up operation, if  $x$  represents the temperature of reactants in a tubular reactor, and if the temperature profile at the terminal time  $T$  has to be as near to a steady-state profile as possible, this type of constraint limits an average deviation from the steady-state profile. For example

$$\int_0^Y [x_1(T, y) - T_m(y)]^2 \, dy < 10$$

where  $x_1(T, y)$  is the actual terminal temperature profile and  $T_m(y)$  is a given function describing a steady-state profile.

#### D. Performance Indices

A performance index gives a measure to the achievement of control effort. By definition an optimal control will produce the minimum value of the performance index. The form of the performance index depends on the objective of the control effort. It may be related to the cost of operation. It may represent the deviation from a steady-state operation. The form of performance index adopted may require compromise between the form motivated physically and the form treatable by the optimal theories.

For example, we might like to have a minimum of the positive quantity:

$$I_1 = \max_{0 \leq t \leq T} x_1(t, Y)$$

but for the sake of mathematical treatment,  $I_1$  may be replaced by  $I_2$

$$I_2 = \int_0^T x_1^2(t, Y) dt$$

If the cost of producing species 2 from species 1 is the objective of minimization, and if the cost of the production is linearly proportional to control  $u$  and to concentrations  $x_1$ ,  $-x_2$  in the end product, then the performance index might be expressed as

$$I_3 = \int_0^T [x_1(t, Y) - c_1 x_2(t, Y)] dt + \int_0^T \int_0^Y c_2 u(t, y) dy dt$$

where  $c_1$  and  $c_2$  imply cost relative to a unit cost of  $x_1(t, Y)$ .

The integrand in a performance index does not have to be linear with respect to state and control variables. The performance index which will be considered in this work has a general form.

$$I = \int_0^T \int_0^Y f_0(t, y, x(t, y), u(t, y), u^t(t), u^y(y)) dy dt \\ + \int_0^T g_0(t, x(t, 0), x(t, Y), u^t(t)) dt$$

$$+ \int_0^Y h_0(y, x(0, y), x(T, y), u^Y(y)) dy$$

### E. Description of the Problem

The main question raised in this work is "what is the 'best' way to control a tubular reactor if the control and the system are subjected to certain constraints?" For the purpose of answering the question, we have to define the meaning of 'best' by the minimum of a certain quantity called a performance index; at the same time, we have to develop the dynamic model of the system; finally, we have to define the various constraints imposed on the system. Then, an optimal control is sought for the system.

In searching for optimal control of a tubular reactor system, one may approximate the system by a lumped system with many state variables and then find the optimal control by the application of optimal theories on lumped systems. Dynamic programming and the Maximum Principle are the two most widely used methods for lumped system solutions.

Because the lumped system which approximates a distributed system, inherently consists of many state variables, a direct application of dynamic programming is out of the question due to the requirements of excessive calculation time and large memory capacity of a computing machine. The application of the Maximum Principle coupled with an efficient method of

iterative maximum search appears to be a better way of handling this problem.

Instead of approximating the tubular reactor system by a lumped system, one may develop optimal control theorems for the distributed system, then find the optimal control by the application of these theorems. This kind of direct formulation maintains the rigor of the problem statement. Though the practical calculation may involve approximation analogous to the lumped problem, the method of solution is not necessarily limited to the physical lumping.

A general optimal problem for a tubular reactor system will be stated below. Necessary conditions for various forms of control variable will be given in the next section. Details of the derivation and proofs are given in Appendices A to C. The derivation of the expression for increment  $\Delta I$  follows that of Sirazetdinov (19), the pattern of proof was originally given by Rozonoër (9).

The dynamic behavior of a tubular reactor system can be adequately described by a set of  $n$  linear first order partial differential equations:

$$\frac{\partial x_i}{\partial t} = f_i(t, y, x, u', \frac{\partial x}{\partial y})$$

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

(IIIE. 1)



where

$$f_i = -v_i(t) \frac{\partial x_i}{\partial y} + r_i(t, y, x, u^i) \quad (\text{IIIE. 2})$$

The vector  $x=(x_1, x_2, \dots, x_n)$  is an  $n$ -dimensional vector, called the state vector. It represents the state of the system at the instant  $t$  and space  $y$ , or the state at the coordinate  $(t, y)$ . The vector  $u=(u_1, u_2, \dots, u_r)$  is an  $r$ -dimensional vector, called the control vector. The axial velocities  $v_i$  can either be given functions or be elements of control vector.

The control vector  $u$  has  $r$  constraints:

$$\phi_k(u) \leq 0 \quad k=1, 2, \dots, r \quad (\text{IIIE. 3})$$

These constraints specify control region  $U$ . Set  $U$  is the closed, bounded set with  $\phi_k=0 (k=1, 2, \dots, r)$  as its boundaries. An admissible control  $u$  is defined as a piecewise continuous function that has values from set  $U$ . The closed set  $U$  makes the problem non-classical, for  $u$  can assume values on the boundary of  $U$ . The piecewise continuous control is considered because it is sufficient for engineering application, even though Pontryagin considered a somewhat broader class of controls in his basic development (8).

In the present work, the control vector  $u$  consists of  $r$  elements which can be formed into five subgroups  $u^B(t)$ ,  $u^t(t)$ ,  $u''(t, y)$ ,  $u^y(y)$  and  $u^I(y)$ .

The subgroup  $u^B(t)$  is a function of time and is called boundary control as its elements exert their action through the boundary conditions of the system. The element  $u_j^t(t)$  of subgroup  $u^t$  exerts its control on the interval of space coordinate  $Y_{ja} \leq y \leq Y_{jb}$  at the instant of time  $t$ .  $Y_{ja}$  and  $Y_{jb}$  are constants which depend on subscript  $j$ . The element  $u_k''(t, y)$  of subgroup  $u''$  exerts its control on the point  $y$  at time  $t$ . The element  $u_k^y(y)$  of subgroup  $u^y$  exerts its action on a point  $y$  over the time interval between  $T_{ka}$  and  $T_{kb}$ .  $T_{ka}$  and  $T_{kb}$  are constants which depend on subscript  $k$ . The subgroup  $u^I(y)$  is a function of coordinate  $y$  and is called initial control. The elements of initial control exert their action through initial conditions of the system. The notation  $u'$  in the equation (IIIE. 1) consists of subgroups  $u^t$ ,  $u''$  and  $u^y$ .

The initial conditions of the system are given by:

$$x_i(0, y) = x_{i0}(y, u^I) \quad i=1, 2, \dots, n \quad (\text{IIIE. 4})$$

The boundary conditions are given by:

$$x_i(t, 0) = x_{i1}(t, u^B) \quad i=1, 2, \dots, n' \quad (\text{IIIE. 5})$$

$$x_i(t, Y) = x_{i1}(t, u^B) \quad i=n'+1, n'+2, \dots, n \quad (\text{IIIE. 6})$$

where  $n'$  is a positive integer less than or equal to  $n$ ;  $x_{i0}$  are given functions of  $y$  and  $u^I$ ;  $x_{i1}$ , given functions of  $t$  and  $u^B$ .

In addition to constraints (IIIE. 3), there are integral

constraints of the following forms:

$$\int_0^T \int_0^Y f_i(t, y, x, u') \, dy dt \leq L_i$$

$$i = n+1, n+2, \dots, n+q \quad (\text{IIIE. 7})$$

$$\int_0^T g_i(t, x(t, 0), x(t, Y), u^B, u^t) \, dt \leq M_i$$

$$i = 1, 2, \dots, s' \quad (\text{IIIE. 8})$$

$$\int_0^Y h_i(y, x(0, y), x(T, y), u^I, u^Y) \, dy \leq M_i'$$

$$i = 1, 2, \dots, s'' \quad (\text{IIIE. 9})$$

where  $T$  is the end of time under considerations;  $Y$  is the end point of space coordinate;  $L_i$ ,  $M_i$  and  $M_i'$  are constants.

The performance index of the system has the form

$$I = -w_0 \int_0^T g_0(t, x(t, 0), x(t, Y), u^B, u^t) \, dt$$

$$- w'_0 \int_0^Y h_0(y, x(0, y), x(T, y), u^I, u^Y) \, dy$$

$$- p_0 \int_0^T \int_0^Y f_0(t, y, x(t, y), u') \, dy dt \quad (\text{IIIE. 10})$$

where  $w_0$ ,  $w'_0$  and  $p_0$  are constants with value  $-1$ .

The admissible control which yields the minimum of the performance index  $I$  is called optimal control. In view of the system stated above, one may pose a general optimal problem

given below.

Problem: For this tubular reactor system, find control vector  $u = (u^B(t), u^t(t), u''(t, y), u^Y(y), u^I(y))$  which minimizes the performance index  $I$ .

The different forms of control have their own characteristics, thus different optimum theorems are developed. Since every element of control can be varied independently of others, it is possible to consider one element or one form of control at a time. We consider the theorems developed in the next section as an extension to the Sirazetdinov's work (19), though Katz (18) has also obtained a weak form of partial result.

#### F. Optimum Theorems - Extension of Sirazetdinov's Work

For the tubular reactor system, we introduce the Hamiltonian function  $H$ .

$$H = \sum_{j=0}^{n+q} p_j f_j \quad (\text{IIIF. 1})$$

The first  $n$  elements  $p_i$  ( $i=1, 2, \dots, n$ ) of the auxiliary variable  $p$  are defined by the equations:

$$\frac{\partial p_i}{\partial t} = -v_i \frac{\partial p_i}{\partial y} - \sum_{j=0}^{n+q} p_j \frac{\partial f_j}{\partial x_i} \quad i=1, 2, \dots, n \quad (\text{IIIF. 2})$$

The terminal and boundary conditions for  $p_i$  are given by

$$p_i(T, Y) = \sum_{j=0}^{s''} w'_j \frac{\partial h_j}{\partial x_i(T, Y)}$$

$$i=1, 2, \dots, n \quad (\text{IIIF. 3})$$

$$p_i(t, Y) = \frac{1}{v_i} \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_i(t, Y)}$$

$$i=1, 2, \dots, n' \quad (\text{IIIF. 4})$$

$$p_i(t, 0) = - \frac{1}{v_i} \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_i(t, 0)}$$

$$i=n'+1, n'+2, \dots, n \quad (\text{IIIF. 5})$$

The Lagrange multipliers for the system, the remaining  $p_i$ 's and  $w_i$ 's,  $w'_i$ 's are given by

$$p_i(t, Y) = c_i \quad i=n+1, n+2, \dots, n+q \quad (\text{IIIF. 6})$$

$$w_i = d_i \quad i=1, 2, \dots, s' \quad (\text{IIIF. 7})$$

$$w'_i = d'_i \quad i=1, 2, \dots, s'' \quad (\text{IIIF. 8})$$

where  $c_i$ ,  $d_i$  and  $d'_i$  are constants. The constants must be so chosen that they are consistent with the constraint conditions (IIIE. 7), (IIIE. 8) and (IIIE. 9) and with the requirement that performance index  $I$  is minimum. In general, a numerical search would be required.

Equations (IIIE. 1) and (IIIF. 2) can also be written in terms of  $H$  as

$$\frac{\partial x_i}{\partial t} = \frac{\partial H}{\partial p_i} \quad i=1,2,\dots,n \quad (\text{IIIF. 9})$$

$$\frac{\partial p_i}{\partial t} = -v_i \frac{\partial p_i}{\partial y} - \frac{\partial H}{\partial x_i} \quad i=1,2,\dots,n \quad (\text{IIIF. 10})$$

Another four Hamiltonian functions are introduced. Every Hamiltonian function corresponds to a form of control parameter discussed before

$$H^t = \sum_{j=0}^{s'} w_j g_j + \int_0^Y H \, dy \quad (\text{IIIF. 11})$$

$$\begin{aligned} H^B = & \sum_{j=1}^{n'} [p_j(t,0) v_j(t) + \sum_{k=0}^{s'} w_k \frac{\partial g_k}{\partial x_j(t,0)}] x_{j1}(t, u^B) \\ & + \sum_{j=n'+1}^n [-p_j(t,Y) v_j(t) + \sum_{k=0}^{s'} w_k \frac{\partial g_k}{\partial x_j(t,Y)}] x_{j1}(t, u^B) \\ & + \sum_{j=0}^{s'} w_j g_j \end{aligned} \quad (\text{IIIF. 12})$$

$$H^Y = \sum_{j=0}^{s''} w_j^! h_j + \int_0^T H \, dt \quad (\text{IIIF. 13})$$

$$\begin{aligned}
H^I = & \sum_{j=1}^n [p_j(0,y) + \sum_{k=0}^{s''} w'_k \frac{\partial h_k}{\partial x_j(0,y)}] x_{j0}(y, u^I) \\
& + \sum_{j=0}^{s''} w'_j h_j
\end{aligned} \tag{IIIF. 14}$$

For the tubular reactor system, the following theorems are asserted. Proof of the theorems is given in Appendices A to C.

Theorem 1: For the system with  $u''$  as its control variable, if  $u''(t,y)$  is the optimal control, then at almost any  $t(0 \leq t \leq T)$  and almost any  $y(0 \leq y \leq Y)$ ,  $H$  attains its absolute maximum with respect to  $u''$  at  $u'' = u''(t,y)$ . i.e.,

$$H(t,y,u''(t,y)) = \sup_{u'' \in U} H(t,y,u'') \tag{IIIF. 15}$$

Theorem 2: For the system with  $u^t$  as its control variable, if  $u^t(t)$  is the optimal control, then at almost any  $t(0 \leq t \leq T)$ ,  $H^t$  attains its absolute maximum with respect to  $u^t$  at  $u^t = u^t(t)$ .

Corollary: For the system with  $u^Y$  as its control variable, if  $u^Y(y)$  is the optimal control, then at almost any  $y(0 \leq y \leq Y)$ ,  $H^Y$  attains its absolute maximum with respect to  $u^Y$  at  $u^Y = u^Y(y)$ .

Theorem 3: For the system with  $u^B$  as its control variable, if  $u^B(t)$  is the optimal control, then at almost any  $t(0 \leq t \leq T)$ ,  $H^B$  attains its relative maximum with respect to  $u^B$  at  $u^B = u^B(t)$ .

Corollary: For the system with  $u^I$  as its control variable, if  $u^I(y)$  is the optimal control, then at almost any  $y(0 \leq y \leq Y)$ ,  $H^I$  attains its relative maximum with respect to  $u^I$  at  $u^I = u^I(y)$ .

Linear system: The system is considered linear if  $f_i$ ,  $g_i$  and  $h_i$  have the following forms:

$$f_i = -a_i(t) \frac{\partial x_i}{\partial y} + \sum_{j=1}^n b_{ij}(t, y) x_j + r_i(t, y, u')$$

$$i=1, 2, \dots, n \quad (\text{IIIF. 16})$$

$$f_i = \sum_{j=1}^n b_{ij}(t, y) x_j + r_i(t, y, u')$$

$$i=n+1, n+2, \dots, n+q \quad (\text{IIIF. 17})$$

$$g_i = \sum_{j=1}^n c_{ij}(t) x_j(t, Y) + \sum_{j=1}^n d_{ij}(t) x_j(t, 0)$$

$$+ r'_j(t, u^B, u^t) \quad i=0, 1, \dots, s' \quad (\text{IIIF. 18})$$



$$h_i = \sum_{j=1}^n c_{ij}'(y) x_j(T, y) + \sum_{j=1}^n d_{ij}'(y) x_j(0, y) + r_i''(y, u^I, u^Y) \quad i=0, 1, \dots, s'' \quad (\text{IIIF. 19})$$

Theorem 4: For the linear system, the maximum principles of Theorems 1, 2, 3 and Corollaries are both necessary and sufficient.

In the derivation of the theorems, the state variables  $x_i$  and auxiliary variables  $p_i$  are assumed to be piecewise continuous. The only permissible lines of discontinuity are the curves represented by

$$y - \int_0^t v_i \, dt = c_i \quad (\text{IIIF. 20})$$

where  $c_i$  are constants.

The consideration of a system with piecewise continuous  $x_i$  and  $p_i$  instead of a system with continuous  $x_i$  and  $p_i$  has practical significance since one may wish to treat a system with a discontinuous boundary condition such as a unit step function. Also, this simplifies the expression of a suitable performance index, because some expressions of performance indices may give rise to discontinuity of auxiliary variables and require optimal theorems which are derived with the assumption that the auxiliary variables are piecewise continuous. In previous work by Sirazetdinov, continuity of

variables  $x$  and  $p$  was implied.

The necessary conditions obtained for the optimal system require the absolute maxima of certain functions for control elements belonging to subgroups  $u^t$ ,  $u''$  and  $u^y$ , while only relative maxima of certain functions are required for control elements belonging to subgroups  $u^B$  and  $u^I$ .

Compared with Sirazetdinov's optimum theorems (19), the extension given above can be applied to a tubular reactor system with many state variables, while his theorems can only treat a system with one state variable. In addition to the optimum theorems for three forms of control  $u^t(t)$ ,  $u''(t,y)$ ,  $u^y(y)$  which are present in Sirazetdinov's work, we have found optimum theorems for boundary control  $u^B(t)$  and initial control  $u^I(y)$ . The inclusion of boundary control is especially important because of its practical application. For example, one may try to manipulate the inlet concentrations of reactants in a system in order to obtain a desired quality in the product.

Besides the magnitude constraints for control variables, three forms of integral constraints are incorporated in the system and thus the optimum theorems in this extension can be applied to the systems with these constraints, while Sirazetdinov's theorems can not. Moreover, we adopt an unified performance criterion for this extension, and the optimum theorems in the extension can be applied to a variety of

problems.

### G. Piecewise Constant Control

If the tubular reactor system discussed in the section IIIIE can have only a special form of time dependent control - a spatial independent function piecewise constant over different time intervals, a theorem similar to but weaker than Theorem 2 of section IIIF can be asserted.

This form of control is simpler and hence is more practical than a control with continuously changing magnitude. Also, in an iterative search for an optimal control, this form of control is simpler and requires less calculation compared to the continuously changing control function. Therefore, we shall derive an optimum theorem corresponding to this special form of function.

The element  $u^t$  is defined over N periods of time by

$$u_i^t(t) = \begin{cases} a_{i1} & T_0 \leq t < T_1 \\ a_{i2} & T_1 \leq t < T_2 \\ \dots\dots\dots & \\ \dots\dots\dots & \\ a_{iN} & T_{N-1} \leq t < T_N \end{cases} \quad i=1,2,\dots,r \quad (\text{IIIG. 1})$$

where  $a_{ij}$ 's are any arbitrary constant values within the region U.

We introduce a function

$$H_j^{t*} = \int_{T_{j-1}}^{T_j} H^t dt \quad j=1,2,\dots,N \quad (\text{IIIG. 2})$$

Theorem 5: If the admissible control  $u^t = u^t(t)$  yields the minimum of the functional  $I$ , then  $H_j^{t*}$  ( $j=1,2,\dots,N$ ) attains a relative maximum with respect to  $u^t$  at  $u^t = u^t(t)$ .

Since every variation of control  $\Delta u^t$  has to extend over a distance  $Y_{ia} \leq Y \leq Y_{ib}$ , and has to persist for a duration of time between  $T_{j-1}$  and  $T_j$ ,  $\Delta u^t$  has to be of certain smallness in order that the maximum principle for this control holds true. The maximum attained in the theorem stated above is only a relative one.

The proof of Theorem 5 is given in Appendix D.

## H. Possible Applications

The theorems developed in this thesis can be used for finding optimal control of a system governed by a set of linear first order partial differential equations. These include systems with configuration similar to a tubular reactor, such as packed towers, double tube heat exchangers, etc.

Some possible applications will be listed.

- (1). Find an optimal control  $u(t,y)$  for a tubular reactor.

The control  $u(t,y)$  is a function of both time and dis-

tance.

- (2). Find an optimal control  $u(t)$  for a tubular reactor.

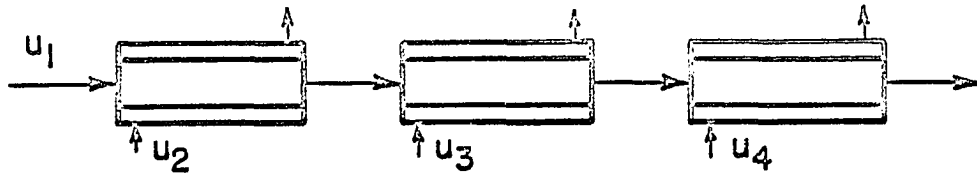
The control  $u(t)$  has  $r$  control elements. The  $j$ th element  $u_j$  exerts its control over a region  $Y_{ja} \leq y \leq Y_{jb}$ . The control regions for a different control element might partly or completely overlap one another. If there is no overlap between different control regions, then the system becomes a reactor with  $r$  control zones in series.

- (3). Find an optimal control  $u(t)$  for tubular reactors with heat exchangers.

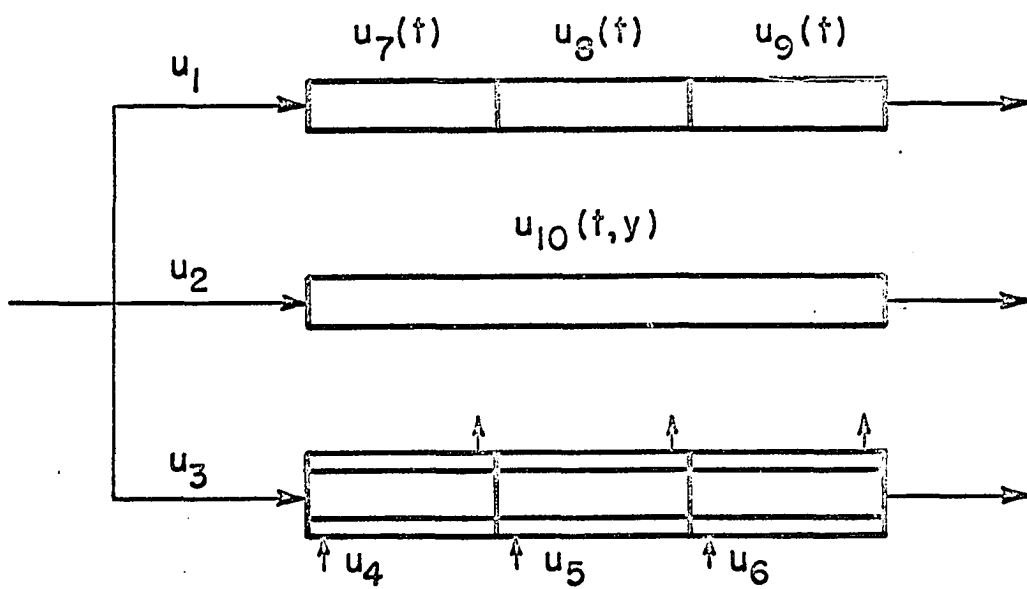
The  $u_j$ 's represent the flow rates of reactant fluid and coolant. See Figure (1a). This kind of arrangement may include tubular reactors coupled together thermally.

- (4). Find an optimal control  $u$  for a parallel-series reactor.

The control variables in this case may include flow rate  $u(t)$  ( $i=1,2,\dots,6$ ) and other zone control  $u_i(t)$  ( $i=7,8,9$ ), and ideal control  $u_{10}(t,y)$ . See Figure (1b) for the control zones for different control elements. The optimization of a group of reactors such as shown on Figure (1b) should involve considering the system as a whole. It is well known that the optimum control of the subdivided small plants may not lead to the optimum of the whole system. The present theorems are in a form which applies to over-all optimization of an entire system.



(a). 3-zone tubular reactor with heat exchanger



(b). Parallel - series reactors

Figure 1. Examples of applications

(5). Find an optimal control for a multi-stage system when there is a great number of stages.

The dynamics of a multi-stage system approach that of a plug-flow system as the number of stages increases. Hence, it is possible to find an "almost" optimal control for the system through the application of the theorems if the number of stages involved is quite large.

(6). Find a best way to manipulate the inlet concentration of one reactant for a tubular reactor operating at a pre-assigned temperature.

#### IV. PRESENTATION AND DISCUSSION OF RESULTS

##### A. Introduction

Several applications of the optimal theories developed in the previous chapter are given here.

For the first application, the theories are applied to an unsteady-state tubular reactor system with a single reversible chemical reaction. Conditions which must be satisfied by the unconstrained and constrained forms of temperature policies are obtained.

The second case is the flow rate control of a heat exchanger system. We find that bang-bang control is the mode of optimum operation.

As to the third case, we have linearized a general system with respect to a steady-state condition.

Finally, the numerical study of several problems are given. Among them are problems concerning tubular reactor systems with familiar first order consecutive reaction A to B to C.

The four problems studied numerically are the optimal control of the ideal reactor where control is function of both time and distance and that of one-, two-, three-zone reactors where a tubular reactor is subdivided into one, two, and three control zones respectively. In a multi-zone reactor, the control variable in each zone, the temperature of reactants or that of heating medium, is constrained to a



special form - function of time only. Thus, there is a single uniform reactant temperature (or heating medium temperature) at any time in each control zone. This form of control can be approximated by condensing steam vapor outside the tube, by heating in a furnace, or by very high flow rate of heating medium. The multi-zone control is a realizable control action, and the investigation of this form of control will be useful for possible future application to industrial processes.

#### B. An Unsteady-State Tubular Reactor with a Single Reaction $A \rightleftharpoons B$

The condition which must be satisfied by the optimum temperature policy of this system is obtained and is compared with the analogous condition for a steady-state reactor, which has been obtained by Aris (3).

Also, a constrained optimum temperature policy for the system is obtained. The temperature policy takes the form that at any instant, there is a single uniform reactor temperature.

The dynamics of this system is expressed by the single equation

$$\frac{\partial x}{\partial t} = -v \frac{\partial x}{\partial y} + r(x, T_r) \quad (\text{IVB. 1})$$

where

$$r(x, T_r) = k_1(a-x) - k_2x \quad (\text{IVB. 2})$$

$x$  is the concentration of B;  $a-x$ , the concentration of A; axial velocity  $v$  is assumed constant. The rate constants  $k_1$  and  $k_2$  are related to reactant temperature by the expression:

$$k_i = k_{i0} \exp(-E_i/RT_r) \quad (\text{IVB. 3})$$

where  $k_{i0}$  and  $E_i$  are constants;  $R$  is universal gas constant;  $T_r$ , reactant temperature.

The reactant temperature is considered a control variable and has upper and lower limits.

$$T_a \leq T_r \leq T_b \quad (\text{IVB. 4})$$

The initial and boundary conditions for the system are given by

$$x(0, y) = x_0(y) \quad (\text{IVB. 5})$$

$$x(t, 0) = x_1(t) \quad (\text{IVB. 6})$$

The so-called 'best' is achieved by the maximization of the output of the reactor, or the minimization of the functional  $I$ .

$$I = - \int_0^T vx(t, y) dt \quad (\text{IVB. 7})$$

We shall introduce the Hamiltonian  $H$

$$H = p(-v \frac{\partial x}{\partial y} + k_1(a-x) - k_2x) \quad (\text{IVB. 8})$$

where  $p$  is defined by

$$\frac{\partial p}{\partial t} = -v \frac{\partial p}{\partial y} + (k_1 + k_2)p \quad (\text{IVB. 9})$$

$$p(T, y) = 0 \quad (\text{IVB. 10})$$

$$p(t, Y) = 1 \quad (\text{IVB. 11})$$

The solutions of the equations (IVB. 9) through (IVB. 11) are

$$p(t, y) = 0 \quad \text{for } y \leq Y - v(T - t) \quad (\text{IVB. 12})$$

$$p(t, y) = \exp \left[ - \int_t^{t+(Y-y)/v} (k_1 + k_2) \, ds \right] \\ \text{for } y > Y - v(T - t) \quad (\text{IVB. 13})$$

For a general expression  $r(x, T_r)$ ,  $p(t, y)$  is given by

$$p(t, y) = 0 \quad \text{for } y \leq Y - v(T - t) \quad (\text{IVB. 14})$$

$$p(t, y) = \exp \left( - \int_t^{t+(Y-y)/v} \frac{\partial r}{\partial x} \, ds \right) \\ \text{for } y > Y - v(T - t) \quad (\text{IVB. 15})$$

There are two problems to be considered:

(a). Find the optimal temperature  $T_r$  as a function of  $t$  and  $y$ .

In order to minimize  $I$ ,  $H$  is maximized with respect to  $T_r$ .

Since  $p$  is non-negative at all points of coordinate

(t,y), we can write

$$\max_{T_a \leq T_r \leq T_b} H = -pv \frac{\partial x}{\partial y} + p \left[ \max_{T_a \leq T_r \leq T_b} r(x, T_r) \right] \quad (\text{IVB. 16})$$

The equation (IVB. 16) states that the optimum temperature  $T_r$  has to be so chosen that it maximizes the local reaction rate at any instant. This condition for an unsteady-state reactor is the same as that obtained by Aris (3) for the optimum temperature policy of a steady-state tubular reactor with single reaction. This is an expected result for the equation (IVB. 16) is still applicable to a steady-state case, in which the functions  $x(t,y)$  and  $p(t,y)$  are reduced to time independent form  $x_s(y)$  and  $p_s(y)$ .

(b). Find the optimum temperature  $T_r$  as a function of  $t$  only.

In order to minimize the performance index  $I$ , the Hamiltonian  $H^t$ , which is defined below has to attain an absolute maximum with respect to  $T_r$ .

$$H^t = vx(t,Y) + \int_0^Y p \left[ -v \frac{\partial x}{\partial y} + r(x, T_r) \right] dy \quad (\text{IVB. 17})$$

$$\max_{T_a \leq T_r \leq T_b} H^t = \text{constant} + \max_{T_a \leq T_r \leq T_b} \int_0^Y pr(x, T_r) dy \quad (\text{IVB. 18})$$

The equation (IVB. 18) shows that the temperature  $T_r$  has to be chosen to maximize the integral. The integrand is formed by the product of the auxiliary variable  $p$  and the rate

of formation of  $x$  at any time  $t$  and at point  $y$ . The variable  $p$  serves as a weighting function when summing all the rates at time  $t$ . Since  $p$  is not a constant function,  $T_r$  is not necessarily chosen to maximize the total reaction rate at any instant. This is different from the result in problem (a).

For the linear system with  $r$  given by the equation (IVB. 2), we shall assume that  $T_r$  does not vary too much from time to time.

Let  $\overline{k_1}$  and  $\overline{k_2}$  be the average values of  $k_1$  and  $k_2$  respectively, we have

$$p(t,y) = 0 \quad \text{for } y \leq Y-v(T-t) \quad (\text{IVB. 19})$$

$$p(t,y) = \exp [-(\overline{k_1} + \overline{k_2})(Y-y)/v] \\ \text{for } y > Y-v(T-t) \quad (\text{IVB. 20})$$

The equation (IVB. 20) shows that for  $y > Y-v(T-t)$ ,  $p(t,y)$  increases monotonically from  $\exp[-(\overline{k_1} + \overline{k_2})Y/v]$  to 1 as the distance  $y$  increases from 0 to  $Y$ . This implies that in the process of deciding the best  $T_r$ , the reaction rate of a stream element is weighted heavier if element's position is nearer to the outlet.

The equation (IVB. 19) shows that the stream elements which will never show up at the outlet before time  $T$  are completely ignored in the process of deciding the optimal tempera-

ture  $T_r$ . This is an expected result.

For the linear system, the optimal control is given by:

(b1).  $c_2 \leq 1$

$$T_r = T_b \quad \text{if} \quad \int_0^Y p(a-x-c_1 c_2 k_1^{c_2-1} x) dy > 0 \quad (\text{IVB. 21})$$

$$T_r = T_a \quad \text{if} \quad \int_0^Y p(a-x-c_1 c_2 k_1^{c_2-1} x) dy < 0 \quad (\text{IVB. 22})$$

(b2).  $c_2 > 1$

$$T_r = T_r^O \quad \text{if} \quad T_a < T_r^O < T_b \quad (\text{IVB. 23})$$

$$T_r = T_b \quad \text{if} \quad T_r^O > T_b \quad (\text{IVB. 24})$$

$$T_r = T_a \quad \text{if} \quad T_r^O < T_a \quad (\text{IVB. 25})$$

where  $c_1$ ,  $c_2$  and  $T_r^O$  are defined as:

$$c_1 = k_{20} (k_{10})^{-E_2/E_1} \quad (\text{IVB. 26})$$

$$c_2 = E_2/E_1 \quad (\text{IVB. 27})$$

$$T_r^O = \frac{-E_1}{\text{Rln} \left\{ \frac{\int_0^Y p(a-x) dy}{c_1 c_2 \int_0^Y p x dy} \cdot \frac{1}{k_{10}^{c_2-1}} \right\}} \quad (\text{IVB. 28})$$

In actual calculation to obtain numerical values of optimal temperature  $T_r$ , the variables  $x$  and  $p$  are implicit functions of optimal temperature policy. Therefore, in order to obtain an optimal temperature policy, an efficient iterative search method such as one which will be explained in the section IVF has to be utilized.

### C. Flow Rate Control of a Heat Exchanger

Flow rate control of a heat exchanger with constant wall temperature is studied in this section. A constant wall temperature might be approximated by condensing steam vapor or by extremely rapid flow of heating medium. The flow rate of process fluid is manipulated in a best fashion in the process of shifting the fluid's outlet temperature to a new level. The optimal control will be found to be a bang-bang control. Koppel (21) has tried to obtain the same result utilizing a sub-optimal control shown in Figure (2b).

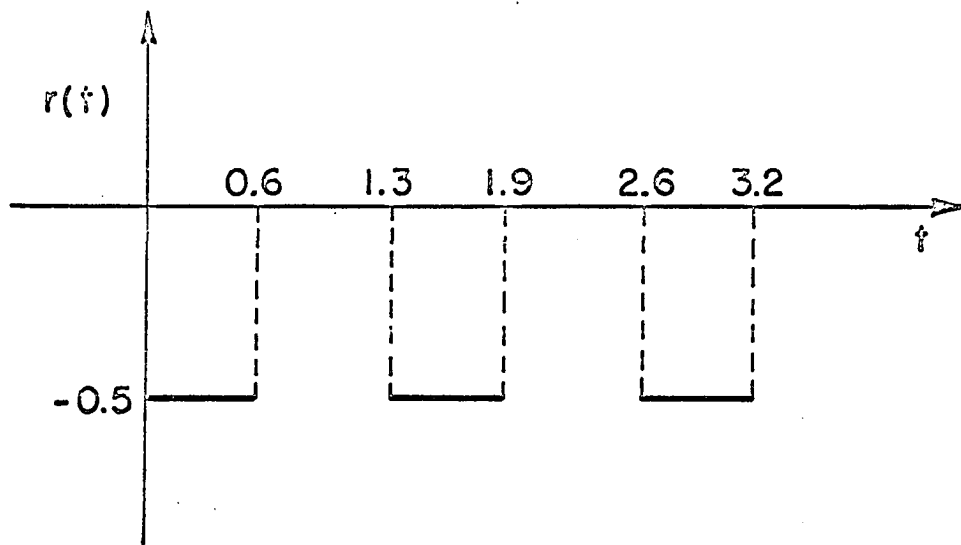
The process dynamics of the system can be represented by the equation

$$\frac{\partial \theta}{\partial t} + [1+r(t)] \frac{\partial \theta}{\partial y} = -c[1+br(t)]\theta \quad (\text{IVC. 1})$$

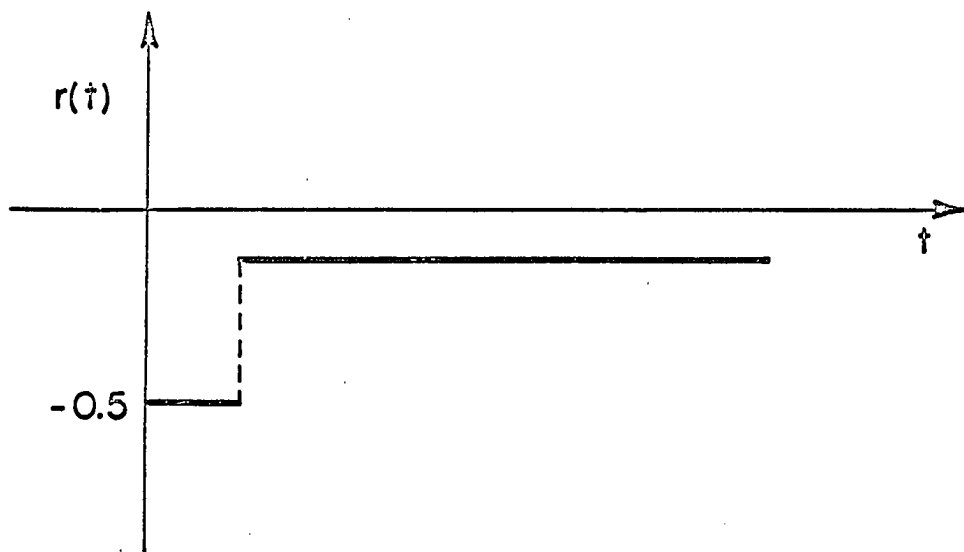
with boundary condition

$$\theta(t,0) = 1 \quad (\text{IVC. 2})$$

and initial condition



(2a). Optimal control



(2b). Sub-optimal control suggested by Koppel

Figure 2. Optimal and sub-optimal controls of heat exchanger



$$\theta(0,y) = e^{-cy} \quad (\text{IVC. 3})$$

where  $\theta$  is the normalized temperature of the fluid;  $b$ ,  $c$ , constants;  $r$ , the manipulatable portion of the fluid velocity  $[1+r]$ .

The transformation of variable by the definition given by the following equation suggested by Koppel (22):

$$x(t,y) = \frac{1}{c} \int_{\theta(t,y)}^1 \frac{ds}{s} - y \quad (\text{IVC. 4})$$

reduces the equations (IVC. 1) through (IVC. 3) to

$$\frac{\partial x}{\partial t} + [1+r(t)] \frac{\partial x}{\partial y} = -(1-b)r(t) \quad (\text{IVC. 5})$$

$$x(t,0) = 0 \quad (\text{IVC. 6})$$

$$x(0,y) = 0 \quad (\text{IVC. 7})$$

For the present problem the value of  $b$  is taken as 0.2 and the control  $r$  is bounded by

$$-0.5 \leq r \leq 0.5 \quad (\text{IVC. 8})$$

We would like to manipulate  $r(t)$  so that the variable  $x(t,1)$  at the outlet will be as near to 0.24 as possible, the performance criterion to be minimized takes the form:

$$I = \int_0^{\infty} [0.24 - x(t,1)]^2 dt \quad (\text{IVC. 9})$$

where 1 is the space coordinate of heat exchanger's outlet.

By solving equations (IVC. 5) through (IVC. 7), the state of the system is found to be:

$$x(t,y) = -(1-b) \int_0^t r(s) ds \quad \text{for } t + \int_0^t r(s) ds < y$$

(IVC. 10)

$$x(t,y) = - (1-b) \int_{t_0(y')}^t r(s) ds$$

$$\text{for } t + \int_0^t r(s) ds \geq y \quad \text{(IVC. 11)}$$

where

$$y' = y - t - \int_0^t r(s) ds \quad \text{(IVC. 12)}$$

and  $t_0(y')$  satisfies the relation

$$t_0(y') = -y' - \int_0^{t_0(y')} r(s) ds \quad \text{(IVC. 13)}$$

The Hamiltonian  $H^t$  for the system is formed.

$$H^t = -[0.24 - x(t,1)]^2 - \int_0^1 p[(1+r) \frac{\partial x}{\partial y} + (1-b)r] dy$$

(IVC. 14)

The auxiliary variable  $p$  in the above equation is defined by:

$$\frac{\partial p}{\partial t} + [1+r(t)] \frac{\partial p}{\partial y} = 0 \quad \text{(IVC. 15)}$$

$$p(t,1) = \frac{2[0.24-x(t,1)]}{1+r(t)} \quad (\text{IVC. 16})$$

$$p(\infty, y) = 0 \quad (\text{IVC. 17})$$

The optimal control  $r(t)$  has to maximize the Hamiltonian  $H^t$ . Therefore, the control  $r(t)$  has to assume the form

$$r(t) = -0.5 \quad \text{if} \quad \int_0^1 p\left(\frac{\partial x}{\partial y} + 1 - b\right) dy > 0 \quad (\text{IVC. 18})$$

$$r(t) = 0.5 \quad \text{if} \quad \int_0^1 p\left(\frac{\partial x}{\partial y} + 1 - b\right) dy < 0 \quad (\text{IVC. 19})$$

Since the performance index is given by (IVC. 9), the value of  $x(t,1)$  will be equal to 0.24 after it reaches this value providing this condition can be satisfied by a certain operation. This operation is possible for the present problem. If we define  $T_1$  as the time when the value of  $x(t,1)$  reaches value 0.24, then  $p(t,1)$  is zero after time  $T_1$ . Therefore, the auxiliary variable  $p(t,y)$  is identically zero after time  $T_1$  and  $p(t,y)$  is either positive or zero before time  $T_1$ .

At time zero,  $x(t,y)$  is identically zero and  $p(t,y)$  is non-negative. Therefore, the integral criterion in (IVC. 18) is larger than zero and the control variable  $r$  takes on the value -0.5. The control variable takes on this value until the integral becomes zero.

The control corresponding to zero value of the integral,

namely,

$$\int_0^1 p \left( \frac{\partial x}{\partial y} + 1 - b \right) dy = 0 \quad (\text{IVC. 20})$$

is called singular control and cannot be obtained by the application of the optimum theorems developed in the present work. Due to the form of initial and boundary conditions chosen for the heat exchanger problem, the value of  $x(t,1)$  can be maintained at the magnitude 0.24 by a suitable manipulation of the flow rate after time  $T_1$ .

$$x(t,1) = 0.24 \quad (\text{IVC. 21})$$

The condition (IVC. 21) makes the auxiliary variable  $p$  identically zero after time  $T_1$  and the condition (IVC. 20) is met, so that the corresponding control is a singular control. For the present problem, the optimum control up to time 0.6 is obtained by the application of the optimum theorems given in the present work. The optimum control after time 0.6 is obtained by fulfilling condition (IVC. 21) through an adequate choice of the function  $r(t)$ .

The optimal control for the heat exchanger system is a bang-bang control as is shown in Figure (2a). The sub-optimal control in the form suggested by Koppel (21) is shown in Figure (2b). The optimal state variable profiles at different instants of time are shown in Figure 3. The optimal control is a bang-bang control because of the special form of initial

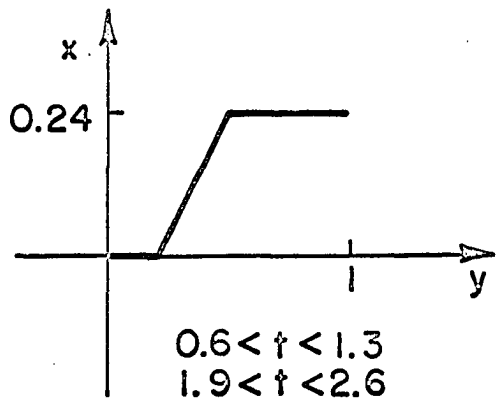
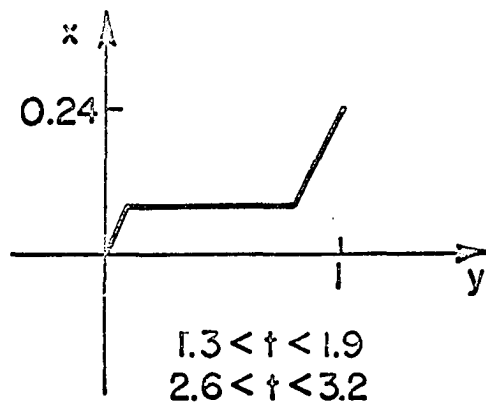
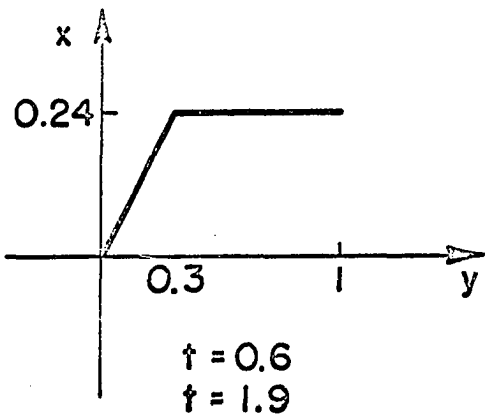
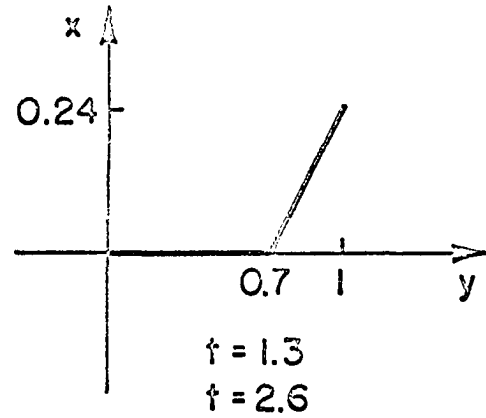
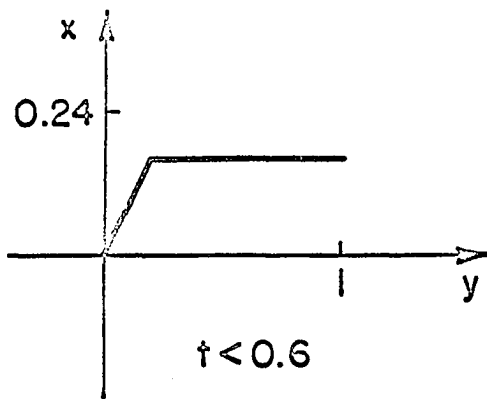


Figure 3. Optimal state variable profiles of heat exchanger

and boundary conditions in addition to the linearity of the system. It may assume any other modes of control if the initial and boundary conditions are of different form in spite of the linearity of the system. On examination of the proposed control strategy, it is obvious that we found the unique optimum control for this problem.

#### D. Linearization of a General Tubular Reactor System

Although a tubular reactor system never operates under a steady-state condition, the deviation of the system's state from steady-state is not very large for usual operation. Thus, for a regulatory control process, it is reasonable to linearize the state of the system with respect to a steady-state condition and obtain the necessary regulatory action by analysis of this simplified model. Analysis is simplified due to consideration of a simpler model than the original one.

A tubular reactor system with a general rate expression will be considered. The reactant flow rate  $u_1$  and coolant flow rate  $u_2$  are considered as two elements of the control vector. The other elements of the control vector might be temperature, pressure or some other controllable parameters.

$$\frac{\partial x_i}{\partial t} = - u_j(t) \frac{\partial x_i}{\partial y} + r_i(x,u)$$

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

$$j=1 \quad \text{for } i=1,2,\dots,n-1$$

$$j=2 \quad \text{for } i=n \quad (\text{IVD. 1})$$

the initial conditions and boundary conditions for the state vector are:

$$x_i(0,y) = x_{i0}(y) \quad i=1,2,\dots,n \quad (\text{IVD. 2})$$

$$x_i(t,0) = x_{i1}(t) \quad i=1,2,\dots,n \quad (\text{IVD. 3})$$

In the condition of steady-state, the values of the state vector  $x_s$  and control vector  $u_s$  can be related by the equations

$$0 = -u_{sj} \frac{dx_{si}}{dy} + r_i(x_s, u_s)$$

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

$$j=1 \quad \text{for } i=1,2,\dots,n-1$$

$$j=2 \quad \text{for } i=n \quad (\text{IVD. 4})$$

and initial conditions

$$x_{si}(0) = d_{si} \quad i=1,2,\dots,n \quad (\text{IVD. 5})$$

where  $d_{si}$ 's are constants. The variables  $x_s$  and  $u_s$  are functions of  $y$  only.

Let

$$x_{Di}(t, y) = x_i(t, y) - x_{si}(y)$$

$$i=1, 2, \dots, n \quad (\text{IVD. 6})$$

$$u_{Dj}(t, y) = u_j(t, y) - u_{sj}(y)$$

$$j=1, 2, \dots, r \quad (\text{IVD. 7})$$

If the values of  $x_{Di}$ 's and  $u_{Dj}$ 's are assumed to be small, then the system (IVD. 1) can be approximated by a linear system of  $x_{Di}$ 's,  $u_{Dj}$ 's with reference to time independent  $x_{si}$ 's,  $u_{sj}$ 's.

The  $u_{Dj}$ 's have lower and upper bounds due to constraints imposed on  $u$  and due to the range of applicability of the linearized system.

$$u_{Dja} \leq u_{Dj} \leq u_{Djb} \quad j=1, 2, \dots, r \quad (\text{IVD. 8})$$

$$\frac{\partial x_{Di}}{\partial t} = -a_j \frac{\partial x_{Di}}{\partial y} + \sum_{k=1}^n b_{ik}(y) x_{Dk} + \sum_{k=1}^r c_{ik}(y) u_{Dk}$$

$$i=1, 2, \dots, n \quad (\text{IVD. 9})$$

where

$$a_j = u_{sj} \quad j=1, 2 \quad (\text{IVD. 10})$$



$$b_{ik}(y) = \left. \frac{\partial r_i}{\partial x_k} \right|_{(x_s, u_s)}$$

$$i=1,2,\dots,n; k=1,2,\dots,n \quad (\text{IVD. 11})$$

$$c_{ik}(y) = \left. \frac{\partial r_i}{\partial u_k} \right|_{(x_s, u_s)} - \frac{r_i(x_s, u_s)}{u_{sj}} \delta_{kj}$$

$$i=1,2,\dots,n; k=1,2,\dots,r \quad (\text{IVD. 12})$$

In the equations (IVD. 9) and (IVD. 12),  $j$  is designated by:

$$j=1, \quad \text{if } i=1,2,\dots,n-1$$

$$j=2, \quad \text{if } i=n$$

$\delta_{kj}$  is the Kronecker delta; the coefficients  $a_j$ 's are constant; other coefficients are functions of  $y$  only.

If we desire to minimize the deviation of the system from the steady-state condition, the performance index to be minimized can be expressed as:

$$\begin{aligned} I = & -w_0 \int_0^T g_0(t, x_D(t, 0), x_D(t, Y), u_D^t(t)) dt \\ & -w'_0 \int_0^Y h_0(y, x_D(0, y), x_D(T, y)) dy \\ & -p_0 \int_0^T \int_0^Y f_0(t, y, x_D, u_D) dy dt \end{aligned} \quad (\text{IVD. 13})$$

where  $w_0$ ,  $w'_0$  and  $p_0$  are constants with value -1.

The Hamiltonians  $H$  and  $H^t$  for this system are

$$H = + p_o f_o + \sum_{i=1}^n p_i (-a_j \frac{\partial x_{Di}}{\partial y} + \sum_{k=1}^n b_{ik} x_{Dk} + \sum_{k=1}^r c_{ik} u_{Dk}) \quad (\text{IVD. 14})$$

$$H^t = - g_o + \int_0^Y H dy \quad (\text{IVD. 15})$$

The auxiliary variables  $p_i (i=1,2,\dots,n)$  are defined by:

$$\frac{\partial p_i}{\partial t} = - a_j \frac{\partial p_i}{\partial y} - \sum_{k=1}^n p_k b_{ki} \quad i=1,2,\dots,n \quad (\text{IVD. 16})$$

$$p_i(T,Y) = \frac{w_o' \partial h_o}{\partial x_{Di}(T,Y)} \quad i=1,2,\dots,n \quad (\text{IVD. 17})$$

$$p_i(t,Y) = \frac{1}{a_j} \frac{w_o \partial g_o}{\partial x_{Di}(t,Y)} \quad i=1,2,\dots,n \quad (\text{IVD. 18})$$

The control vector  $u_D = (u_D^t, u_D'')$  has  $m$  elements  $u_{Dj}^t$  and  $r-m$  elements  $u_{Dk}''$ .  $u_D^t$  is the collection of  $m$  elements  $u_{Dj}^t$ , which are function of time only.  $u_D''$  is the collection of  $r-m$  elements  $u_{Dk}''$ , which are functions of both time and distance.

$H$  has to assume an absolute maximum value with respect to

$u_{Dj}''$  and so does  $H^t$  with respect to  $u_{Dk}^t$ . If  $f_0$  and  $g_0$  are also linear functions of  $u_D$  and  $u_D^t$  respectively, then the optimal control is a bang-bang control.

Assume  $f_0$  and  $g_0$  have the following forms

$$f_0 = \sum_{k=1}^n x_{Dk}^2 + \sum_{k=1}^r c_{ok}(t, y) u_{Dk} \quad (\text{IVD. 19})$$

$$g_0 = \sum_{k=1}^n b_k'(t) x_{Dk}^2(t, Y) + \sum_{k=1}^m d_{ok}(t) u_{Dk}^t \quad (\text{IVD. 20})$$

Then, the optimal control  $u_D$  is

$$u_{Dk}^t = u_{Dka}^t$$

if

$$-d_{ok} + \int_0^Y \sum_{i=0}^n p_i c_{ik} dy < 0$$

$$k=1, 2, \dots, m \quad (\text{IVD. 21})$$

$$u_{Dk}'' = u_{Dka}''$$

if

$$\sum_{i=0}^n p_i c_{ik} < 0 \quad k=m+1, m+2, \dots, r \quad (\text{IVD. 22})$$

$$u_{Dk}^t = u_{Dkb}^t$$

if

$$-d_{ok} + \int_0^Y \sum_{i=0}^n p_i c_{ik} dy > 0$$

$$k=1, 2, \dots, m \quad (\text{IVD. 23})$$

$$u''_{Dk} = u''_{Dkb}$$

if

$$\sum_{i=0}^n p_i c_{ik} > 0 \quad k=m+1, m+2, \dots, r \quad (\text{IVD. 24})$$

The optimal control for other performance criteria can be obtained similarly.

### E. Numerical Studies

The most flexible control in a tubular reactor system is an unconstrained function of time and space coordinates. For example, if the temperature of the heating medium is considered as the control variable of a system, in order to achieve the best result at any time, we would like to assign an optimum temperature profile of heating medium along the reactor. The profile changes its shape as time passes. Since, in practice, it is impossible to carry out this kind of control, we shall consider a control constrained to a special form. Consider a one-zone reactor in which the zone temperature may change from time to time, but at any time there is only one temperature uniform throughout the reactor. This form of control might be approximated by condensing steam under time-varying pressure. If the reactor is split into two zones and if the temperature at each zone may change with time but is uniform throughout the zone, the result is a realizable two-zone control reactor. A three-zone control reactor is defined similarly.

In the numerical studies, the performance of an ideal reactor and one-, two-, three-zone reactors are compared. The ideal reactor gives a scale for the utmost achievement. The performance of multi-zone reactors are compared with each other and with the ideal reactor. The one-zone control is the simplest control among all the cases and gives poorest performance. The two-zone and three-zone controls are more complex than the one-zone control and give better or equal performances.

In general, the three-zone control is better than two-zone control, and two-zone control is again better than one-zone control. However, the careful evaluation of separate processes is necessary, as we can find an example where two-zone control is better than three-zone control. Moreover, this statement is correct only if the same cost of control is assessed to the various cases. If the added complexity of control increases the cost of operation, the better yield obtained by more flexible control is not necessarily worthwhile. The choice of control then should depend on both the yield of the chemical reaction and the cost of control action.

The performance of all the cases studied numerically are judged by the average yield of product  $p$  at the end of a tubular reactor over the periods of time from zero to  $T$ . Therefore, the performance index to be minimized can be expressed as

$$I = - \frac{1}{T} \int_0^T x_p(t, Y) dt$$

where  $x_p$  is the concentration of a product;  $Y$ , the space coordinate of the outlet of the reactor. The adoption of this performance index implies a negligible variation of cost of operation for different controls. In all the numerical examples, the terminal time  $T$  for the optimum operation is taken as one residence time of the reactor. The details of various control reactors and their control operation are given below.

The reactors under consideration have length  $Y$  and among them, the multi-zone reactors have control zones of equal length. The controls for the reactors have to take a value from a control region bounded by an upper bound and a lower bound. For an ideal reactor, any piecewise continuous function of time and space coordinate with its magnitude between these bounds can be applied. However, for a multi-zone control, the control in each zone is restricted to a special form - a piecewise constant function of time. The total time  $T$  is subdivided into twenty-four periods of equal length, and the control variable in each zone assumes twenty-four constant values over twenty-four periods of time. Theorem 1 and Theorem 5 of Chapter III are utilized in seeking for optimal control for ideal reactor and multi-zone reactors.

The procedure of obtaining optimum control is given in section F. The computation was by IBM System/360, and the

computer flow diagram is given in section G. The results of calculations are described and discussed in section H.

#### F. Procedure of Calculation

The system under consideration is described by a set of linear first order partial differential equations

$$\frac{\partial x_i}{\partial t} = -v_i(t) \frac{\partial x_i}{\partial y} + r_i(x, u)$$

$$i=1, 2, \dots, n \quad (\text{IVF. 1})$$

with initial and boundary conditions

$$x_i(0, y) = a_{i0}(y) \quad i=1, 2, \dots, n \quad (\text{IVF. 2})$$

$$x_i(t, 0) = a_{i1}(t) \quad i=1, 2, \dots, n \quad (\text{IVF. 3})$$

The independent variables will be transformed by the formulas

$$y = y'_i + \int_0^s v_i(t) dt$$

$$i=1, 2, \dots, n \quad (\text{IVF. 4})$$

$$t = s \quad (\text{IVF. 5})$$

After the transformation of the independent variables, equations (IVF. 1) reduce to

$$\frac{\partial x_i(s, y'_i)}{\partial s} = r_i(x, u)$$

$$i=1, 2, \dots, n \quad (\text{IVF. 6})$$

By the same transformation of the independent variables, the equations for the auxiliary variables (IIIF. 10) become

$$\frac{\partial p_i(s, y_i^!)}{\partial s} = - \frac{\partial H(s, y_i^!, x, x_y, p, u)}{\partial x_i}$$

$i=1, 2, \dots, n$  (IVF. 7)

Hence, following the path of constant  $y_i^!$ , a Runge-Kutta-Gill system abstracted in Appendix E can be utilized for the numerical evaluation of  $x_i$  and  $p_i$  at different value of  $s$ .

Since the optimal control is the desired solution, it is not known beforehand. Therefore, a reasonable but arbitrary guess of  $u$  is used for the calculation of  $x_i$  and  $p_i$ . The  $x_i$  are obtained from forward integration.  $I$  is then calculated.

After all the required values of  $x_i(t, y)$  are obtained,  $p_i(t, y)$  are obtained by backward integration, using the known values of  $p_i(T, y)$ ,  $p_i(t, Y)$ , control  $u$  and state variable  $x_i(t, y)$ .

For an ideal reactor, we have the following equations:

$$H = \sum_{k=1}^n p_k f_k \quad (\text{IVF. 8})$$

$$I = \int_0^T g(t, x(t, Y)) dt \quad (\text{IVF. 9})$$

$$\Delta I = - \int_0^T \int_0^Y \frac{\partial H}{\partial u} \Delta u dy dt + o(\Delta u^2) \quad (\text{IVF. 10})$$

where  $o(\Delta u^2)$  represents the remainder term, which has order of



smallness  $\Delta u^2$ .

For an N-zone reactor, we have assigned piecewise constant functions of time to control variables, and an increment  $\Delta I$  corresponding to a change of control can be expressed as a sum of the terms consisting of product of  $\Delta u_{ij}$  and partial derivatives of  $H_{ij}^{t*}$  with respect to  $u_{ij}$ .

$$\Delta I = - \sum_{i=1}^N \sum_{j=1}^{24} \frac{\partial H_{ij}^{t*}}{\partial u_{ij}} \Delta u_{ij} + O(\Delta u_{ij}^2) \quad (\text{IVF. 11})$$

$$H_{ij}^{t*} = \int_{T_{j-1}}^{T_j} \int_{(i-1)Y/N}^{iY/N} H \, dy \, dt$$

$$i=1 \text{ to } N; \quad j=1, 2, \dots, 24 \quad (\text{IVF. 12})$$

where  $\Delta u_{ij}$  is the variation of the control  $u_i$  of the  $i$ th zone in the  $j$ th period of time, from  $T_{j-1}$  to  $T_j$ .

For an ideal reactor,  $\frac{\partial H}{\partial u}$  is then evaluated and the change of control variable  $\Delta u$  takes a value proportional to  $\frac{\partial H}{\partial u}$ .

$$\Delta u = c^* \frac{\partial H}{\partial u} \quad (\text{IVF. 13})$$

where  $c^*$  is a suitable positive number.

Then, for this variation  $\Delta u$ ,  $\Delta I$  becomes

$$\Delta I = - \int_0^T \int_0^Y c^* \left( \frac{\partial H}{\partial u} \right)^2 dy \, dt + O(\Delta u^2) \quad (\text{IVF. 14})$$

As increment  $\Delta I$  is negative for the variation  $\Delta u$ , the value of  $I$  decreases until it reaches a possible local minimum

or an absolute minimum.

For an N-zone reactor,  $\frac{\partial H_{ij}^{t*}}{\partial u_{ij}}$  are evaluated instead, and the changes of control  $\Delta u_{ij}$  take values proportional to  $\frac{\partial H_{ij}^{t*}}{\partial u_{ij}}$ .

$$\Delta u_{ij} = c^{**} \frac{\partial H_{ij}^{t*}}{\partial u_{ij}} \quad i=1 \text{ to } N; \quad j=1,2,\dots,24 \quad (\text{IVF. 15})$$

where  $c^{**}$  is a suitable positive number.

The increment  $\Delta I$  for this variation of control is

$$\Delta I = - \sum_{i=1}^N \sum_{j=1}^{24} c^{**} \left( \frac{\partial H_{ij}^{t*}}{\partial u_{ij}} \right)^2 + O(\Delta u_{ij}^2) \quad (\text{IVF. 16})$$

The value of  $I$  again decreases until it reaches a local minimum or an absolute minimum.

The control vector is adjusted repeatedly. Each time the new values of  $x_i$ ,  $p_i$ ,  $I$ ,  $\frac{\partial H}{\partial u}$  or  $\frac{\partial H_{ij}^{t*}}{\partial u_{ij}}$  are evaluated. This process repeats until the increment of  $I$  is less than a certain small number.

#### G. Computer Flow Diagram

The computer programs were written in Basic Programming Support Fortran IV Language and were run on the IBM System/360 computer. The value of  $I$  approaches a local minimum or an absolute minimum by computation. Each program was divided into one main program and several subprograms. The three sub-

programs calculated variables  $x$ ,  $p$  and improved control  $u^*$ . The main program supplied the logic of sequential calculation. Figure 4 shows a flow diagram of the computer program. The details on calculating  $x$ ,  $p$  and improved control  $u^*$  were given in previous section. The method of obtaining optimal control consisted of step by step improvements on control  $u$  (or  $u_{ij}$ ) with the step change  $\Delta u$  (or  $\Delta u_{ij}$ ) proportional to magnitude of partial derivative  $\frac{\partial H}{\partial u}$  (or  $\frac{\partial H_{ij}^{t*}}{\partial u_{ij}}$ ). In other words, a weak form of the maximum theorems derived in the present work was utilized. The weak form was used because the time duration for step change  $\Delta u$  was not infinitesimally small in the numerical calculations.

#### H. Result and Discussion

The optimum start-up operations of various control reactors with three different chemical processes are studied numerically. The average yield of a product over one residence time is the quantity to be maximized. This amounts to the minimization of the performance index  $I$ .

$$I = \frac{-1}{T} \int_0^T x_p(t, Y) dt$$

where  $T$  is one residence time;  $Y$ , space coordinate of the outlet of reactor.

The adoption of this index gives an uniform measure of

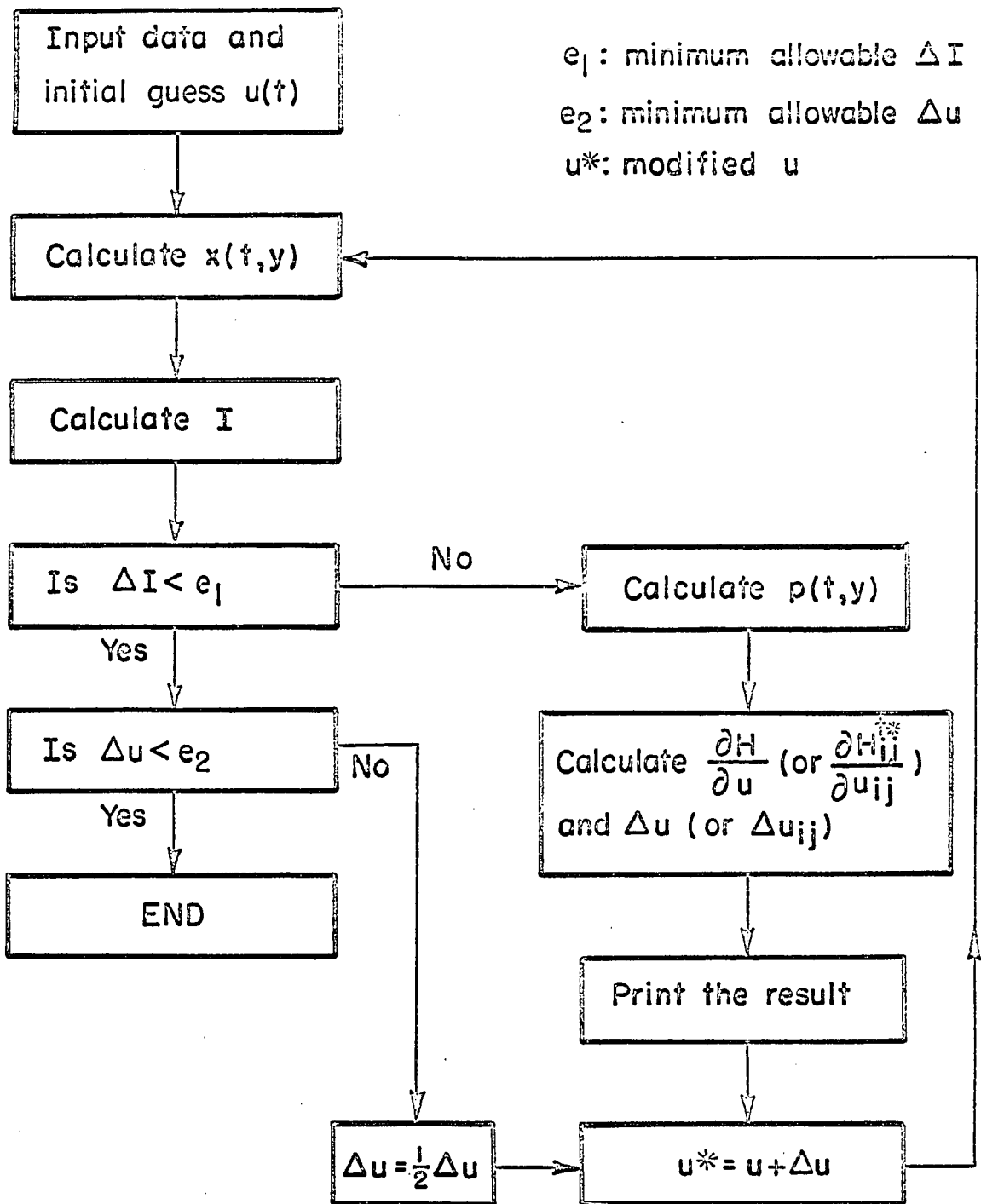


Figure 4. Computer flow diagram

achievement, which depends only on the physical data of the chemical processes. In a practical plant optimization, the cost of control operation and control equipment should also be included. A more complicated operation indicates a higher cost. Even in the design stage, higher computation cost is observed for more complicated zoned control as is shown in Figure 5. But the inclusion of control cost etc. complicates the problem. Furthermore, despite exclusion of control cost, the performance index  $I$  gives a reliable measure of achievement as long as the control cost is relatively low.

We have arbitrarily chosen one residence time as the duration of time for optimization. There is some question as to the best choice of the optimization time period  $T$ . Clearly a time  $T$  very much smaller than one residence time would not lead to very meaningful answers. On the other hand, a very long period  $T$  increases the computation cost too much. So we have chosen  $T$  equal to one residence time as an intuitive compromise between the requirements of shorter times for lower computation costs and longer times for more meaningful results.

We have chosen the piecewise constant function of time as the desired mode of control in multi-zone reactors. It would be too optimistic to ask a machine to operate precisely on a continuously changing optimal control pattern, not to mention the cost of calculation and operation. The practical

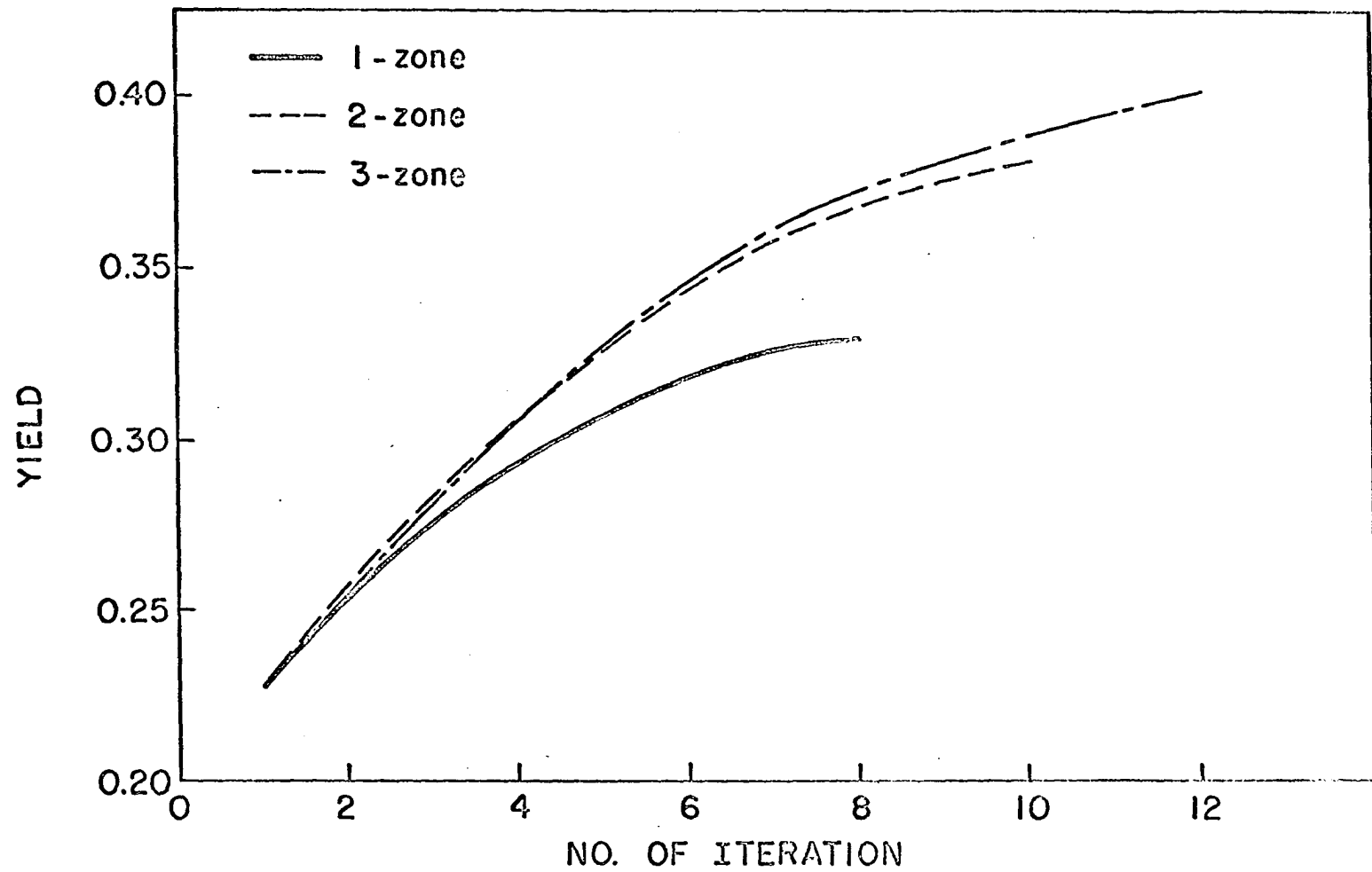


Figure 5. Yield vs. number of iteration

operation may not require such a precise operation either, since unknown factors of the processes result in a set of physical data accurate only up to a certain degree.

At any time, there are numerous fluid elements occupying different positions in a tubular reactor. All the elements have different residence time in the reactor and may have different chemical compositions. Therefore, different elements need different temperature policies to attain the maximum yield. If every fluid element in the reactor has to be subjected to the same temperature policy, and if the maximum yield is sought, there is a necessity of compromise among different elements in deciding the common temperature policy.

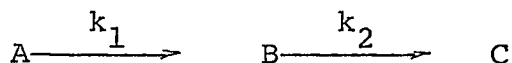
In an one-zone reactor, the optimum temperature at any time  $t$  is the result of compromise among all the fluid elements in the reactor at that time. Likewise, the optimum temperatures in the first zone and the second zone in a two-zone reactor are the results of compromise among the fluid elements in the first zone and in the second zone respectively. Comparing with a one-zone reactor, in the two-zone reactor we need only consider one half as many fluid elements in the process of obtaining an optimum temperature. As a result, the temperature policy obtained in the two-zone reactor satisfies the elements' need better than that in the one-zone reactor. Therefore, we expect a higher yield from two-zone reactor than from one-zone reactor. By the same reasoning,

we expect a higher yield from three-zone reactor than from one-zone reactor.

The ideal reactor has the highest possible yield since every fluid element in the reactor can have its own temperature policy.

The numerical differences in performance for different form of control depend on the specific chemical process. We consider the first order consecutive reaction A to B to C; ethylene oxide production - a process with two competitive reactions; and formaldehyde production. The details on the results for various processes are given in the following.

- (a). The optimum start-up operations of tubular reactor with the first order consecutive reaction A to B to C.



The yields of substance B were maximized in start-up operations of tubular reactors. This reaction under steady-state operation was originally studied by Bilous and Amundson (23,24). They obtained an optimum temperature profile for a tubular reactor of a given length. Later, the same profile was again obtained by Aris (3), Lee (25) and Fan (26). Aris used dynamic programming to obtain the optimal solution. Lee and Fan applied Pontryagin's Maximum Principle. Coward and Jackson (27) used this reaction as an example to stress that the Maximum Principle is only a necessary condition of an



optimal operation.

In this study, the optimum unsteady state operation of a reactor was considered. The yield of substance B was maximized over a period of one residence time. The optimum reactant temperature policies for ideal reactor, and one-, two-, three-zone reactors were obtained.

The equations which represent the dynamics of the process are

$$\frac{\partial x_1}{\partial t} = -v \frac{\partial x_1}{\partial y} - k_1 x_1$$

$$\frac{\partial x_2}{\partial t} = -v \frac{\partial x_2}{\partial y} + k_1 x_1 - k_2 x_2$$

The initial and the boundary conditions for the variables are

$$x_1(0, y) = 0.95$$

$$x_1(t, 0) = 0.95$$

$$x_2(0, y) = 0.05$$

$$x_2(t, 0) = 0.05$$

where  $x_1$  is the concentration of component A;  $x_2$  is the concentration of component B; the axial velocity  $v$  is assumed constant; in numerical calculation  $k_1$  replaces reactant temperature as the control variable. The variable  $k_2$  is function of  $k_1$ .

$$k_2 = c_1 k_1^{c_2}$$

where  $c_1$  and  $c_2$  are positive constants related to  $a_{ij}$  in Table 1.

$$c_1 = \frac{a_{21}}{\frac{a_{22}}{a_{12}}}$$

$$c_2 = \frac{a_{22}}{a_{12}}$$

The functional to be minimized has a form

$$I = - \frac{1}{T} \int_0^T x_2(t, Y) dt$$

The Hamiltonian  $H_T$  for the system is formed.

$$H_T = p_1 \left( -v \frac{\partial x_1}{\partial y} - k_1 x_1 \right) + p_2 \left( -v \frac{\partial x_2}{\partial y} + k_1 x_1 - k_2 x_2 \right)$$

The auxiliary variables  $p_i$ 's are defined by the equations:

$$\frac{\partial p_1}{\partial t} = -v \frac{\partial p_1}{\partial y} + (p_1 - p_2) k_1$$

$$\frac{\partial p_2}{\partial t} = -v \frac{\partial p_2}{\partial y} + p_2 k_2$$

and the terminal as well as the boundary conditions:

$$p_i(T, Y) = 0 \quad i=1,2$$

$$p_1(t, Y) = 0$$

$$p_2(t, Y) = \frac{1}{vT}$$

For the purpose of optimization, we can consider only the terms which contain the control variable.  $H$  is the function which is the part of  $H_T$  containing the control variable.

$$H = -p_1 k_1 x_1 + p_2 (k_1 x_1 - k_2 x_2)$$

For an ideal tubular reactor  $H$  has to be maximized with respect to  $k_1$ . The optimal control yields absolute maximum of  $H$ .  $H$  might assume absolute maximum at a local maximum. In this case

$$\frac{\partial H}{\partial k_1} = 0 \quad \text{if } k_{1a} < k_1 < k_{1b}$$

where  $k_{1a}$  and  $k_{1b}$  are lower and upper bound of  $k_1$ .

$H$  might assume the absolute maximum at extreme points of control. At these points, the derivatives of  $H$  with respect to  $k_1$  have following properties:

$$\frac{\partial H}{\partial k_1} \geq 0 \quad \text{if } k_1 = k_{1b}$$

$$\frac{\partial H}{\partial k_1} \leq 0 \quad \text{if } k_1 = k_{1a}$$

For one-, two-, three-zone reactors,  $H_i^t$  has to be maxi-

mized with respect to  $k_{1i}(t)$ . The subscript  $i$  ranges from 1 to  $N$ .  $N$  is the total number of control zones in the reactor. The function  $k_{1i}(t)$  is the value of control  $k_1$  in the  $i$ th zone.

$$H_i^t = \int_{(i-1)Y/N}^{iY/N} [-p_1 k_{1i} x_1 + p_2 (k_{1i} x_1 - k_{2i} x_2)] dy$$

$$i=1 \text{ to } N$$

The optimal  $k_{1i}$  can either correspond to a local maximum of  $H_i^t$  or be at the extreme points. The derivative

$\frac{\partial H_i^t}{\partial k_{1i}}$  at the optimal point has properties the same as that of  $\frac{\partial H}{\partial k_1}$  . i.e.

$$\frac{\partial H_i^t}{\partial k_{1i}} = 0 \quad \text{if } k_{1a} < k_{1i} < k_{1b}$$

$$\frac{\partial H_i^t}{\partial k_{1i}} \geq 0 \quad \text{if } k_1 = k_{1b}$$

$$\frac{\partial H_i^t}{\partial k_{1i}} \leq 0 \quad \text{if } k_1 = k_{1a}$$

If the control variable is the temperature of the heating medium  $T_h$ , we need an additional equation to describe the dynamics of the system.

$$\frac{\partial x_3}{\partial t} = -v \frac{\partial x_3}{\partial y} + c_3 (T_h - x_3) + c_4 (-k_1 x_1 \Delta H_1 - k_2 x_2 \Delta H_2)$$

where  $x_3$  is the temperature of the reactants;  $\Delta H_j$  is the heat of  $j$ th reaction.

$$c_3 = \frac{2U_T}{R_t \rho C_p}$$

$$c_4 = \frac{1}{\rho C_p}$$

$$k_i = a_{i1} \exp (-a_{i2}/x_3)$$

where  $U_T$  is the overall heat transfer coefficient;  $R_t$ , diameter of the tube;  $\rho$ , average density of the reactants;  $C_p$ , average heat capacity of the reactant.

The initial and boundary conditions for variable  $x_3$  are

$$x_3(0, y) = 350^0\text{K}$$

$$x_3(t, 0) = 350^0\text{K}$$

The control  $T_h$  is bounded.

$$T_{ha} \leq T_h \leq T_{hb}$$

The Hamiltonian  $H_{T_h}$  is formed for this system

$$H_{T_h} = p_1 \left( -v \frac{\partial x_1}{\partial y} - k_1 x_1 \right) + p_2 \left( -v \frac{\partial x_2}{\partial y} + k_1 x_1 - k_2 x_2 \right)$$

$$+ p_3 \left[ -v \frac{\partial x_3}{\partial y} + c_3(T_h - x_3) + c_4(-k_1 x_1 \Delta H_1 - k_2 x_2 \Delta H_2) \right]$$

The equations for the auxiliary variables  $p_i$ 's are

$$\frac{\partial p_1}{\partial t} + v \frac{\partial p_1}{\partial y} = (p_1 - p_2 + p_3 c_4 \Delta H_1) k_1$$

$$\frac{\partial p_2}{\partial t} + v \frac{\partial p_2}{\partial y} = (p_2 + p_3 c_4 \Delta H_2) k_2$$

$$\begin{aligned} \frac{\partial p_3}{\partial t} + v \frac{\partial p_3}{\partial y} = & (p_1 - p_2 + p_3 c_4 \Delta H_1) x_1 \frac{\partial k_1}{\partial x_3} \\ & + (p_2 + p_3 c_4 \Delta H_2) x_2 \frac{\partial k_2}{\partial x_3} + p_3 c_3 \end{aligned}$$

The terminal and boundary conditions for the auxiliary variables are given by:

$$p_i(T, y) = 0 \quad i=1, 2, 3$$

$$p_i(t, Y) = 0 \quad i=1, 3$$

$$p_2(t, Y) = \frac{1}{vT}$$

Only the term containing control  $T_h$  is considered directly in optimization.

$$H' = p_3 c_3 T_h$$

$$H'_i{}^t = \int_{(i-1)Y/N}^{iY/N} p_3 c_3 T_h \, dy \quad i=1 \text{ to } N$$

Since the Hamiltonians  $H'$  and  $H'_i{}^t$  are linear with respect to the control variable,  $T_h$ , we can conclude that the optimal control must be a bang-bang control.

For an ideal reactor we know that the optimal control is

$$T_h = T_{ha} \quad \text{if } p_3 c_3 < 0$$

$$T_h = T_{hb} \quad \text{if } p_3 c_3 > 0$$

Similarly, for 1-, 2-, 3-zone reactor we have

$$T_{hi} = T_{ha} \quad \text{if } \int_{(i-1)Y/N}^{iY/N} p_3 c_3 \, dy < 0$$

$$T_{hi} = T_{hb} \quad \text{if } \int_{(i-1)Y/N}^{iY/N} p_3 c_3 \, dy > 0$$

$$i=1 \text{ to } N$$

Since the special control function - a piecewise constant function described previously was calculated numerically for the one-, two-, three-zone case, the Hamiltonian functions which have to attain relative maxima for reactant temperature control and heating medium temperature control are  $H_{ij}^{t*}$ ,  $H'_{ij}^{t*}$  respectively.

$$H_{ij}^{t*} = \int_{T_{j-1}}^{T_j} H_i^t \, dt \quad i=1 \text{ to } N; \quad j=1, 2, \dots, 24$$

$$H_{ij}^{t*} = \int_{T_{j-1}}^{T_j} H_i^t dt \quad i=1 \text{ to } N; \quad j=1,2,\dots,24$$

The mode for the heating medium control is not necessarily a bang-bang control for this function.

The input data and corresponding yields are presented in Table 1.

The study of start-up operations of reactors with this reaction is subdivided into three cases:

(a1). The reactant temperature control;  $\frac{a_{22}}{a_{12}} \leq 1$ ;

(a2). The reactant temperature control;  $\frac{a_{22}}{a_{12}} > 1$ ;

(a3). The heating medium temperature control.

The results for these three cases are given below.

(a1). The reactant temperature control;  $\frac{a_{22}}{a_{12}} \leq 1$ .

Runs number 1 through 8 belong to this case. Only the residence time for runs number 1 through 4 differs from that for runs number 5 through 8. Other data are unchanged.

The optimum temperature profiles for different reactors are shown on Figures 6 and 8. The main feature of the temperature profiles is that they do not change with time.

Since we know that different fluid elements in the reactor have different temperature policies, we expect that the temperature policy for one-zone control would consist of



Table 1. Input data and the optimum yields for reaction A to B to C

Run no.	Reactor type	Residence time	Yield	Limits on control variable*		Rate constants <sup>1</sup>			
				Upper limit	Lower limit	a <sub>11</sub>	a <sub>21</sub>	a <sub>12</sub>	a <sub>22</sub>
1	Ideal	0.6	0.402	400 <sup>0</sup> K	300 <sup>0</sup> K	0.128E07 <sup>2</sup>	0.400E01	0.490E04	0.000E00
2	1-zone	"	0.330	"	"	"	"	"	"
3	2-zone	"	0.382	"	"	"	"	"	"
4	3-zone	"	0.402	"	"	"	"	"	"
5	Ideal	0.4	0.390	"	"	"	"	"	"
6	1-zone	"	0.369	"	"	"	"	"	"
7	2-zone	"	0.390	"	"	"	"	"	"
8	3-zone	"	0.383	"	"	"	"	"	"
9	Ideal	1.0	0.462	"	"	0.200E14	0.800E16	0.900E04	0.110E05
10	1-zone	"	0.437	"	"	"	"	"	"
11	2-zone	"	0.457	"	"	"	"	"	"

\*Runs number 1 through 15, control variables are reactant temperatures; runs number 16 through 19 control variables are temperature of heating medium.

<sup>1</sup> $k_i = a_{i1} \exp(-a_{i2}/T_r)$  per unit time, where  $T_r$  is the temperature of the reactants.

<sup>2</sup> $0.128E07 = 0.128 \cdot 10^7$ .

Table 1 (Continued)

Run no.	Reactor type	Residence time	Yield	Limits on control variable*		Rate constants <sup>1</sup>			
				Upper limit	Lower limit	a <sub>11</sub>	a <sub>21</sub>	a <sub>12</sub>	a <sub>22</sub>
12	3-zone	"	0.460	"	"	"	"	"	"
13	1-zone	1.0	0.535	400 <sup>0</sup> K	300 <sup>0</sup> K	0.400E14	0.800E16	0.900E04	0.110E05
14	2-zone	"	0.559	"	"	"	"	"	"
15	3-zone	"	0.566	"	"	"	"	"	"
16 <sup>3</sup>	1-zone	0.6	0.306	380 <sup>0</sup> K	300 <sup>0</sup> K	0.128E07	0.400E01	0.490E04	0.000E00
17	3-zone	"	0.333	"	"	"	"	"	"
18	1-zone	"	0.344	400 <sup>0</sup> K	300 <sup>0</sup> K	"	"	"	"
19	3-zone	"	0.436	"	"	"	"	"	"

<sup>3</sup>For runs number 16 through 19, control variables are heating medium temperature, other data needed are:

$$c_3 = 20$$

$$c_4 = 0.001$$

$$\Delta H_1 = -50,000$$

$$\Delta H_2 = -200,000.$$

some intermediate temperature between its upper and lower bounds. But Figures 6 and 8 show that there is no compromise and the fluid elements which are nearer to the inlet of the reactor are ignored in the process of deciding a common temperature policy.

The temperature profiles for two ideal reactors (runs number 1 and 5) also show that the ideal controls consist of two segments: the first segment nearer to the inlet is at the lowest possible temperature; the second segment nearer to the outlet is at the highest possible temperature and has a residence time 0.2. This indicates that the zone control reactor should be designed in such a way that the last zone of reactor should have a residence time 0.2. This also implies that a possible optimal control operation should be considered at the reactor's design stage.

The yields are shown in Table 2. The comparison of yields among the first set (runs number 1 through 4) shows that the three-zone reactor has the same yield as the ideal reactor and is the best among the zone control reactors; the two-zone reactor is inferior to the three-zone reactor but is superior to the one-zone reactor. However, the comparison of yields among the second set (runs number 5 through 8) shows that the two-zone reactor has the same yield as the ideal reactor and is the best among zone control reactors; the three-zone reactor is inferior to the two-zone reactor but is superior

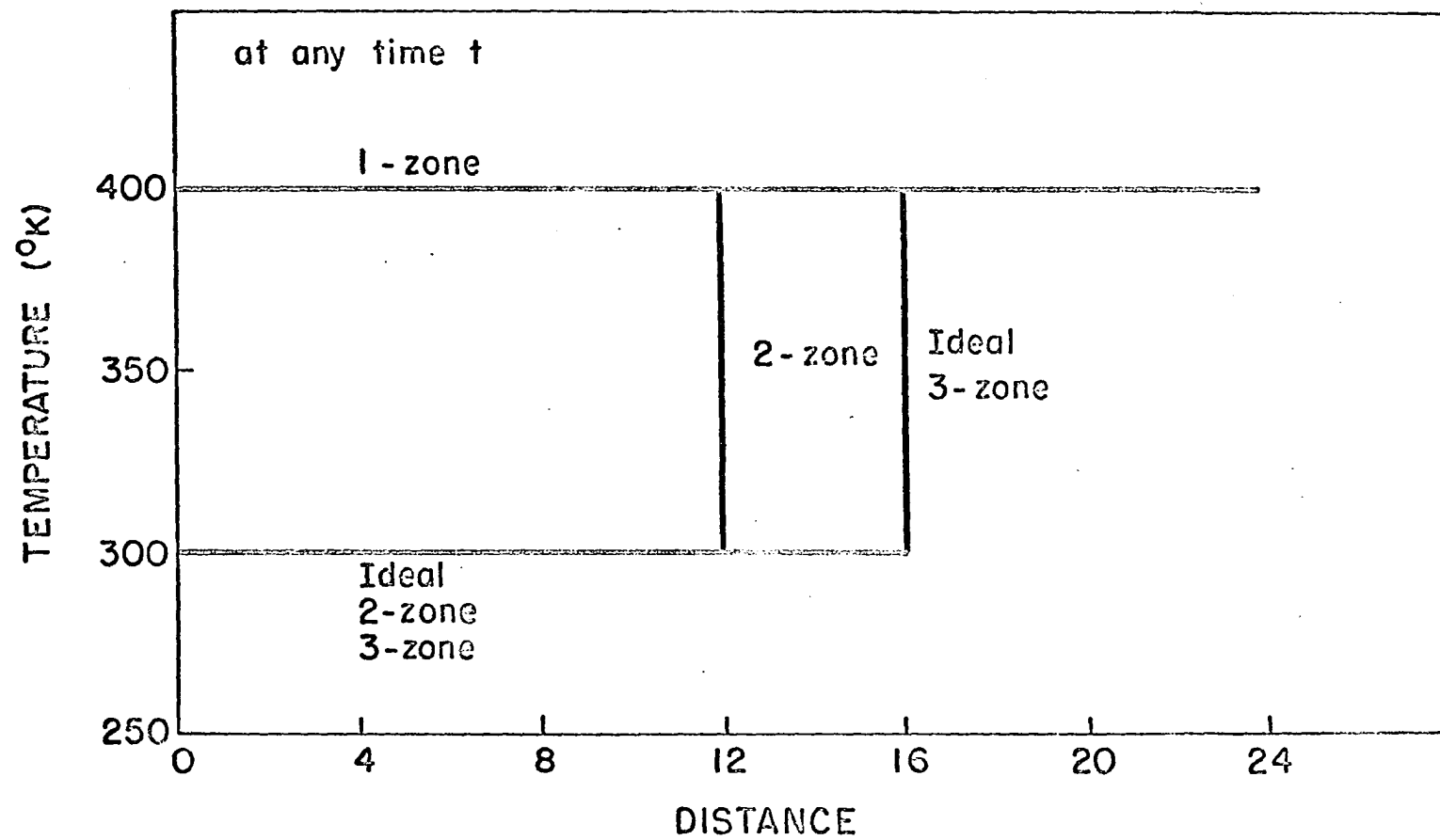


Figure 6. Temperature profiles of various tubular reactors  
(runs number 1 through 4)

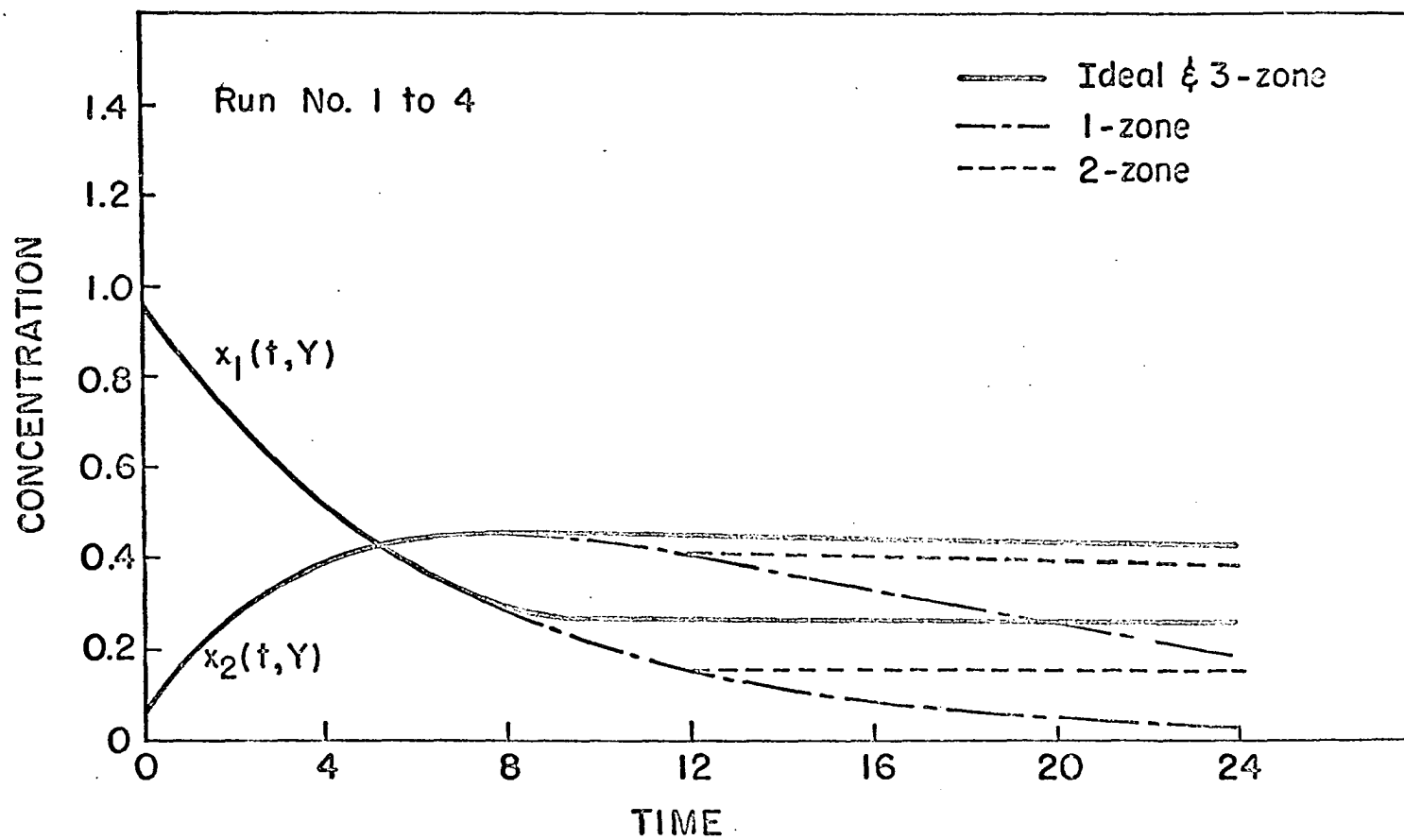


Figure 7. Output of various tubular reactors

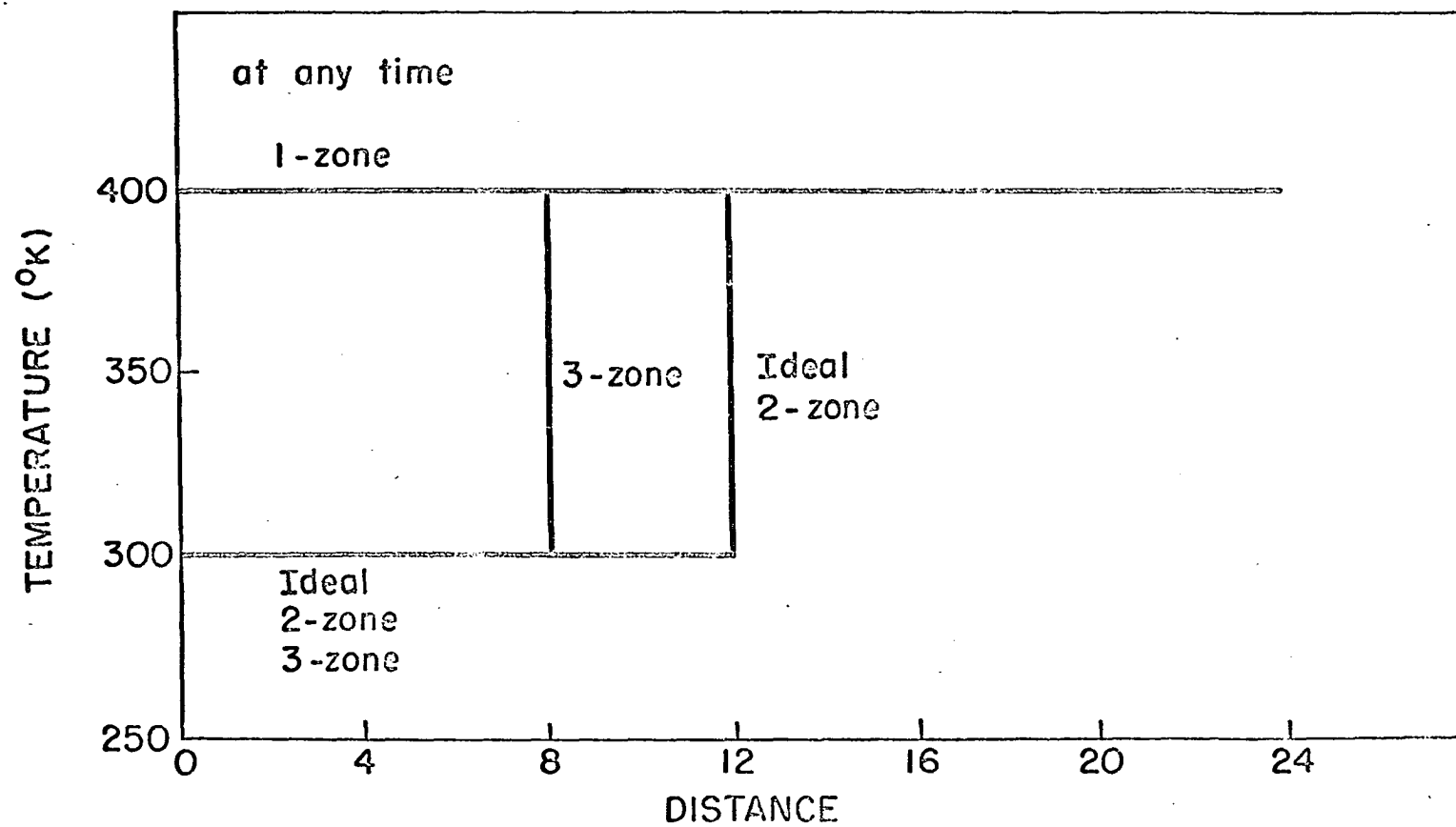


Figure 8. Temperature profiles of various tubular reactors  
(runs number 5 through 8)

Table 2. Yields for reaction A to B to C; case (a1), reactant temperature control

Run no.	Reactor type	Residence time	Yield
1	Ideal	0.6	0.402
2	1-zone	"	0.330
3	2-zone	"	0.382
4	3-zone	"	0.402
5	Ideal	0.4	0.390
6	1-zone	"	0.369
7	2-zone	"	0.390
8	3-zone	"	0.383

to the one-zone reactor.

From the results of this case, we conclude that a more complex control operation does not necessarily result in a better yield.

The outlet concentrations of the reactors for runs number 1 through 4 are shown on Figure 7. Curves for all reactors coincide with each other at the first eight periods of time. This fact corresponds to the coincidence of one third of the profiles near the ends of the reactors. The curves for the one zone case follow that of the two zone case another four periods of time. This also corresponds to the coincidence of another one sixth length of the profile between two reactors. These curves deviate more and more toward the end of twenty-four periods. This is an expected result in view of the differences in control strategy which occur near the reactor inlet. The fluid elements nearer to the inlets of the

reactors experience more difference in the control action before they finally emerge at the outlet of the reactor.

(a2). The reactant temperature control;  $\frac{a_{22}}{a_{12}} > 1$ .

The runs number 9 through 15 belong to this case. The temperature profiles for various reactors are shown on Figures 9 and 10. We find that they change with time. We also find that the zone temperature profiles try to approximate the ideal profile on some weighted average. The better approximation is obtained by more complex and more flexible controls. The yields are given in Table 3. For the first set of data (runs number 9 through 12), the three-zone reactor obtains a yield equivalent to 99.5% of the ideal yield and is the best among the zone control reactors. For this case, we conclude that three-zone reactor is better than a two-zone reactor and the two-zone reactor is better than a one-zone reactor.

From the results of above two cases, we find the simple criterion  $\frac{a_{22}}{a_{12}} \leq 0$  or  $\frac{a_{22}}{a_{12}} > 0$ , determines whether the optimal, ideal and zoned temperature strategies are time-invariant or time-variable. This criterion is the same as one used by others to determine whether bang-bang or continuously variable temperature strategies are optimal in a batch reactor. When the ratio  $\frac{a_{22}}{a_{12}}$  is less than or equal to zero, the Hamiltonians  $H$  and  $H_1^t$  have no local maximum. For this case, the ideal reactor is expected to have a bang-bang strategy; a zoned



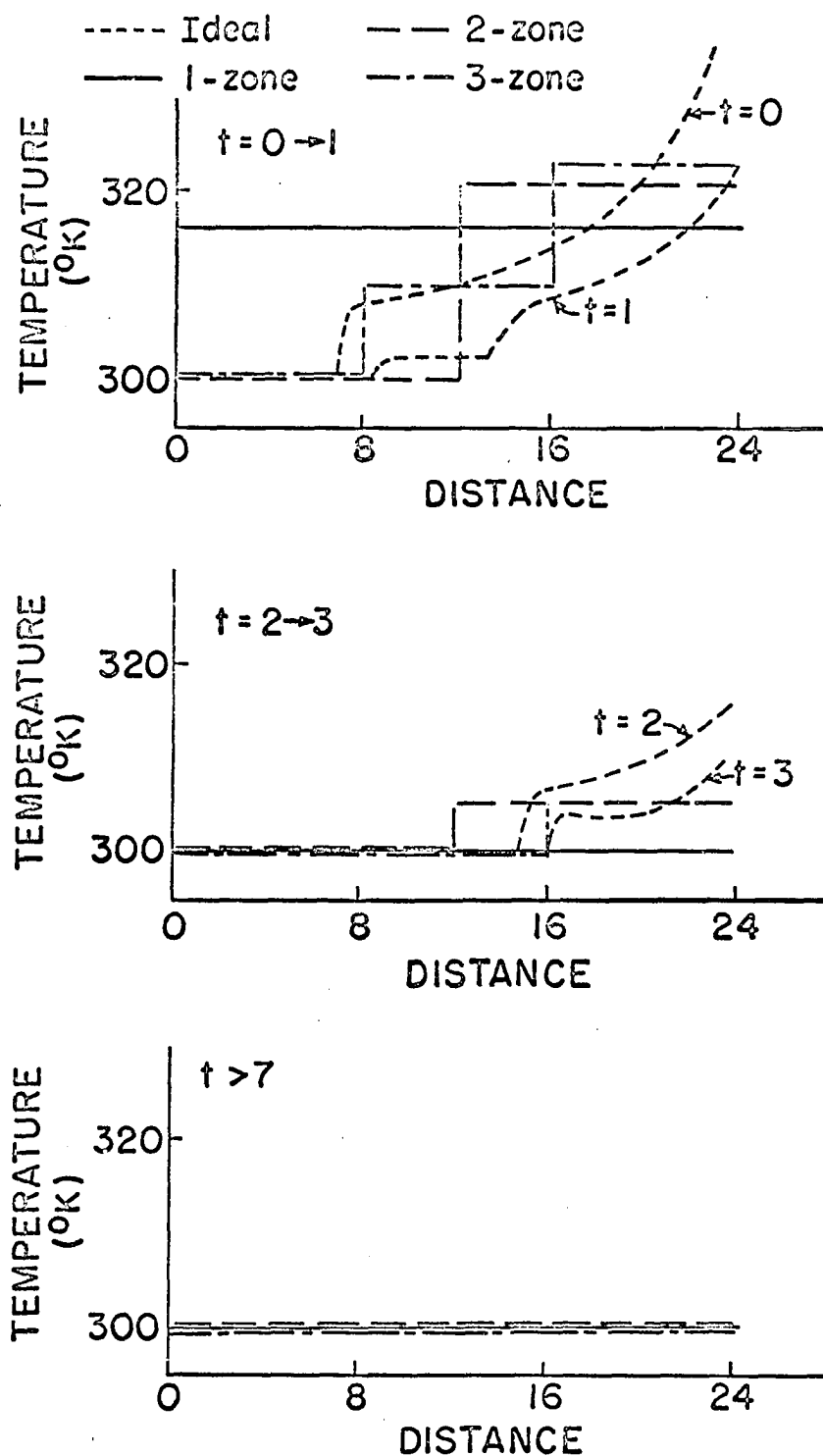


Figure 9. Temperature profiles of various tubular reactors (runs number 9 through 12)

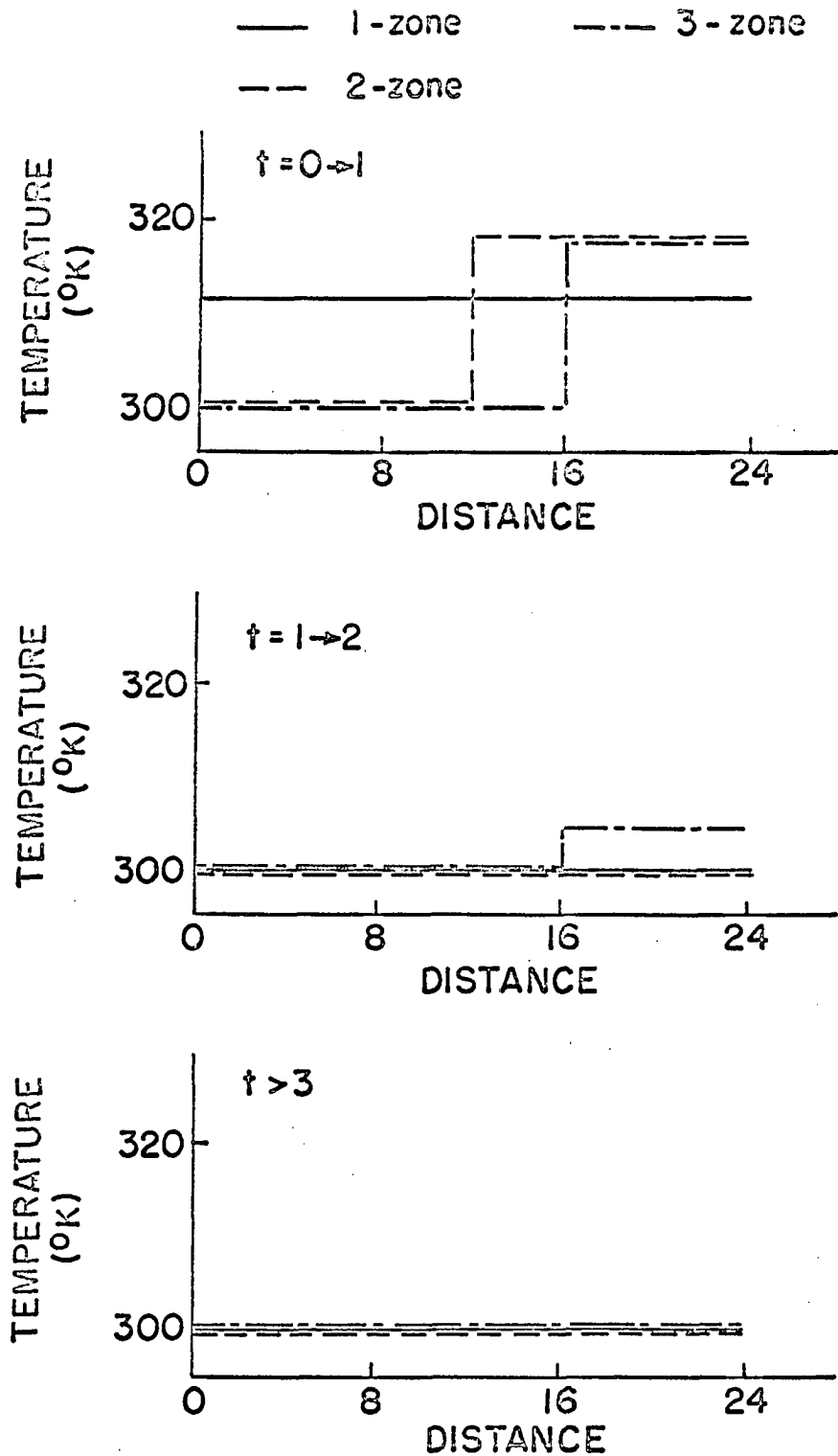


Figure 10. Temperature profiles of various tubular reactors (runs number 13 through 15)

Table 3. Yields for reaction A to B to C; case (a2), reactant temperature control

Run number	Reactor type	Residence time	Yield
9	Ideal	1.0	0.462
10	1-zone	"	0.437
11	2-zone	"	0.457
12	3-zone	"	0.460
13	1-zone	"	0.535
14	2-zone	"	0.559
15	3-zone	"	0.566

reactor with a piecewise constant control may have a bang-bang strategy. In our results, we find the bang-bang strategies and also a time-invariant character of the strategies.

(a3). Heating medium temperature control

Runs number 16 through 19 belong to this case.

A direct manipulation of reactant temperature is impossible in an actual situation. But reactant temperature may be manipulated indirectly by varying the temperature of heating medium. Therefore, the study of heating medium temperature control is a necessary step for practical application. The efficiency of heat transfer as evidenced by the relative magnitude of  $c_3$  and  $c_4$  plays a very important role in this case. If  $c_3$  is very large compared to  $c_4$ , then the difference between reactant temperature  $x_3$  and heating medium temperature is very small and we can almost reproduce the

desired zone reactant temperature. On the other hand, if  $c_3$  is very small compared to  $c_4$ , we have almost no control over reactant temperature, the reactant temperature is essentially the adiabatic temperature of the reactor with main source of heat from reaction heats. An intermediate value  $c_3$  corresponds to a situation with a partial control of reactant temperature.

The numerical studies correspond to the cases with partial control of reactant temperature. The temperature profiles of the heating medium are shown in Figures 11 and 13. The reactant temperature profiles are shown in Figures 12 and 14. The reactant temperature profiles have shapes analogous to those of the heating medium profiles. The lower and upper limits of reactant temperature reached in runs number 16 and 17 are  $300^{\circ}\text{K}$  and  $400^{\circ}\text{K}$ . Since the zone reactant temperature control reactors, runs number 2 to 4, have equal limits on reactant temperatures and the same kinetic data for chemical reaction, the best reactant temperature profile among these three runs - that of three-zone reactor, is also shown in Figure 12. The three-zone heating medium control apparently can simulate better the three-zone reactant temperature control than the one-zone heating medium control and thus gives a better yield. The yields for the one-zone and three-zone heating medium control reactors are 0.306 and 0.333 which are respectively 76.0% and 83.8% of the yield of the three-zone

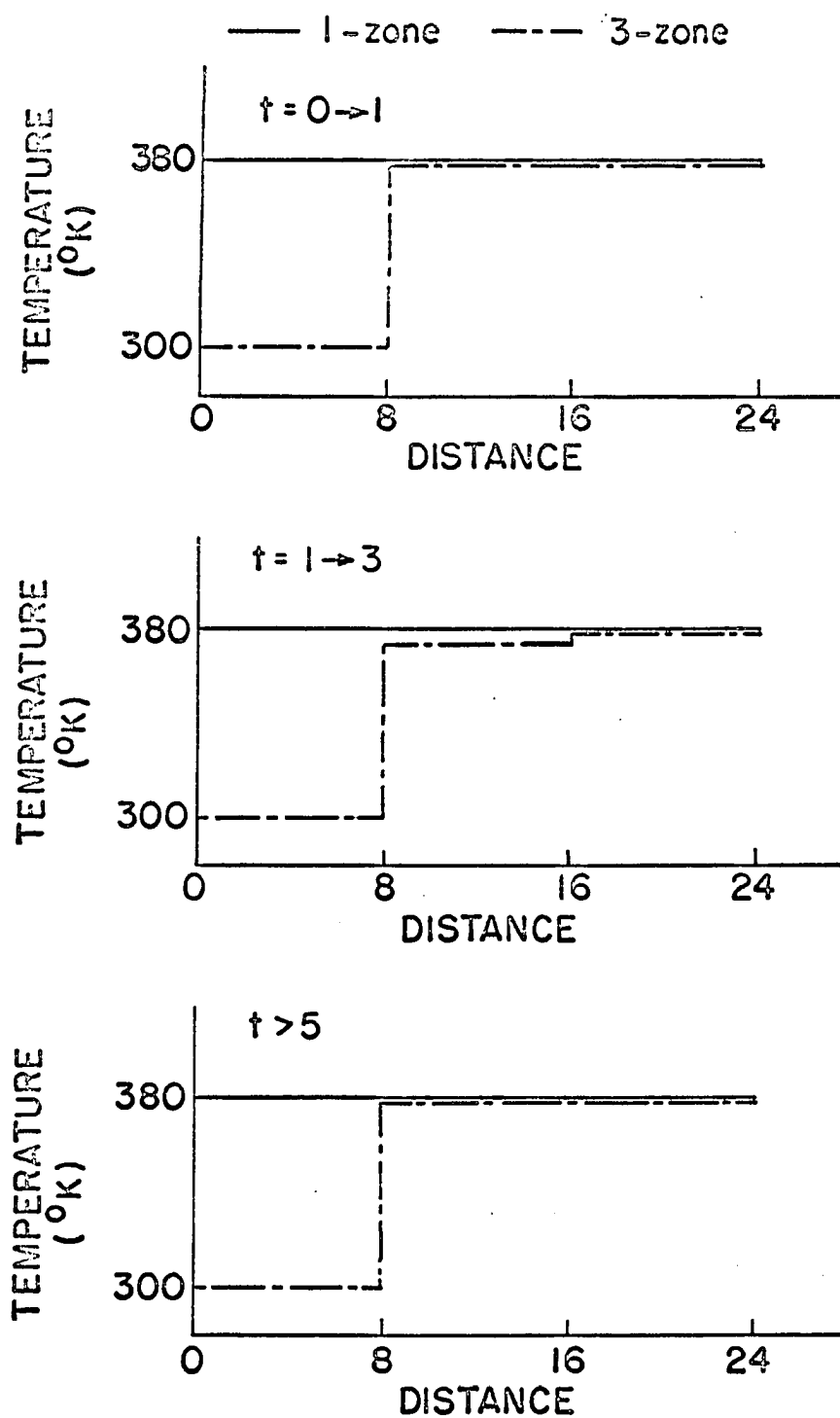


Figure 11. Temperature profiles for heating medium, 1-zone and 3-zone reactors (runs number 16 and 17)

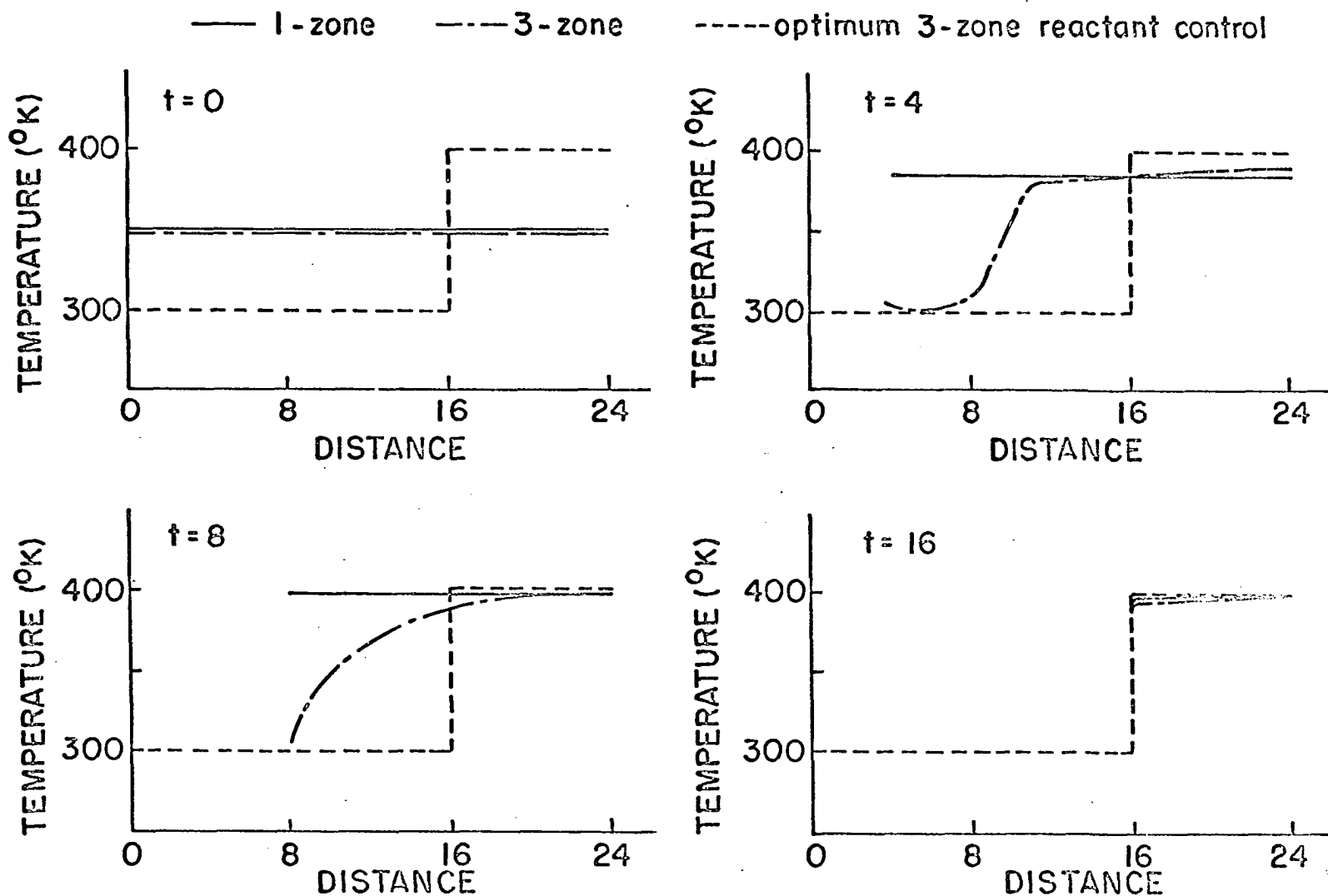


Figure 12. Reactant temperature profiles for multi-zone heating medium control reactors (runs number 16 and 17) and 3-zone reactant temperature control reactor (run number 4)

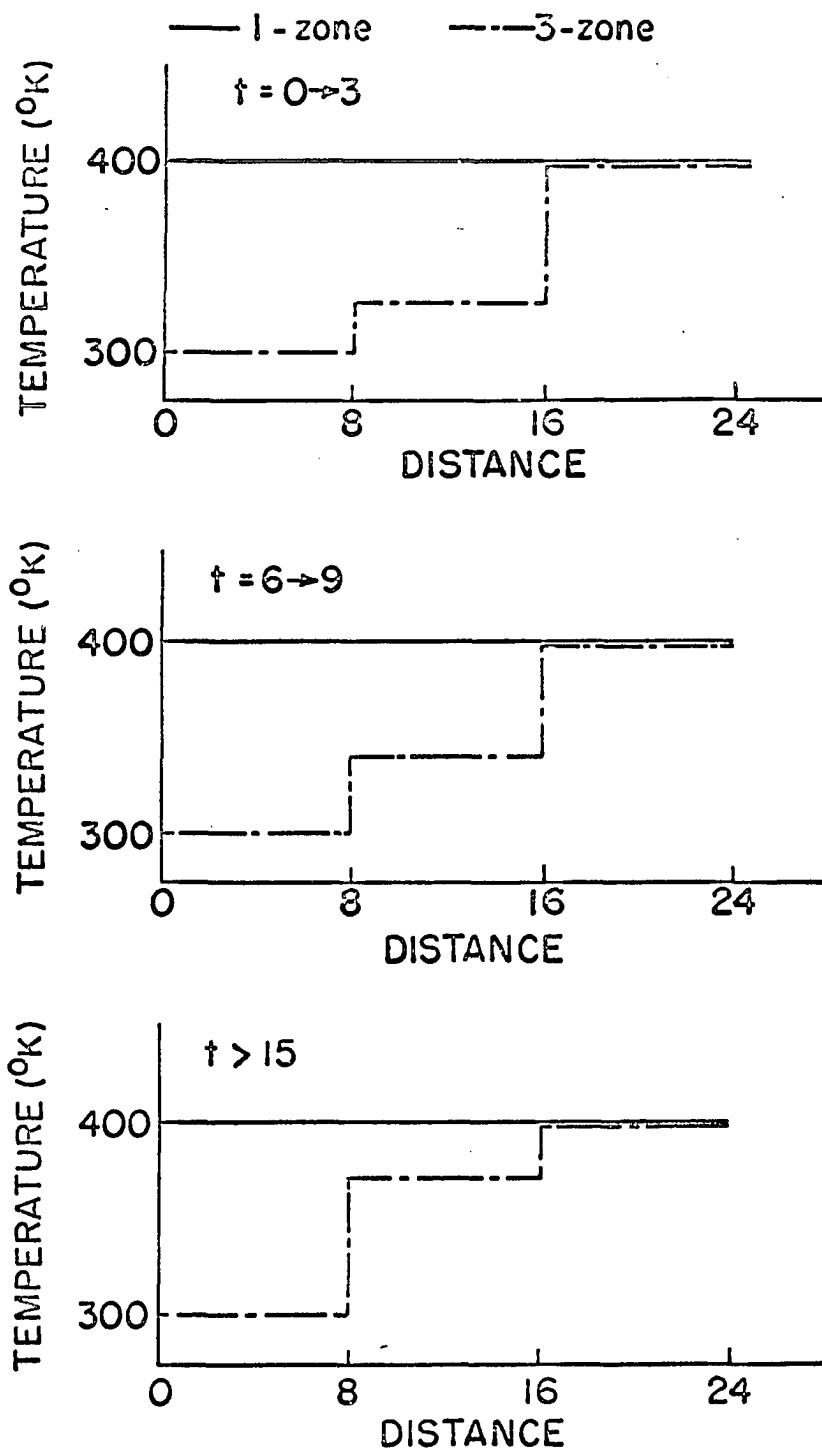


Figure 13. Temperature profiles for heating medium, 1-zone and 3-zone reactors (runs number 18 and 19)

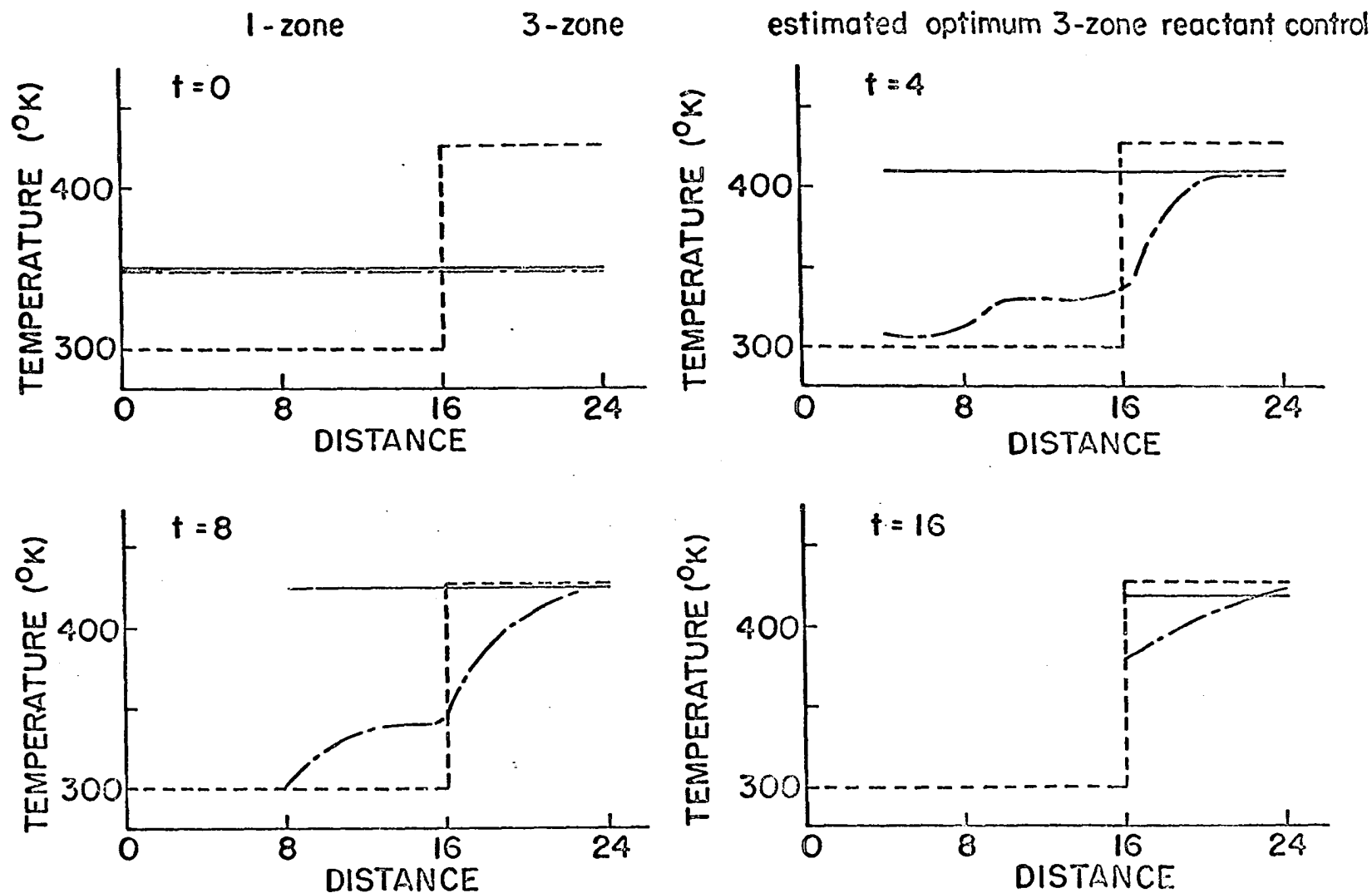


Figure 14. Reactant temperature profiles for multi-zone heating medium control reactors (runs number 18 and 19) and 3-zone reactant temperature control reactor (estimate)



reactant temperature control reactor. An estimate of the optimum reactant temperature profile for the three-zone reactant temperature control reactor is shown in Figure 14, assuming the same lower and upper limits for reactant temperatures reached in runs number 18 and 19. The three-zone heating medium control apparently also can simulate better the three-zone reactant control and thus gives a better yield. The yields for the one-zone and the three-zone reactors are respectively 0.344 and 0.436. Since the yield of the three-zone reactant temperature control reactor with same limits on the reactant temperature is not calculated, no comparison can be made with respect to the last yield. However, it appears that the comparison may be similar to that for runs number 16 and 17 with respect to run number 4.

Table 4. Yields for reaction A to B to C; case (a3), heating medium temperature control

Run number	Reactor type	Residence time	Yield
16	1-zone	0.6	0.306
17	3-zone	"	0.333
18	1-zone	"	0.344
19	3-zone	"	0.436

(b). The optimum start-up operations of tubular reactors for ethylene oxide production.

The data for ethylene oxidation to ethylene oxide is obtained from Slinko et al. (28). They studied ethylene oxide

production for one to three stages of complete mixing reactors, using dynamic programming.

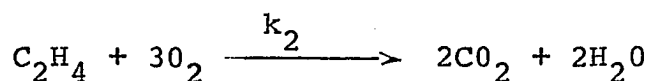
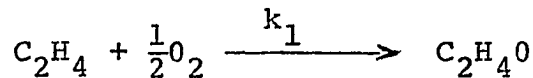
The present numerical study of this process is concerned with optimum start-up operation. We seek the reactant temperature strategy which attains the maximum average yield of ethylene oxide over one residence time for the ideal, one-, two- and three-zone reactors.

The performance index to be minimized is expressed by

$$I = - \frac{1}{T} \int_0^T x_2(t, Y) dt$$

where  $x_2$  is the ethylene oxide concentration;  $T$ , one residence time of the reactor;  $Y$ , space coordinate of the outlet of the reactor.

The chemical process of ethylene oxidation can be expressed by two parallel reactions.



$$k_1 = 6.5 \cdot 10^6 \exp (-15,200/RT_r) \text{sec}^{-1}$$

$$k_2 = 3.0 \cdot 10^8 \exp (-19,800/RT_r) \text{sec}^{-1}$$

The reactant temperature  $T_r$  is bounded by

$$483^0K \leq T_r \leq 543^0K$$

The equations for the unsteady-state tubular reactor are

$$\frac{\partial x_1}{\partial t} = -v \frac{\partial x_1}{\partial y} - (k_1 + k_2) \frac{x_1}{1 + a(1 - x_1/c_0)}$$

$$\frac{\partial x_2}{\partial t} = -v \frac{\partial x_2}{\partial y} + k_1 \frac{x_1}{1 + a(1 - x_1/c_0)}$$

where  $x_1$  is the concentration of ethylene; axial velocity  $v$  is assumed constant;  $a$  is an empirical constant;  $c_0$  can be considered as an empirical constant.

$$a = 5.7$$

$$c_0 = 0.100$$

The initial and boundary conditions of the state variable are

$$x_1(0, y) = 0.1$$

$$x_1(t, 0) = 0.1$$

$$x_2(0, y) = 0.0$$

$$x_2(t, 0) = 0.0$$

The Hamiltonian  $H$  which includes only terms containing the control variable is

$$H = [-p_1(k_1 + k_2) + p_2 k_1] \frac{x_1}{1 + a - ax_1/c_0}$$

The auxiliary variables  $p_i$ 's are defined by

$$\frac{\partial p_1}{\partial t} + v \frac{\partial p_1}{\partial y} = [p_1(k_1 + k_2) - p_2 k_1] \frac{1 + a}{(1 + a - ax_1/c_0)^2}$$

$$\frac{\partial p_2}{\partial t} + v \frac{\partial p_2}{\partial y} = 0$$

$$p_i(T, Y) = 0 \quad i=1, 2$$

$$p_i(t, Y) = 0$$

$$p_2(t, Y) = \frac{1}{vT}$$

Hamiltonian functions  $H_i^t$  for the  $i$ th zone of the  $N$ -zone reactor are given by

$$H_i^t = \int_{(i-1)Y/N}^{iY/N} (-p_1^{k_{1i}} - p_1^{k_{2i}} + p_2^{k_{1i}}) \frac{x_1}{1+a-ax/c_0} dy \quad i=1 \text{ to } N$$

The Hamiltonians for the piecewise constant control are then,

$$H_{ij}^{t*} = \int_{T_{j-1}}^{T_j} H_i^t dt \quad i=1 \text{ to } N; \quad j=1, 2, \dots, 24$$

In the numerical calculation, a residence time of 10 seconds is used for all the reactors.

The optimum temperature profiles for the ideal reactor and one-, two-, and three-zone reactors are shown in Figure 15.

The zone temperature profiles approximate an ideal reactor profile and a more complex and more flexible control can do this better. Thus, a more complex and more flexible control gives a better yield. The yields of one-, two-, three-zone reactors and ideal reactor are 0.0591, 0.0610, 0.0619 and 0.0636 respectively. The three-zone reactor obtains a

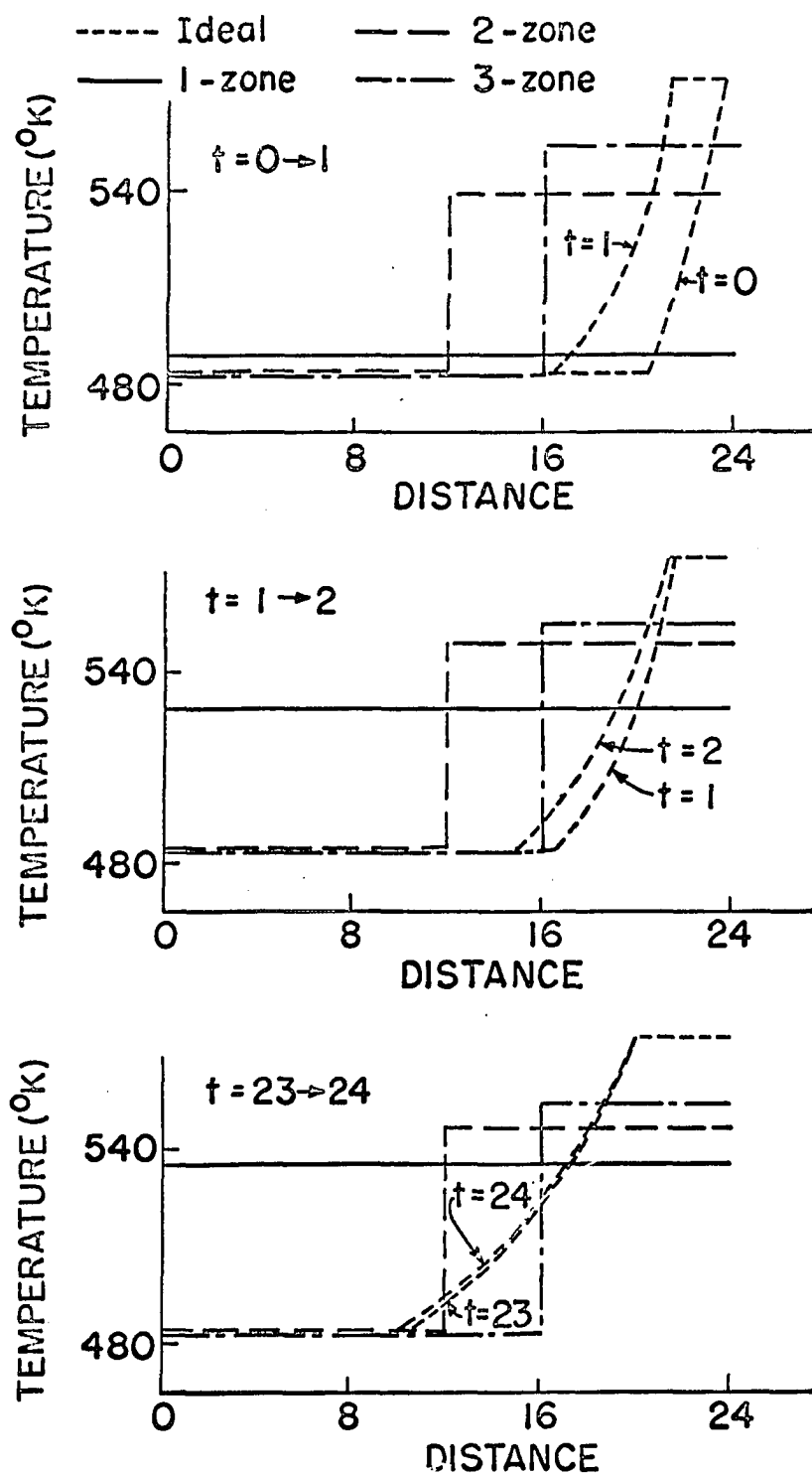


Figure 15. Temperature profiles of various tubular reactors, ethylene oxide production

yield equivalent to 97.2% of the ideal yield and is the best among the zone control reactors.

Table 5. Yields for ethylene oxide production

Reactor type	Yields
Ideal	0.0636
1-zone	0.0591
2-zone	0.0610
3-zone	0.0619

(c). The start-up operations of tubular reactors for formaldehyde production.

The oxidation of methanol to formaldehyde was also studied. The data was obtained from Polanovshaya and Topchieva (29). The kinetic expression given in the reference (29) is not a function of extensive properties of the system. Therefore, the data was analyzed and was fitted into a suitable kinetic expression. Again a start-up operation was optimized by choosing the best possible reactant temperatures for ideal, one-, two- and three-zone reactors. A maximum production of formaldehyde over one residence time of the reactor is desired. The performance index to be minimized can be expressed as

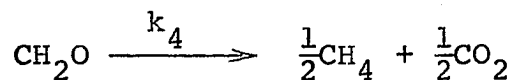
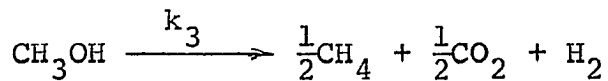
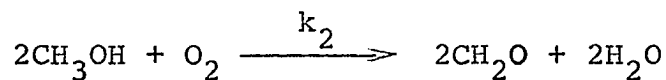
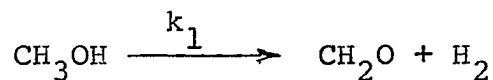
$$I = -\frac{1}{T} \int_0^T (1 - e^{-c(T-t)}) x_3(t, Y) dt$$

where

$$c = 1,500$$

and  $x_3$  is concentration of formaldehyde;  $T$ , one residence time of reactor;  $Y$ , space coordinate of the outlet of the reactor.

Four main reactions of the process are



The tubular reactor system can be represented by the set of equations

$$\frac{\partial x_1}{\partial t} = -v \frac{\partial x_1}{\partial y} - (k_1 + k_3)x_1 - 2k_2x_1^2x_2$$

$$\frac{\partial x_2}{\partial t} = -v \frac{\partial x_2}{\partial y} - k_2x_1^2x_2$$

$$\frac{\partial x_3}{\partial t} = -v \frac{\partial x_3}{\partial y} + k_1x_1 + 2k_2x_1^2x_2 - k_4x_3$$

with initial and boundary conditions

$$x_1(0,y) = 0.815 \cdot 10^{-5} \text{ g mole/cm}^3$$

$$x_1(t,0) = 0.815 \cdot 10^{-5} \text{ g mole/cm}^3$$

$$x_2(0,y) = 0.182 \cdot 10^{-5} \text{ g mole/cm}^3$$

$$x_2(t,0) = 0.182 \cdot 10^{-5} \text{ g mole/cm}^3$$

$$x_3(0,y) = 0.0 \text{ g mole/cm}^3$$

$$x_3(t,0) = 0.0 \text{ g mole/cm}^3$$

where  $x_1$  is the concentration of methanol;  $x_2$ , concentration of oxygen; axial velocity  $v$  is assumed constant.

The rate constants  $k_i$  have following forms of temperature dependence.

$$k_1 = 1.358 \cdot 10^{11} \exp(-29,800/RT_r) \text{ min}^{-1}$$

$$k_2 = 4.07 \cdot 10^{22} \exp(-27,500/RT_r) \text{ min}^{-1} (\text{g mole/cm}^3)^{-2}$$

$$k_3 = 1.871 \cdot 10^5 \exp(-9,470/RT_r) \text{ min}^{-1}$$

$$k_4 = 9.40 \cdot 10^6 \exp(-18,770/RT_r) \text{ min}^{-1}$$

The allowable temperature is bounded by

$$723^0\text{K} \leq T_r \leq 823^0\text{K}$$

The Hamiltonian  $H$  which includes only the terms containing



the control variable is:

$$H = -p_1[(k_1+k_3)x_1 + 2k_2x_1^2x_2] - p_2k_2x_1^2x_2 \\ + p_3(k_1x_1 + 2k_2x_1^2x_2 - k_4x_3)$$

The auxiliary variables  $p_i$ 's are defined by

$$\frac{\partial p_1}{\partial t} + v \frac{\partial p_1}{\partial y} = p_1(k_1+k_3+4k_2x_1x_2) + 2p_2k_2x_1x_2 \\ - p_3(k_1+4k_2x_1x_2)$$

$$\frac{\partial p_2}{\partial t} + v \frac{\partial p_2}{\partial y} = (2p_1+p_2-2p_3)k_2x_1^2$$

$$\frac{\partial p_3}{\partial t} + v \frac{\partial p_3}{\partial y} = p_3k_4$$

$$p_i(T, Y) = 0 \quad i=1, 2, 3$$

$$p_i(t, Y) = 0 \quad i=1, 2$$

$$p_3(t, Y) = (1-e^{-c(T-t)})/vT$$

$H_{ij}^t$ 's for the optimal piecewise constant function control are

$$H_{ij}^t = \int_{T_{j-1}}^{T_j} \int_{(i-1)Y/N}^{iY/N} H \, dy \, dt$$

$$i=1 \text{ to } N; \quad j=1, 2, \dots, 24$$

In the numerical study of this process, a residence time of 0.0018 minute is used for one-zone and three-zone reactors.

No increase of the yield is observed for the three-zone reactant temperature control reactor over the one-zone reactant temperature control reactor. Both attain a yield of 0.522.

This shows that a one-zone reactor is a good control, there is no need for installing a complicated control system.

The optimum temperature policies obtained are:

One-zone control:

Use  $823^{\circ}\text{K}$  for the first seventeen periods then use  $723^{\circ}\text{K}$  for the next seven periods.

Three-zone control:

Zone 1: Use  $823^{\circ}\text{K}$  for the first twelve periods, then use  $723^{\circ}\text{K}$  for the next twelve periods.

Zone 2: Same as zone 1.

Zone 3: Use  $823^{\circ}\text{K}$  for the first eighteen periods, then use  $723^{\circ}\text{K}$  for the remaining six periods.

## V. CONCLUSION

The following conclusions can be made from the preceeding study on tubular reactor system:

1. An extension of the Maximum Principle developed in this work provides an effective basis for the computation of optimal control in unsteady-state, plug-flow systems subjected to constrained control, such as zoned tubular reactors, and heat exchangers.
2. A detailed example has been developed involving flow rate control of a heat exchanger, such that the optimal strategy in the absence of continuing disturbances is periodic adjustment of flow rate.
3. Zoned temperature control may provide a practical means to simplify design and operation of tubular reactors without significant loss of yield.
4. Optimal zoned temperature profiles for a tubular reactor do not match in any simple way the ideal continuous temperature profiles in general.
5. Four specific reactions have been studied, for an unsteady-state tubular reactor system.
  - a.  $A \rightarrow B \rightarrow C$  (consecutive reactions). A simple criterion can be used to determine if the optimal zoned temperature strategy is time-invariant or time-variable. This criterion is the same as one used by others to deter-

mine if bang-bang or continuously variable temperature strategies are optimal in a batch reactor.

Usually more zones permit better performance, whether direct reactant temperature control is used, or heating medium temperature control. But in one case, two zones are superior to three zones.

- b.  $A \rightleftharpoons B$  (reversible reaction). In the ideal reactor, the best choice of reactant temperature is that temperature which will maximize local rate. In the zoned-reactor no similar principle applies.
- c. Ethylene oxide (parallel reactions). In this system, a three-zone reactor permits yield near the ideal yield even though the zoned temperature profiles does not appear to closely approximate the ideal temperature profile.
- d. Formaldehyde (multiple reactions). In this system, a one-zone reactor allows the same yield as a three-zone reactor.

## VI. NOMENCLATURE

The following nomenclature applies to all preceeding sections. For some terms, more than one meaning has been assigned. These terms are defined within the text of the section for which they apply.

$A'$ :	cross sectional area for the flow of heating medium or coolant
$a$ :	subscript for lower limit
$a_i, a_{ij}$ :	functions; constants
$b$ :	subscript for upper limit
$b_i, b'_i, b_{ik}$ :	functions
$c, c_i, c'_i$ :	constants
$c_{ij}, c'_{ij}$ :	functions
$C_p$ :	average heat capacity of reactant
$C'_p$ :	average heat capacity of heating medium or coolant
$d_j$ :	constants
$d_{si}$ :	functions
$E$ :	energy of activation
$e_i$ :	remainders; small constants
$f$ :	an $(n+q)$ dimensional vector function which has elements $f_i$ 's
$g, g', g_i$ :	functions
$H, H^t, H^B, H^Y,$ $H^I, H^{t*}_{ij}$	: Hamiltonians

$-\Delta H_j$ :	heat of generation (cal per unit of jth reaction)
$h$ :	constant
$h_i$ :	functions
$I, I_i$ :	performance index
$i$ :	integer
$j$ :	integer
$k, k_r$	integers
$k_i, k_{io}$ :	rate constants
$k_{in}$ :	variable
$L_i^!$ :	constants
$M, M_i, M_i^!$ :	constants
$m$ :	integer
$N$ :	integer; the last stage; total number of zones
$n, n'$ :	integer
$\bar{n}$ :	outer normal
$P$ :	pressure
$p, p'$ :	an $(n+q)$ dimensional auxiliary vector
$p_s$ :	auxiliary variable $p$ under a steady-state condition
$q_{in}$ :	variable
$R$ :	universal gas constant
$R_j$ :	rate of reaction for jth reaction
$R_t$ :	radius of tabular reactor
$r$ :	integer; adjustable part of flow rate
$r_i, r_i^!, r_i''$ :	functions; rates of reactions

$S$ :	space coordinate; space domain
$S_1, S_2$ :	hypersurface
$s$ :	dummy variable; transformed time variable
$s', s''$ :	integer
$s_{ij}$ :	stoichiometric coefficient
$T$ :	end of time; a constant
$T_h$ :	the temperature of the heating medium or coolant
$T_k$ :	constants
$T_r$ :	the reactant temperature
$t$ :	time variable
$t^*$ :	A fixed point of time
$U$ :	a closed bounded set in $r$ dimensional space with $\phi_k(u)=0$ ( $k=1,2,\dots,r$ ) as its boundary
$U_T$ :	over-all heat transfer coefficient
$u$ :	control vector which is an $r$ -dimensional vector with elements $u_1, u_2, \dots, u_r$
$u'$ :	collection of $u^t, u'', u^y$
$u^*$ :	modified control of $u$
$u'', u''(t, y)$ :	control vector which is function of both time $t$ and space $y$
$u^B(t)$ :	boundary control which is present only in boundary conditions
$u^I(y)$ :	initial control which is present in initial conditions
$u^t, u^t(t)$ :	control vector which is function of time only
$u^{t*}$ :	modified control of $u^t$
$u^y, u^y(y)$ :	control vector which is function of space only
$u_D$ :	the control vector which is defined as $u - u_s$

$u_s$ :	the control vector for the steady state case
$v, v_i$ :	linear velocities of fluids
$w_i, w_i'$ :	constants
$X$ :	constant
$x$ :	state vector which is an n-dimensional vector with elements $x_1, x_2, \dots, x_n$ ; independent variable
$x_D$ :	state vector which is defined as $x - x_s$
$x_s$ :	the state vector under steady state condition which has elements $x_{si}$ 's
$Y$ :	end of space; a constant
$y$ :	space variable; distance from inlet; dependent variable
$y^*$ :	a fixed point of space
$y_i'$ :	transformed space variable
$z$ :	a 3 n-dimensional vector, $z = (x_1, x_2, \dots, x_n, p_1, p_2, \dots, p_n, x_{1y}, x_{2y}, \dots, x_{ny})$
$\alpha$ :	a positive constant
$\delta$ :	a small region
$\delta_{ij}$ :	Kronecker delta; $\delta_{ij}=1$ if $i=j$ , $\delta_{ij}=0$ if $i \neq j$
$\epsilon$ :	belong to
$\theta$ :	normalized temperature
$\theta_i$ :	constant vectors
$\rho$ :	average density of reactants
$\rho'$ :	average density of heating medium or coolant
$\sum$ :	summation
$\phi, \phi', \phi_k$ :	functions



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## VIII. ACKNOWLEDGMENTS

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IX. APPENDIX A: DERIVATION OF  
EXPRESSION FOR INCREMENT  $\Delta I$

Let  $S$  be the region  $0 \leq t \leq T$ ,  $0 \leq y \leq Y$ . The admissible control vector  $u$  is a piecewise continuous function with its range in set  $U$ . The  $U$  is a closed bounded set with its boundaries  $\phi_k(u)=0$  ( $k=1,2,\dots,r$ ). The control vector  $u$  has  $r$  elements. They are formed into five subgroups  $u^B, u^t, u'', u^Y, u^I$  and are also arranged in such order. Subgroups  $u^B, u^t, u'', u^Y, u^I$  respectively have  $m', m-m', m''-m, m''-m''$  and  $r-m''$  elements. The notation  $u'$  in the following derivation contains subgroups  $u^t, u''$  and  $u^Y$ . Subgroup  $u^B$  represents boundary control whose elements are functions of time only. Subgroup  $u^t$  consists of elements which are also functions of time only but do not appear in the boundary conditions. Subgroup  $u''$  consists of elements which are both functions of time and distance. Subgroup  $u^Y$  consists of elements which are functions of space  $y$  only and do not appear in the initial conditions. Subgroup  $u^I$  consists of elements which are functions of  $y$  only but appear only in the initial conditions.

Vectors  $x$  and  $p$  are piecewise continuous and have piecewise continuous partial derivatives with respect to  $t$  and  $y$ . The only permissible lines of discontinuity for the elements  $x_k$  and  $p_k$  of vectors  $x$  and  $p$  are the curves represented by:

$$y - \int_0^t v_k(t) dt = c_k$$

where  $c_k$  are constants. Though it is possible to have many lines of discontinuity for  $x$  and  $p$ , the optimum theorems will be obtained for a system with  $n$  lines of discontinuity. However, the same optimum theorems can be obtained for a system with any number of lines of discontinuity by applying the same procedure of derivation. Now, let  $c_{k-}$  and  $c_{k+}$  be two constants with infinitesimal difference in their value, and let  $C_{k-}$  and  $C_{k+}$  be the corresponding curves on  $t, y$ -plane, then along these curves  $x_k$  and  $p_k$  are continuous. But along a line of constant time, there is a jump of values for  $x_k$  or  $p_k$  if one passes from curve  $C_{k-}$  to  $C_{k+}$  or vice versa. Along a constant distance line, one may also observe a jump of values for  $x_k$  and  $p_k$ , if one passes from curve  $C_{k-}$  to  $C_{k+}$ .

The optimum theorems derived with the assumption of piecewise continuous  $x_i$  and  $p_i$  have much wider scope of application than the similar theorems derived with the assumption of continuous  $x_i$  and  $p_i$ . In an original derivation, we had assumed continuous  $x_i$  and  $p_i$  and found that the performance index had to be defined in such a way that the following conditions were fulfilled.

$$\lim_{t \rightarrow T-} p_i(t, 0) = \lim_{y \rightarrow 0+} p_i(T, y)$$

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

We also noticed that the original theorems could not be applied to a system with discontinuous boundary conditions such as a unit step function.

Functions  $f_i (i=0,1,\dots,n+q)$ ,  $g_i (i=0,1,\dots,s')$ ,  $\frac{\partial f_i}{\partial x_k}$  ( $i=0,1,\dots,n+q$ ;  $k=1,2,\dots,n$ ),  $\frac{\partial g_i}{\partial x_k}$  ( $i=0,1,\dots,s'$ ;  $k=1,2,\dots,n$ ) and  $\frac{\partial h_i}{\partial x_k}$  ( $i=0,1,\dots,s''$ ;  $k=1,2,\dots,n$ ) are assumed to be continuous with respect to their arguments. Functions  $x_{i0}(y, u^I)$  and  $x_{i1}(t, u^B)$  are continuously differentiable functions. The linear velocities  $v_i$  can either be given time functions or be elements of the control vector.

Let  $u$  be the optimal control of the problem. Let  $x+\Delta x$  and  $p+\Delta p$  be respectively the state vector and auxiliary vector corresponding to the modified control  $u^*$  (i.e.,  $u+\Delta u$ ). The modified control  $u^*$  keeps the following integrals constant:

$$\int_0^T \int_0^Y f_i \, dy \, dt$$

$$i=n+1, n+2, \dots, n+q \quad (A. 1)$$

$$\int_0^T g_i \, dt$$

$$i=1, 2, \dots, s' \quad (A. 2)$$

$$\int_0^Y h_i \, dt$$

$$i=1, 2, \dots, s'' \quad (A. 3)$$

The increment of functional I due to this change in control is

$$\begin{aligned}
 \Delta I = & - \int_0^T w_0 [g_0(t, x(t, 0) + \Delta x(t, 0), x(t, Y) \\
 & + \Delta x(t, Y), u^{B*}, u^{t*}) - g_0(t, x(t, 0), x(t, Y), u^B, u^t)] dt \\
 & - \int_0^Y w'_0 [h_0(y, x(0, y) + \Delta x(0, y), x(T, y) + \Delta x(T, y), u^{I*}, u^{Y*}) \\
 & - h_0(y, x(0, y), x(T, y), u^I, u^Y)] dy \\
 & - \int_0^T \int_0^Y p_0 [f_0(t, y, x + \Delta x, u^{I*}) - f_0(t, y, x, u^I)] dy dt
 \end{aligned}
 \tag{A. 4}$$

Since the increments of the integrals in (A. 1), (A. 2), (A. 3) are zero, the increment  $\Delta I$  can also be written as

$$\begin{aligned}
 \Delta I = & - \int_0^T \sum_{k=0}^{s^I} w_k [g_k(t, x(t, 0) + \Delta x(t, 0), x(t, Y) + \Delta x(t, Y), u^{B*}, u^{t*}) \\
 & - g_k(t, x(t, 0), x(t, Y), u^B, u^t)] dt \\
 & - \int_0^Y \sum_{k=0}^{s^u} w'_k [h_k(y, x(0, y) + \Delta x(0, y), x(T, y) + \Delta x(T, y), u^{I*}, u^{Y*}) \\
 & - h_k(y, x(0, y), x(T, y), u^I, u^Y)] dy
 \end{aligned}$$



$$\begin{aligned}
& - \int_0^T \int_0^Y p_0 [f_0(t, y, x + \Delta x, u'^*) - f_0(t, y, x, u')] dy dt \\
& - \int_0^T \int_0^Y \sum_{k=n+1}^{n+q} p_k [f_k(t, y, x + \Delta x, u'^*) - f_k(t, y, x, u')] dy dt
\end{aligned}
\tag{A. 5}$$

The Lagrange multipliers  $w_k$ ,  $w'_k$  and  $p_j$  ( $j=n+1$  to  $n+q$ ) are constants which are so chosen that they are consistent with the constraints imposed on integrals (A. 2), (A. 3) and (A. 1), and the requirement that the functional  $I$  is minimum.

A function  $H$  is defined as:

$$H = \sum_{k=0}^{n+q} p_k f_k \tag{A. 6}$$

The functions  $f_k$  ( $k=1, 2, \dots, n$ ) are related to the partial derivatives of  $x_k$  with respect to time by equations (A. 7), and are defined by equation (A. 7a).

$$\frac{\partial x_k}{\partial t} = f_k \quad k=1, 2, \dots, n \tag{A. 7}$$

$$f_k = -v_k(t) \frac{\partial x_k}{\partial y} + r_k(t, y, x, u') \tag{A. 7a}$$

$k=1, 2, \dots, n$

The equations (A. 7) can also be written in term of  $H$  as:

$$\frac{\partial x_k}{\partial t} = \frac{\partial H}{\partial p_k} \quad k=1,2,\dots,n \quad (\text{A. 8})$$

The auxiliary variable  $p_k$  ( $k=1,2,\dots,n$ ) are defined by the equations

$$\frac{\partial p_k}{\partial t} = -v_k \frac{\partial p_k}{\partial y} - \frac{\partial H}{\partial x_k} \quad k=1,2,\dots,n \quad (\text{A. 9})$$

with initial and boundary conditions

$$p_k(t, Y) = \sum_{j=0}^{s'} \frac{w_j}{v_k} \frac{\partial g_j(t, x(t, 0), x(t, Y), u^B, u^t)}{\partial x_k(t, Y)} \quad k=1,2,\dots,n' \quad (\text{A. 10})$$

$$p_k(t, 0) = - \sum_{j=0}^{s'} \frac{w_j}{v_k} \frac{\partial g_j(t, x(t, 0), x(t, Y), u^B, u^t)}{\partial x_k(t, 0)} \quad k=n'+1, n'+2, \dots, n \quad (\text{A. 11})$$

$$p_k(T, Y) = \sum_{j=0}^{s''} w_j' \frac{\partial h_j(y, x(0, y), x(T, Y), u^I, u^Y)}{\partial x_k(T, Y)} \quad k=1,2,\dots,n \quad (\text{A. 12})$$

Integrating (A. 6) and making use of (A. 7), we obtain

$$\int_0^T \int_0^Y [p_0 f_0 + \sum_{k=1}^n p_k \frac{\partial x_k}{\partial t} + \sum_{k=n+1}^{n+q} p_k f_k - H] dy dt = 0 \quad (\text{A. 13})$$

The variation of the expression (A. 13) due to modification of control is

$$\begin{aligned}
& \int_0^T \int_0^Y \{ p_0 [f_0(t, y, x + \Delta x, u'^*) - f_0(t, y, x, u')] \\
& + \sum_{k=n+1}^{n+q} p_k [f_k(t, y, x + \Delta x, u'^*) - f_k(t, y, x, u')] \\
& + \sum_{k=1}^n p_k \frac{\partial \Delta x_k}{\partial t} + \sum_{k=1}^n \Delta p_k \frac{\partial x_k}{\partial t} + \sum_{k=1}^n \Delta p_k \frac{\partial \Delta x_k}{\partial t} \\
& - [H(t, y, x, x_y, p, u'^*) - H(t, y, x, x_y, p, u')] \\
& - \sum_{k=1}^n \left( \frac{\partial H}{\partial x_k} \Delta x_k + \frac{\partial H}{\partial x_{ky}} \Delta x_{ky} + \frac{\partial H}{\partial p_k} \Delta p_k \right) \} dy dt \\
& - e_1 = 0 \tag{A. 14}
\end{aligned}$$

The derivatives of  $H$  are estimated at the point  $(t, y, x, x_y, p, u)$ . The remainder  $e_1$  is given by

$$\begin{aligned}
e_1 = & \int_0^T \int_0^Y \sum_{k=1}^n \sum_{j=1}^n \left( \frac{\partial^2 H_a}{\partial x_j \partial x_k} \Delta x_j \Delta x_k + \frac{\partial^2 H_a}{\partial x_j \partial p_k} \Delta x_j \Delta p_k \right. \\
& \left. + \frac{\partial^2 H_a}{\partial x_{jy} \partial p_k} \Delta x_{jy} \Delta p_k \right) dy dt
\end{aligned}$$

$$\begin{aligned}
& + \int_0^T \int_0^Y \sum_{j=m'+1}^{m'''} \sum_{k=1}^n \left( \frac{\partial^2 H_b}{\partial x_k \partial u_j} \Delta x_k + \frac{\partial^2 H_b}{\partial x_{ky} \partial u_j} \Delta x_{ky} \right. \\
& \left. + \frac{\partial^2 H_b}{\partial p_k \partial u_j} \Delta p_k \right) \Delta u_j \, dy dt \quad (A. 15)
\end{aligned}$$

The second derivatives of  $H_a$  and  $H_b$  in  $e_1$  are estimated at the point  $(t, y, x + \theta_1 \Delta x, x_y + \theta_2 \Delta x_y, p + \theta_3 \Delta p, u + \Delta u)$  and  $(t, y, x, x_y, p, u + \theta_4 \Delta u)$  respectively.  $\theta_i$ 's are constant vectors with elements having values between 0 and 1.

The application of Green's Theorem gives the equation

$$\begin{aligned}
\int_0^T \int_0^Y \sum_{k=1}^n p_k \frac{\partial \Delta x_k}{\partial t} \, dy dt &= - \int_0^T \int_0^Y \sum_{k=1}^n \Delta x_k \frac{\partial p_k}{\partial t} \, dy dt \\
&+ \int_0^Y \sum_{k=1}^n [p_k(T, Y) \Delta x_k(T, Y) - p_k(0, Y) \Delta x_k(0, Y)] \, dy \\
&+ \sum_{k=1}^n \int_{C_{k-}} p_k \Delta x_k \, dy - \sum_{k=1}^n \int_{C_{k+}} p_k \Delta x_k \, dy \quad (A. 16)
\end{aligned}$$

The initial conditions of  $\Delta x_k$ 's are given by

$$\begin{aligned}
\Delta x_k(0, Y) &= x_{k0}(Y, u^{I*}) - x_{k0}(Y, u^I) \\
&k=1, 2, \dots, n \quad (A. 17)
\end{aligned}$$

Similarly, we have

$$\int_0^T \int_0^Y \sum_{k=1}^n \frac{\partial H}{\partial x_{ky}} \Delta x_{ky} \, dy dt = \int_0^T \int_0^Y \sum_{k=1}^n \Delta x_k v_k \frac{\partial p_k}{\partial y} \, dy dt$$

$$\begin{aligned}
& - \int_0^T \sum_{k=1}^n [p_k(t, Y) v_k(t) \Delta x_k(t, Y) \\
& - p_k(t, 0) v_k(t) \Delta x_k(t, 0)] dt + \sum_{k=1}^n \int_{C_{k-}} p_k v_k \Delta x_k dt \\
& - \sum_{k=1}^n \int_{C_{k+}} p_k v_k \Delta x_k dt \quad (A. 18)
\end{aligned}$$

On the curves  $C_{k-}$  and  $C_{k+}$ , the following equality holds

$$dy = v_k dt \quad k=1, 2, \dots, n \quad (A. 19)$$

The boundary conditions of  $\Delta x_k$ 's are given by

$$\begin{aligned}
\Delta x_k(t, 0) &= x_{k1}(t, u^{B*}) - x_{k1}(t, u^B) \\
&k=1, 2, \dots, n' \quad (A. 20)
\end{aligned}$$

$$\begin{aligned}
\Delta x_k(t, Y) &= x_{k1}(t, u^{B*}) - x_{k1}(t, u^B) \\
&k=n'+1, n'+2, \dots, n \quad (A. 21)
\end{aligned}$$

If we define

$$e_2 = \int_0^T \int_0^Y \sum_{k=1}^n \Delta p_k \frac{\partial \Delta x_k}{\partial t} dy dt \quad (A. 22)$$

we have

$$e_2 = \int_0^T \int_0^Y \sum_{k=1}^n \left[ \sum_{j=m'+1}^{m'''} \frac{\partial^2 H_c}{\partial p_k \partial u_j} \Delta p_k \Delta u_j \right]$$

$$+ \sum_{j=1}^n \left( \frac{\partial^2 H_c}{\partial p_k \partial x_j} \Delta p_k \Delta x_j + \frac{\partial^2 H_c}{\partial p_k \partial x_{jy}} \Delta p_k \Delta x_{jy} \right) dy dt \quad (A. 23)$$

The second derivatives of  $H_c$  in the expression for  $e_2$  are estimated at the point  $(t, y, x + \theta_5 \Delta x, x_y + \theta_6 \Delta x_y, p, u + \theta_7 \Delta u)$ .  $\theta_i$ 's are constant vectors with elements having values between 0 and 1.

Equations (A. 5), (A. 14) and (A. 16) through (A. 22) are combined to form

$$\begin{aligned} \Delta I = & - \int_0^T \left\{ \sum_{k=1}^{n'} [p_k(t, 0) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, 0)}] \Delta x_{k1} \right. \\ & + \sum_{k=n'+1}^n [-p_k(t, Y) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, Y)}] \Delta x_{k1} \Big\} dt \\ & - \int_0^Y \sum_{k=1}^n [p_k(0, y) + \sum_{j=0}^{s''} w'_j \frac{\partial h_j}{\partial x_k(0, y)}] \Delta x_{k0} dy \\ & - \int_0^T \sum_{k=0}^{s'} w_k \Delta g_k dt - \int_0^Y \sum_{k=0}^{s''} w'_k \Delta h_k dy - \int_0^T \int_0^Y \Delta H dy dt \\ & - \int_0^T \int_0^Y \left[ \sum_{k=1}^n \left( \frac{\partial p_k}{\partial t} + v_k \frac{\partial p_k}{\partial y} + \frac{\partial H}{\partial x_k} \right) \Delta x_k \right. \\ & \left. - \sum_{k=1}^n \left( \frac{\partial x_k}{\partial t} - \frac{\partial H}{\partial p_k} \right) \Delta p_k \right] dy dt \end{aligned}$$

$$\begin{aligned}
& - \int_0^T \sum_{k=1}^{n'} \left[ \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, Y)} - p_k(t, Y) v_k(t) \right] \Delta x_k(t, Y) dt \\
& - \int_0^T \sum_{k=n'+1}^n \left[ \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, 0)} \right. \\
& \left. + p_k(t, 0) v_k(t) \right] \Delta x_k(t, 0) dt \\
& - \int_0^Y \sum_{k=1}^n \left[ \sum_{j=0}^{s''} w_j' \frac{\partial h_j}{\partial x_k(T, Y)} - p_k(T, Y) \right] \Delta x_k(T, Y) dy
\end{aligned}$$

$$- e_1 + e_2 - e_3 \quad (\text{A. 24})$$

where

$$\Delta x_{kl} = x_{kl}(t, u^{B*}) - x_{kl}(t, u^B)$$

$$k=1, 2, \dots, n \quad (\text{A. 25})$$

$$\Delta x_{k0} = x_{k0}(Y, u^{I*}) - x_{k0}(Y, u^I)$$

$$k=1, 2, \dots, n \quad (\text{A. 26})$$

$$\Delta g_k = g_k(t, x(t, 0), x(t, Y), u^{B*}, u^{t*})$$

$$- g_k(t, x(t, 0), x(t, Y), u^B, u^t)$$

$$k=0, 1, \dots, s' \quad (\text{A. 27})$$

$$\Delta h_k = h_k(y, x(0, y), x(T, y), u^I, u^Y)$$

$$- h_k(y, x(0, y), x(T, y), u^I, u^Y)$$

$$k=0, 1, \dots, s'' \quad (\text{A. 28})$$

$$\Delta H = H(t, y, x, x_y, p, u^I) - H(t, y, x, x_y, p, u^I) \quad (\text{A. 29})$$

The derivatives of  $g$  and  $h$  are taken at points  $(t, x(t, 0), x(t, y), u^B, u^t)$  and  $(y, x(0, y), x(T, y), u^I, u^Y)$  respectively.

The term  $e_3$  in equation (A. 24) is given by

$$\begin{aligned} e_3 = & \int_0^T \sum_{j=1}^m \sum_{k=0}^{s'} \sum_{i=1}^n w_k \left[ \frac{\partial^2 g_{ka}}{\partial x_i(t, y) \partial u_j} \Delta x_i(t, y) \right. \\ & \left. + \frac{\partial^2 g_{kb}}{\partial x_i(t, 0) \partial u_j} \Delta x_i(t, 0) \right] \Delta u_j dt \\ & + \int_0^T \sum_{k=0}^{s'} \sum_{i=1}^n \sum_{j=1}^n w_k \left[ \frac{\partial^2 g_{kc}}{\partial x_i(t, y) \partial x_j(t, y)} \Delta x_i(t, y) \Delta x_j(t, y) \right. \\ & + \frac{\partial^2 g_{kc}}{\partial x_i(t, y) \partial x_j(t, 0)} \Delta x_i(t, y) \Delta x_j(t, 0) \\ & \left. + \frac{\partial^2 g_{kc}}{\partial x_i(t, 0) \partial x_j(t, 0)} \Delta x_i(t, 0) \Delta x_j(t, 0) \right] dt \end{aligned}$$



$$\begin{aligned}
& + \int_0^Y \sum_{j=m''+1}^r \sum_{k=0}^{s''} \sum_{i=1}^n w'_k \left[ \frac{\partial^2 h_{ka}}{\partial x_i(0,Y) \partial u_j} \Delta x_i(0,Y) \right. \\
& + \left. \frac{\partial^2 h_{kb}}{\partial x_i(T,Y) \partial u_j} \Delta x_i(T,Y) \right] \Delta u_j dy \\
& + \int_0^Y \sum_{k=0}^{s''} \sum_{i=1}^n \sum_{j=1}^n w'_k \left[ \frac{\partial^2 h_{kc}}{\partial x_i(T,Y) \partial x_j(T,Y)} \Delta x_i(T,Y) \Delta x_j(T,Y) \right. \\
& + \frac{\partial^2 h_{kc}}{\partial x_i(T,Y) \partial x_j(0,Y)} \Delta x_i(T,Y) \Delta x_j(0,Y) \\
& + \left. \frac{\partial^2 h_{kc}}{\partial x_i(0,Y) \partial x_j(0,Y)} \Delta x_i(0,Y) \Delta x_j(0,Y) \right] dy \quad (A. 30)
\end{aligned}$$

The second derivatives of  $g_{ka}$ ,  $g_{kb}$  and  $g_{kc}$ ,  $h_{ka}$ ,  $h_{kb}$  and  $h_{kc}$  are estimated at the points  $(t, x(t, 0), x(t, Y), u + \theta_8 \Delta u)$ ,  $(t, x(t, 0), x(t, Y), u + \theta_9 \Delta u)$ ,  $(t, x(t, 0) + \theta_{10} \Delta x(t, 0), x(t, Y) + \theta_{11} \Delta x(t, Y), u + \Delta u)$ ,  $(y, x(0, Y), x(T, Y), u + \theta_{12} \Delta u)$ ,  $(y, x(0, Y), x(T, Y), u + \theta_{13} \Delta u)$  and  $(y, x(0, Y) + \theta_{14} \Delta x(0, Y), x(T, Y) + \theta_{15} \Delta x(T, Y), u + \Delta u)$  respectively. The constant vector  $\theta_i$ 's are again vectors having elements with values between 0 and 1.

Substituting equations (A. 8) through (A. 12) into equation (A. 24), we have

$$\begin{aligned}
\Delta I = & - \int_0^T \left\{ \sum_{k=1}^{n'} \left[ p_k(t,0) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t,0)} \right] \Delta x_{k1} \right. \\
& + \sum_{k=n'+1}^n \left[ -p_k(t,Y) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t,Y)} \right] \Delta x_{k1} \Big\} dt \\
& - \int_0^Y \sum_{k=1}^n \left[ p_k(0,y) + \sum_{j=0}^{s''} w'_j \frac{\partial h_j}{\partial x_k(0,y)} \right] \Delta x_{k0} dy \\
& - \int_0^T \sum_{k=0}^{s'} w_k \Delta g_k dt - \int_0^Y \sum_{k=0}^{s''} w'_k \Delta h_k dy \\
& - \int_0^T \int_0^Y \Delta H dy dt + e
\end{aligned} \tag{A. 31}$$

where

$$e = -e_1 + e_2 - e_3 \tag{A. 32}$$

Because every element of control can be varied independent of other elements, we shall consider the variation of one form of control at a time.

If only the elements belong to subgroup  $u''$  is varied, the equation (A. 31) is reduced to

$$\Delta I = - \int_0^T \int_0^Y \Delta H \, dy \, dt + e \quad (\text{A. 33})$$

Similarly, the variations of the elements of subgroups  $u^t$ ,  $u^B$ ,  $u^Y$ ,  $u^I$ , one group at a time, reduce the increment  $\Delta I$  to equations (A. 34), (A. 35), (A. 36) and (A. 37) respectively.

$$\Delta I = - \int_0^T \Delta H^t \, dt + e \quad (\text{A. 34})$$

$$\Delta I = - \int_0^T \Delta H^B \, dt + e \quad (\text{A. 35})$$

$$\Delta I = - \int_0^Y \Delta H^Y \, dy + e \quad (\text{A. 36})$$

$$\Delta I = - \int_0^Y \Delta H^I \, dy + e \quad (\text{A. 37})$$

The Hamiltonian functions  $H^t$ ,  $H^B$ ,  $H^Y$ ,  $H^I$  are defined as

$$H^t = \sum_{j=0}^{s'} w_j g_j + \int_0^Y H \, dy \quad (\text{A. 38})$$

$$\begin{aligned} H^B = & \sum_{k=1}^{n'} \left[ p_k(t, 0) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, 0)} \right] x_{k1}(t, u^B) \\ & + \sum_{k=n'+1}^n \left[ -p_k(t, Y) v_k(t) + \sum_{j=0}^{s'} w_j \frac{\partial g_j}{\partial x_k(t, Y)} \right] x_{k1}(t, u^B) \end{aligned}$$

$$+ \sum_{j=0}^{s'} w_j g_j \quad (\text{A. 39})$$

$$H^Y = \sum_{j=0}^{s''} w_j^! h_j + \int_0^T H \, dt \quad (\text{A. 40})$$

$$H^I = \sum_{k=1}^n \left[ p_k(0, Y) + \sum_{j=0}^{s''} w_j^! \frac{\partial h_j}{\partial x_k(0, Y)} \right] x_{k0}(Y, u^I)$$

$$+ \sum_{j=0}^{s''} w_j^! h_j \quad (\text{A. 41})$$

X. APPENDIX B: BOUND OF  $|e|$ 

$$\frac{\partial x_i}{\partial t} = -v_i(t) \frac{\partial x_i}{\partial y} + r_i(t, y, x, u')$$

$$i=1,2,\dots,n \quad (B. 1)$$

The characteristic equations for the equations (B. 1) are

$$\frac{dt}{ds} = 1 \quad (B. 2)$$

$$\frac{dy}{ds} = v_i \quad i=1,2,\dots,n \quad (B. 3)$$

$$\frac{dx_i}{ds} = r_i(s, y, x, u') \quad i=1,2,\dots,n \quad (B. 4)$$

$$\frac{dx_{it}}{ds} = -x_{iy} \frac{dv_i}{ds} + \frac{\partial r_i}{\partial s} + x_{it} \frac{\partial r_i}{\partial x_i}$$

$$i=1,2,\dots,n \quad (B. 5)$$

$$\frac{dx_{iy}}{ds} = \frac{\partial r_i}{\partial y} + x_{iy} \frac{\partial r_i}{\partial x_i}$$

$$i=1,2,\dots,n \quad (B. 6)$$

Let  $x+\Delta x$  be the state vector of the system corresponding to the control  $u+\Delta u$ , then, we have

$$\frac{d\Delta x_i}{ds} = r_i(s, y, x+\Delta x, u'+\Delta u') - r_i(s, y, x, u')$$

$$i=1,2,\dots,n \quad (B. 7)$$

Lipschitz conditions for the functions  $r_i$ ,  $x_{i1}$  and  $x_{i0}$  are

$$\begin{aligned} |r_i(s, y, x + \Delta x, u' + \Delta u') - r_i(s, y, x, u')| &\leq M_1 \sum_{j=1}^n |\Delta x_j| \\ &+ M_2 \sum_{j=m'+1}^{m''} |\Delta u_j| \\ i=1,2,\dots,n \end{aligned} \quad (B. 8)$$

$$\begin{aligned} |x_{i1}(s, u^B + \Delta u^B) - x_{i1}(s, u^B)| &\leq M_3 \sum_{j=1}^{m'} |\Delta u_j| \\ i=1,2,\dots,n \end{aligned} \quad (B. 9)$$

$$\begin{aligned} |x_{i0}(y, u^I + \Delta u^I) - x_{i0}(y, u^I)| &\leq M_3 \sum_{j=m'''+1}^r |\Delta u_j| \\ i=1,2,\dots,n \end{aligned} \quad (B. 10)$$

Integrating equations (B. 7), taking the absolute value of both sides and using the Lipschitz conditions, we obtain formulas for  $|\Delta x_i|$ .

$$\begin{aligned} |\Delta x_i| &\leq \int_0^t (M_1 \sum_{j=1}^n |\Delta x_j| + M_2 \sum_{j=m'+1}^{m''} |\Delta u_j|) ds \\ &+ M_3 \left( \sum_{j=1}^{m'} |\Delta u_j| + \sum_{j=m'''+1}^r |\Delta u_j| \right) \\ i=1,2,\dots,n \end{aligned} \quad (B. 11)$$

Summing over all  $i$  and strengthening the inequality, we

get

$$\theta \leq \int_0^t M_1 n \theta \, ds + \int_0^T M_2 n \phi \, ds + M_3 n \phi_1 \quad (\text{B. 12})$$

where

$$\theta = \sum_{j=1}^n |\Delta x_j| \quad (\text{B. 13})$$

$$\phi = \sum_{j=m'+1}^{m''} |\Delta u_j| \quad (\text{B. 14})$$

$$\phi_1 = \sum_{j=1}^{m'} |\Delta u_j| + \sum_{j=m''+1}^r |\Delta u_j| \quad (\text{B. 15})$$

Therefore

$$\theta \leq M_4 \int_0^T \phi \, ds + M_5 \phi_1 \quad (\text{B. 16})$$

where

$$M_4 = M_2 n e^{M_1 n T} \quad (\text{B. 17})$$

$$M_5 = M_3 n e^{M_1 n T} \quad (\text{B. 18})$$

Thus we obtain the bounds of  $|\Delta x_i|$ :

$$|\Delta x_i| \leq M_4 \int_0^T \phi \, ds + M_5 \phi_1 \quad (\text{B. 19})$$

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

Bounds of  $|\Delta x_{iy}|$  and  $|\Delta p_i|$  are obtained in an analogous fashion

$$|\Delta x_{iy}| \leq M_6 \int_0^T \phi \, ds + M_7 \phi_1$$

$$i=1,2,\dots,n \quad (\text{B. 20})$$

$$|\Delta p_i| \leq M_8 \int_0^T \phi \, ds + M_9 \phi_1$$

$$i=1,2,\dots,n \quad (\text{B. 21})$$

Introducing a  $3n$ -dimensional vector  $z$  as

$$z \equiv (x_1, x_2, \dots, x_n, p_1, p_2, \dots, p_n, x_{1y}, x_{2y}, \dots, x_{ny})$$

then, we have

$$|\Delta z_i| \leq M \left( \int_0^T \phi' \, ds + \phi_1 \right)$$

$$i=1,2,\dots,3n \quad (\text{B. 22})$$

where  $M$  is the maximum value among  $M_i$  ( $i=4$  to  $9$ ) and

$$\phi'(t) = \max_{0 \leq y \leq Y} \phi(t, y) \quad (\text{B. 23})$$

Second derivatives  $\frac{\partial^2 H}{\partial x_i \partial x_j}$ ,  $\frac{\partial^2 H}{\partial x_j \partial p_k}$ ,  $\frac{\partial^2 H}{\partial x_{jy} \partial p_k}$ ,  $\frac{\partial^2 H}{\partial p_k \partial u_j}$ ,

$\frac{\partial^2 H}{\partial x_k \partial u_j}$ ,  $\frac{\partial^2 H}{\partial x_{ky} \partial u_j}$ ,  $\frac{\partial^2 g_k}{\partial x_i \partial x_j}$ ,  $\frac{\partial^2 g_k}{\partial x_i \partial u_j}$ ,  $\frac{\partial^2 h_k}{\partial x_i \partial x_j}$ , and  $\frac{\partial^2 h_k}{\partial x_i \partial u_j}$  are



all bounded. If the variations of control  $u^I$  and  $u^Y$  are zero, equations (A. 15), (A. 23), (A. 30) and (A. 32) lead to

$$|e| \leq M_{10} \left( \int_0^T \phi' ds + \phi_1 \right)^2 \quad (\text{B. 24})$$

If the variation of the boundary control  $u^B$  is also zero, the bound on  $|e|$  is reduced to

$$|e| \leq M_{10} \left( \int_0^T \phi' ds \right)^2 \quad (\text{B. 25})$$

We shall consider a function  $\phi'(t)$  which is larger than zero only on the interval  $[T_V, T_V + \Delta t]$  and which is zero at all other times.

$$\phi'(t) > 0 \quad \text{for } T_V \leq t \leq T_V + \Delta t \quad (\text{B. 26})$$

$$\phi'(t) = 0 \quad \text{for } 0 \leq t < T_V \text{ and}$$

$$T_V + \Delta t < t \leq T \quad (\text{B. 27})$$

Using Cauchy-Schwartz inequality

$$\left( \int_a^b fg ds \right)^2 \leq \left( \int_a^b f^2 ds \right) \left( \int_a^b g^2 ds \right) \quad (\text{B. 28})$$

we obtain

$$|e| \leq M_{10} \Delta t \int_{T_V}^{T_V + \Delta t} \phi'^2 ds \quad (\text{B. 29})$$

where  $\phi'$  can have any magnitude as long as the modified control  $u^*$  is still in set  $U$ .

If only the variation of boundary control  $u^B$  is non-zero and also it is non-zero only in the interval  $T_V \leq t \leq T_V + \Delta t$ ,

the bound on  $|e|$  is

$$|e| \leq M_{10} \phi_1^2 \quad (\text{B. 30})$$

## XI. APPENDIX C:

## PROOFS OF THEOREMS 1 TO 4

## A. Proof of Theorem 1

A control vector  $u''(t,y)$  which is a vector function of  $t$  and  $y$  will be considered. Let  $S$  be the region  $[0,T] \times [0,Y]$  and  $\delta \in S$  be the region  $[T_v, T_v + \Delta t] \times [Y_v, Y_v + \Delta y]$ . The area of region  $\delta$  tends to zero as its diagonal  $d$  approaches zero.

Let  $u''(t,y)$  be the optimal control. The new control  $u''^*(t,y)$  modifies the control  $u''(t,y)$  in the following fashion:

$$u''^*(t,y) = \begin{cases} u''(t,y) & \text{for } (t,y) \in R - \delta \\ u_v'' & \text{for } (t,y) \in \delta \end{cases} \quad (C1. 1)$$

$$\Delta u'' = u''^*(t,y) - u''(t,y) \quad (C1. 2)$$

where  $u_v''$  may assume any value in the set  $U$ . The variation of control vector,  $\Delta u''$ , in the region  $R - \delta$  is zero; the variation in the region  $\delta$  can be as large as the set  $U$  would permit.

The minimum of the functional  $I$  is obtained by the optimal control vector  $u''(t,y)$ . Then for any variation  $\Delta u''$ , we have

$$\Delta I \geq 0 \quad (C1. 3)$$

For the optimal control  $u''(t,y)$ , Hamiltonian  $H$  has to attain its absolute maximum with respect to control  $u''$ . In

other words, for any variation of control vector,  $\Delta u$ , we can assert

$$\Delta H \leq 0 \quad (\text{Cl. 4})$$

where

$$\Delta H = H(t, y, x, x_y, p, u^*(t, y)) - H(t, y, x, x_y, p, u''(t, y)) \quad (\text{Cl. 5})$$

This result will be proved by assuming that for a control  $u^*$ ,  $\Delta H > 0$ . This assumption will lead to  $\Delta I < 0$ , a conclusion which contradicts the statement that  $u''(t, y)$  is the optimal control. Thus, it is necessary for  $H$  to attain its maximum with respect to  $u$  at optimal control  $u''(t, y)$ .

Since  $H(t, y, x, x_y, p, u)$  are continuous functions of its arguments; and  $u''(t, y), x(t, y), p(t, y), x_y(t, y)$  are piecewise continuous functions, we can find a region  $\delta$  encircling a point  $(t^*, y^*)$  such that functions  $H(t, y, x, x_y, p, u^*)$  and  $H(t, y, x, x_y, p, u''(t, y))$  are continuous, and consequently, uniformly continuous.

Assume at the point  $(t^*, y^*)$  the maximum condition of  $H$  is not satisfied. If the region  $\delta$  is small enough, due to uniform continuity, it is possible to find a positive number  $\alpha$  such that the following inequality holds for the region  $\delta$ .

$$\Delta H > \alpha \quad (\text{Cl. 6})$$

In case that  $(t^*, y^*)$  is the point of discontinuity for function  $u''(t, y)$ , we chose a region  $\delta$  such that  $(t^*, y^*)$  is one of the boundary points. In the interior of  $\delta$ ,  $u''(t, y)$  is

continuous, on the boundary point of  $\delta$ ,  $u''(t,y)$  takes on a value approached from the inside of the set  $\delta$ .

For the variation of control  $\Delta u''$ , we have

$$\Delta I = - \int_0^T \int_0^Y \Delta H \, dy dt + e \quad (Cl. 7)$$

Substituting the equations (Cl. 6) and (B. 29) into (Cl. 7), we have

$$\Delta I \leq - \int_{Y_V}^{Y_V + \Delta Y} \int_{T_V}^{T_V + \Delta t} \left( \alpha - \frac{M_{10}}{\Delta Y} \Delta t \phi'^2 \right) dt dy \quad (Cl. 8)$$

Since  $\phi'^2$  is bounded, the integrand in (Cl. 8) can be made positive by choosing a sufficiently small  $\Delta t$ . Thus,  $\Delta I < 0$ , which contradicts the condition (Cl. 3). Therefore, for  $u''(t,y)$  to be optimal solution of the process, it is necessary that the Hamiltonian  $H$  assumes its absolute maximal value at  $u''(t,y)$  at almost any time  $t$  and almost any  $y$ .

#### B. Proof of Theorem 2

The proof of Theorem 2 is essentially the same as that of Theorem 1.

Since  $u^t$  replaces  $u''$  as control variable of the system, the interval  $[T_V, T_V + \Delta t]$  replaces the region  $\delta$  where the variation of control variable is not zero; also the Hamiltonian function  $H^t$  replaces  $H$  in the proof. If the Hamiltonian function  $H^t$  increases by more than  $\alpha$  (a positive number) due

to the modification of the optimal control, the increment  $\Delta I$  can be expressed as

$$\Delta I \leq - \int_{T_V}^{T_V + \Delta t} (\alpha - M_{10} \Delta t \phi'^2) dt$$

Since  $\phi'$  is bounded, the increment  $\Delta I$  can be made negative by choosing a sufficiently small  $\Delta t$  and this leads to a contradiction that  $I$  is already at its minimum. Therefore, it is necessary that  $H^t$  assumes its absolute maximal value at  $u^t(t)$  at almost any time  $t$ .

### C. Proof of Theorem 3

Proof of Theorem 3 is again essentially the same as that of Theorem 1.

We replace control  $u$ , region  $\delta$  and Hamiltonian  $H$  with control  $u^B$ , interval  $[T_V, T_V + \Delta t]$  and Hamiltonian  $H^B$  respectively. Then, we assume that the Hamiltonian function  $H^B$  increases by more than  $\alpha$  (a positive number) due to the modification of the optimal control. The combination of equations (A. 35), (B. 30) and the assumption  $\Delta H^B > \alpha$  gives

$$\Delta I \leq - (\alpha \Delta t - M_{10} \phi_1^2) \quad (C3. 1)$$

For any fixed infinitesimal  $\Delta t$ , the right hand side of inequality sign can be made negative by choosing a sufficiently small  $\phi_1$  and this leads to  $\Delta I < 0$ , which contradicts that  $I$  is already at its minimum. Therefore, it is necessary

that the Hamiltonian  $H^B$  assumes its relative maximal value at  $u^B(t)$  at almost any time  $t$ .

#### D. Proof of Theorem 4

For the linear system, we have

$$\frac{\partial^2 H}{\partial x_i \partial x_j} = 0 \quad i=1,2,\dots,n; \quad j=1,2,\dots,n \quad (C4. 1)$$

$$\frac{\partial^2 H}{\partial x_i \partial u_j} = 0 \quad i=1,2,\dots,n; \quad j=1,2,\dots,r \quad (C4. 2)$$

$$\frac{\partial^2 H}{\partial x_{iy} \partial u_j} = 0 \quad i=1,2,\dots,n; \quad j=1,2,\dots,r \quad (C4. 3)$$

$$\frac{\partial^2 H}{\partial p_i \partial x_j} = b_{ij} \quad i=0,1,\dots,n+q; \quad j=1,2,\dots,n \quad (C4. 4)$$

$$\frac{\partial^2 H}{\partial p_i \partial x_{jy}} = -a_i \delta_{ij} \quad i=0,1,\dots,n+q; \quad j=1,2,\dots,n \quad (C4. 5)$$

where  $\delta_{ij}$  is the Kronecker delta.

$$\frac{\partial^2 H}{\partial p_i \partial u_j} = \frac{\partial r_i}{\partial u_j} \quad i=0,1,\dots,n+q; \quad j=1,2,\dots,r \quad (C4. 6)$$

$$\frac{\partial^2 g_k}{\partial x_i \partial x_j} = 0 \quad i,j=1,2,\dots,n; \quad k=0,1,\dots,s' \quad (C4. 7)$$

$$\frac{\partial^2 g_k}{\partial x_i \partial u_j} = 0 \quad \begin{array}{l} i=1,2,\dots,n; \quad j=1,2,\dots,r \\ k=0,1,\dots,s' \end{array} \quad (C4. 8)$$

$$\frac{\partial^2 h_k}{\partial x_i \partial x_j} = 0 \quad i,j=1,2,\dots,n; \quad k=0,1,\dots,s'' \quad (C4. 9)$$

$$\frac{\partial^2 h_k}{\partial x_i \partial u_j} = 0 \quad \begin{array}{l} i=1,2,\dots,n; \quad j=1,2,\dots,r \\ k=0,1,\dots,s'' \end{array} \quad (C4. 10)$$

$$\frac{\partial \Delta p_i}{\partial t} = - a_i \frac{\partial \Delta p_i}{\partial y} - \sum_{j=0}^{n+q} \Delta p_j b_{ji} \quad i=1,2,\dots,n \quad (C4. 11)$$

$$\Delta p_i(T, y) = 0 \quad i=1,2,\dots,n \quad (C4. 12)$$

$$\Delta p_i(t, Y) = 0 \quad i=1,2,\dots,n' \quad (C4. 13)$$

$$\Delta p_i(t, 0) = 0 \quad i=n'+1, n'+2, \dots, n \quad (C4. 14)$$

The solution of (C4. 9) through (C4. 12) gives

$$\Delta p_i(t, y) = 0 \quad i=1,2,\dots,n \quad (C4. 15)$$

Substituting the equations (C4. 1) through (C4. 10) and (C4. 15) into equations (A. 15), (A. 23), (A. 30) and using



the equation (A. 32) we obtain

$$e = 0 \qquad \qquad \qquad (C4. 14)$$

Since the remainder term is zero, the maximum principles stated in Theorems 1,2 and 3 and Corollaries are both necessary and sufficient conditions.

XII. APPENDIX D:  
PROOF OF THEOREM 5

The proof of Theorem 5 is similar to that of Theorem 3. The control function is a piecewise constant function of time.

The total time  $[0, T]$  is subdivided into  $N$  intervals  $[T_{j-1}, T_j]$  ( $j=1, 2, \dots, N$ ), with  $T_0=0$ ,  $T_N=T$ . The  $j$ th interval has length  $\Delta T_j = T_j - T_{j-1}$ .

Let  $u^t(t)$  be the optimal control vector for the system. The control  $u^{t*}(t)$  modifies the control  $u^t(t)$  in the following fashion:

$$u^{t*}(t) = \begin{cases} u^t(t) & 0 \leq t < T_{j-1}; \quad T_j < t \leq T \\ u^t(t) + \Delta u^t & T_{j-1} \leq t \leq T_j \end{cases} \quad (D. 1)$$

For a certain small variation  $\Delta u^t$ , we should have

$$\Delta I \geq 0 \quad (D. 2)$$

Let's assume that for this  $\Delta u^t$ , there is a positive number  $\alpha$ , such that

$$\Delta H_j^{t*} > \alpha \quad (D. 3)$$

then, combination of the equations (IIIG. 2), (A. 34) and (D. 3) gives

$$\Delta I \leq -(\alpha - M_{10} \Delta T_j^2 \phi'^2) \quad (D. 4)$$

Since  $\Delta T_j$  is bounded, the right hand side of the inequality sign in the equation (D. 4) can be made negative by

choosing a sufficiently small  $\Delta n^t$  and this leads to  $\Delta I < 0$ , which contradicts the condition (D. 2). Therefore, the functions  $H_j^t$  have to attain their relative maxima for  $j=1,2,\dots,N$ .

XIII. APPENDIX E:  
RUNGE-KUTTA-GILL SYSTEM

Gill (20) proposes a modification of the standard fourth order Runge-Kutta integration process for a set of first order ordinary differential equations. His modification minimizes the storage problem and reduces the accumulation of the round-off errors.

The set of the differential equations has the form:

$$\frac{dx_i}{dt} = f_i(t, x_1, x_2, \dots, x_n)$$

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

He wants to obtain the value of  $x$  corresponding to  $T + h$ , starting from the point  $t=T$ ,  $x=X$ .

$k_{ij}$ 's,  $x_{ij}$ 's and  $q_{ij}$ 's are sequentially calculated. The order of calculation is as follows:

$$k_{i0} = hf_i(T, x_{10}, x_{20}, \dots, x_{n0})$$

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

$$x_{i1} = x_{i0} + \frac{1}{2}k_{i0} \quad i=1, 2, \dots, n$$

$$q_{i1} = k_{i0} \quad i=1, 2, \dots, n$$

$$k_{i1} = hf_i(T + \frac{h}{2}, x_{11}, x_{21}, \dots, x_{n1})$$

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

$$x_{i2} = x_{i1} + (1 - \sqrt{\frac{T}{2}}) (k_{i1} - q_{i1})$$

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

$$q_{i2} = (2 - \sqrt{2})k_{i1} + (-2 + 3\sqrt{\frac{T}{2}})q_{i1}$$

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

$$k_{i2} = hf_i(T + \frac{h}{2}, x_{12}, x_{22}, \dots, x_{n2})$$

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

$$x_{i3} = x_{i2} + (1 + \sqrt{\frac{T}{2}}) (k_{i2} - q_{i2})$$

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

$$q_{i3} = (2 + \sqrt{2})k_{i2} + (-2 - 3\sqrt{\frac{T}{2}})q_{i2}$$

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

$$k_{i3} = hf_i(T + h, x_{13}, x_{23}, \dots, x_{n3})$$

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

$$x_{i4} = x_{i3} + \frac{1}{6} k_{i3} - \frac{1}{3} q_{i3}$$

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

$x_{i4}$  ( $i=1,2,\dots,n$ ) are the solutions required.