Optimal Temperature Control Policy

For Parallel Reactions in a Batch Reactor

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ABSTRACT : This paper studies the problem of the determination of the optimal temperature progression in a batch reactor in which the following reactions take place:

$$R \xrightarrow{k_1} P \qquad R \xrightarrow{k_2} W$$

where R is an expensive raw material, P is the desired product, and W is a waste byproduct. Both reactions are irreversible, and first-order in species R. The kinetic parameters k_1 and k_2 are related to the temperature thru the familiar Arrhenius relation ($k_i = A_i e^{-E_i/\Re T}$ $i = 1,2$). The objective here is the selection of the temperature control policy $T(t)$, subject to the bounds $T_{min} \leq T(t) \leq T_{max}$, such that the concentration of the desired product P is maximized at the end of a fixed batch operating time. It is not difficult to see that the optimal policy is obtained by setting $T(t) \equiv T_{max}$ when $E_2 \leq E_1$. The solution for the case $E_2 > E_1$, however, is not trivial. This problem has been studied in the literature only numerically, and then only for certain values of the parameters E_i's and A_i's. In this paper, a complete mathematical solution is derived using the classical variational theory. The optimal temperature policy is demonstrated to have a constant initial value of T_{min}, then an increasing nonlinear segment, and finally a constant final value $T(t) = T_{max}$. The solution is obtained for arbitrary values of the parameters E_i's and A_i's. The results obtained using a numerical procedure are in good agreement with the exact solution given here.

1 Problem Description

An important optimization problem that arises in the design of batch reactors is the selection of a controllable variable (such as temperature, catalyst concentration, pH, etc.) such that the amount of the desired product obtained at the end of the operation is maximized. A practically important problem is the determination of the optimal temperature progression in a batch reactor in which the following reactions take place:

$$R \xrightarrow{k_1} P$$

$$R \xrightarrow{k_2} W$$

where R is an expensive raw material, P is the desired product, and W is a waste byproduct. Both reactions are irreversible, and first-order in species R. The rate constants k_1 and k_2 are given by

$$k_i = A_i\, e^{-E_i/\Re T} \quad i = 1, 2 \tag{1}$$

In this equation, $\Re$ represents the universal gas constant, E is the activation energy, and A is the preexponential constant. Material balances on species R and P in the batch reactor are

$$\frac{dC_R}{dt} = -(k_1 + k_2)C_R$$

$$\frac{dC_P}{dt} = k_1 C_R$$

$$C_W(t) = C_R(0) - C_R(t) - C_P(t)$$

$$C_R(0) > 0 \quad \text{and } C_P(0) = C_W(0) = 0$$

where C_R denotes the concentration of component R in the reaction medium. The objective is to determine the temperature policy for the batch reactor which maximizes the amount of P at the end of a fixed batch time of 1-hour. The notation will be simplified by the following definitions:

$$x_1 = C_R/C_R(0)$$

$$x_2 = C_P/C_R(0)$$

$$p = E_2/E_1$$

$$\alpha = A_2/A_1^p$$

so that $k_2 = \alpha k_1^p$.

It is clear that $\alpha > 0$ and $p > 0$, as A_i and E_i are positive constants. Furthermore, one may restrict attention to the case $p > 1$. The reason for this can be seen as follows.

$$\frac{dC_W/dt}{dC_P/dt} = \frac{k_2}{k_1} = \frac{A_2}{A_1} \exp\left[\frac{E_1 - E_2}{\Re T(t)}\right] \tag{2}$$

Three possible cases can be considered:

1. p = 1: From equation 2, it can be seen that

$$p = 1 \quad \Longrightarrow \quad E_2 = E_1 \quad \Longrightarrow \quad \frac{d(k_2/k_1)}{dT} = 0$$

That is, a change in temperature changes the rates of formation of both products proportionally. Since the rate of formation of P (and that of W) increases with temperature, one simply sets $T(t) \equiv T_{max}$ to maximize $C_P(1)$ in this case.

2. $p < 1$: In this case, increasing temperature increases the rate of formation of the desired product P more than that of W:

$$p < 1 \quad \Longrightarrow \quad E_2 < E_1 \quad \Longrightarrow \quad \frac{d(k_2/k_1)}{dT} < 0$$

One again sets $T(t) \equiv T_{max}$ to maximize $C_P(1)$.

3. $p > 1$: This is the case of interest here as there is no trivial solution.

$$p > 1 \quad \Longrightarrow \quad E_2 > E_1 \quad \Longrightarrow \quad \frac{d(k_2/k_1)}{dT} > 0$$

That is, high temperatures favor the faster formation of the undesired component W. Thus one cannot set $T(t) \equiv T_{max}$. Furthermore, one cannot use $T(t) \equiv T_{min}$ either, because both reactions would be slow, and very little of both P and W would be present in the reactor at the end of the fixed batch time.

It will be convenient to use $k_1(T(t))$ as the control function. This is permissible because there is a ono-to-one relationship between k_1 and T (see equation 1). The optimization problem can now be stated as follows:

$$\text{Minimize} \quad -x_2(1) = \int_0^1 (-x_2')\, dt$$

such that the following holds:

$$x_1' = -(u + \alpha u^p)\, x_1 \tag{3}$$

$$x_2' = u\, x_1 \tag{4}$$

$$x_1(0) = 1 \tag{5}$$

$$x_2(0) = 0 \tag{6}$$

$$0 \le x_1(t) \le 1 \tag{7}$$

$$0 \le x_2(t) \le 1 \tag{8}$$

$$U_L \le u(t) \le U_U \tag{9}$$

where $0 < U_L < U_U$ represent the bounds on the control function. It is assumed that $p > 1$, and $\alpha > 0$.

2 Derivation of the Exact Solution

The mathematical treatment of this problem is simplified considerably by the following observation.

Claim : *Let x_1 and x_2 be functions defined and continuously differentiable on $[0,\tau]$. If x_1 and x_2 satisfy the differential equations 3 and 4, the initial conditions 5 and 6, and if $u(t) > 0$, $\alpha > 0$, $p > 0$, then the following holds:*

$$0 \le x_1(t) \le 1 \quad \text{and} \quad 0 \le x_2(t) \le 1 \qquad \forall t \in [0,\tau]$$

Proof: Since $x_1(0) = 1$, it follows that

$$x_1(t) = \exp\Big[-\int_0^t (u(\xi) + \alpha u^p(\xi))\, d\xi \Big]$$

Therefore

$$1 \;\ge\; e^{-(U_L + \alpha U_L^p)t} \;\ge\; x_1(t) \;\ge\; e^{-(U_U + \alpha U_U^p)t} \;>\; 0$$

Thus $0 < x_1(t) \le 1$ for all $t \ge 0$. From Equation 4 it follows that $x_2' > 0$. Since $x_2(0) = 0$, it follows that $x_2(t) \ge 0$. Finally it can be noted that

$$x_1' + x_2' = -\alpha\, u^p(t)\, x_1(t) < 0$$

that is, $x_1 + x_2$ is decreasing. Since $x_1(0) + x_2(0) = 1$, and $x_1(t) > 0$, it follows that $x_2(t) < 1$. □

This fact allows the elimination of the inequality constraints on the state functions $x_i(t)$, as these constraints are satisfied whenever the bounds on $u(t)$ are satisfied (in particular, when $u(t) \ge U_L > 0$).

A solution that satisfies the necessary conditions of optimality will be constructed here by utilizing the following results from variational calculus [3, 4].

Theorem : *If for a domain $D \subseteq \mathrm{R}^{2d}$, $f = f(t, X, Z) \in C^1([0,\tau] \times D)$ and X provides a local extremal value for*

$$F(X) = \int_0^\tau f[t, X(t), X'(t)]\, dt$$

on

$$\mathcal{D} = \{X \in (\widehat{C}^1[0,\tau])^d : \; X(0) = X_0 \,; (X(t), X'(t)) \in D\}$$

then except at its corner points, X is C^1 and satisfies the first and second Euler-Lagrange equations:

$$\frac{d}{dt} f_{X'}(t) = f_X[t, X, X'] \tag{10}$$

$$\frac{d}{dt}(f - X' \cdot f_{X'})(t) = f_t(t) \tag{11}$$

At each corner point c, X meets the Weierstrass-Erdmann conditions:

$$f_{X'}(c^-) = f_{X'}(c^+) \tag{12}$$

$$(f - X' \cdot f_{X'})(c^-) = (f - X' \cdot f_{X'})(c^+) \tag{13}$$

Furthermore, X meets the natural boundary conditions

$$f_{X'}(\tau) = \tilde{0} \tag{14}$$

It may be noted that the optimization problem considered in the theorem is an *unconstrained* one. On a fixed interval $[0, \tau]$, a more typical problem in variational calculus is that of minimizing $F(X)$ on the set $\mathcal{D}$ defined in the theorem, subject to Lagrangian constraints of two forms:

a) Equality constraints symbolized by $E(X, X') = \tilde{0}$

b) Inequality constraints symbolized by $C(X, X') \leq \tilde{0}$

The theorem remains applicable, however, by utilizing the following well-known result from variational theory [4]:

Proposition : *If for some continuous Lagrangian multiplier functions P and M, $(X_*, X_*') \in \mathcal{D}$ minimizes*

$$\tilde{F}(X) = F(X) + \int_0^\tau \Big(P(t) \cdot E[X(t), X'(t)] + M(t) \cdot C[X(t), X'(t)] \Big)\, dt \tag{15}$$

on $\mathcal{D}$, it follows that (X_*, X_*') will minimize $F(X)$ on $\mathcal{D}$ under a) *and* b) *provided that for $t \in [0, \tau]$:*

a') $E(X_*, X_*') = \tilde{0}$, *and*

b') $M \geq \tilde{0}$ *with* $M \cdot C(X_*, X_*') \equiv 0$

except possibly at a finite set of values.

The theorem will be applicable to the problem considered here by noting the following:

1. The control function $u(t)$ is continuous because there cannot be a discontinuity in the temperature within a single reactor. Except at its corner points, one expects the optimal control function to be differentiable. Thus, the above theorem will be used by considering $u(t)$ as $x_3(t)$.

2. $u(0)$ is not specified, hence one may modify the theorem by adding the natural boundary condition:

$$\tilde{f}_{x_3'}(0) = \tilde{f}_{u'}(0) = 0$$

This condition, however, is redundant since u' does not appear anywhere in the problem, and $\tilde{f}_{u'}(t) \equiv 0 \ \ \forall t \in [0, \tau]$.

3. As is described in the proposition, the differential equations and the bounds on $u(t)$ will be handled by augmenting the integrand $f(t, X, X')$ by appropriate Lagrangian multiplier functions to obtain a new integrand $\tilde{f}$.

In this problem there are two equality constraints (the differential equations) and a single inequality constraint:

$$U_L \leq u(t) \leq U_U \qquad \Longleftrightarrow \qquad (u(t) - U_L)(u(t) - U_U) \leq 0$$

Hence, using the notation $P = (\rho_1, \rho_2)$ and $M = (\mu)$, the augmented integrand can be written as

$$\tilde{f} = f + \rho_1(t)\,[x_1' + (u + \alpha u^p)x_1] + \rho_2(t)\,[x_2' - ux_1] + \mu(t)\,(u - U_L)(u - U_U)$$

To apply the theorem, first the following is observed:

$$\tau = 1$$

$$f = -x_2'$$

$$\tilde{f}_{x_1'} = \rho_1$$

$$\tilde{f}_{x_2'} = \rho_2 - 1$$

$$\tilde{f}_{x_3'} = \tilde{f}_{u'} \equiv 0$$

$$\tilde{f}_{x_1} = \rho_1(u + \alpha u^p) - \rho_2 u$$

$$\tilde{f}_{x_2} = 0$$

$$\tilde{f}_u = \rho_1(1 + p\alpha u^{p-1})x_1 - \rho_2 x_1 + \mu(2u - U_L - U_U)$$

$$\tilde{f}_t \equiv 0$$

Before writing the necessary conditions in their entirety, considerable simplification in notation can be realized by applying the second component of the first Euler-Lagrange equations (Equation 10):

$$\frac{d(\rho_2 - 1)}{dt} = 0 \qquad \Longrightarrow \qquad \rho_2 = \text{ constant}$$

Furthermore, from the second component of the natural boundary conditions (Equation 14):

$$\tilde{f}_{x_2'}(1) = -1 + \rho_2(1) = 0 \qquad \Longrightarrow \qquad \rho_2(1) = 1$$

hence

$$\rho_2(t) \equiv 1 \qquad \forall t \in [0,1] \tag{16}$$

Using Equation 16 together with the other conditions given by the theorem above, the following set of conditions is obtained.

From Equation 10:

$$\rho_1' = \rho_1(u + \alpha u^p) - u \tag{17}$$

$$0 = x_1[\rho_1(1 + p\alpha u^{p-1}) - 1] + \mu(2u - U_U - U_L) \tag{18}$$

From Equation 11:

$$ux_1[\rho_1(1 + \alpha u^{p-1}) - 1] = \text{ constant } = C \tag{19}$$

From Equation 14:

$$\rho_1(1) = 0 \tag{20}$$

From the Proposition:

$$\mu(t)[u(t) - U_L][u(t) - U_U] \equiv 0 \tag{21}$$

The corner conditions (Equations 12 and 13) simply require that ρ_1, ρ_2 and the quantity in Equation 18 be continuous. The last two are identically equal to 1 and 0 respectively, and the continuity requirement is trivially satisfied. The continuity of ρ_1 (as well as that of μ) is already required in the statement of the proposition, and will be utilized.

Thus the problem is one of finding continuous functions ρ_1, μ, u, and continuously differentiable functions x_1, x_2 such that the differential equations 3,4; the initial conditions 5,6; and the set of optimality conditions expressed by Equations 17 thru 21 are all satisfied. In what follows, the subscript on ρ_1 will be dropped for convenience, as it is already known that $\rho_2 \equiv 1$ on the entire interval [0,1].

The solution strategy used here basically consists of assuming a certain form for the optimal control $u(t)$, and checking if the assumed control policy satisfies the necessary conditions for optimality. The following control function is assumed [1]

$$u(t) = \begin{cases} U_L & \text{on } [0, \tau_1] \\ U_U & \text{on } [\tau_2, 1] \end{cases} \tag{22}$$

The locations of the corner points τ_1, τ_2 and the control policy on (τ_1, τ_2) will be uniquely determined from the necessary conditions.

Left Subinterval $[0, \tau_1]$

The following is obtained from the differential equations 3, 4 and the necessary conditions 17, 18:

$$x_1(t) = e^{-(U_L + \alpha U_L^p)t}$$

[1] That the temperature should be increasing has been argued qualitatively in [1].

$$x_2(t) = \frac{1 - x_1(t)}{1 + \alpha U_L^{p-1}}$$

$$\rho(t) = \frac{\rho(0) - x_2(t)}{x_1(t)}$$

$$\mu(t) = \frac{1}{U_U - U_L}\, x_1(t)\, [\, 1 - \rho(t)\,(1 + p\alpha U_L^{p-1})\,]$$

The only unknown here is $\rho(0)$, and will be determined from the natural boundary condition $\rho(1) = 0$ subsequently.

Middle Subinterval (τ_1, τ_2)

Since $U_L < u(t) < U_U$ on (τ_1, τ_2), it follows from Equation 21 that

$$\mu(t) \equiv 0$$

Substituting this into Equation 18, one can obtain the following relation between ρ and u :

$$\rho(t) = \frac{1}{1 + p\,\alpha\, u^{p-1}(t)}$$

Using Equation 19, x_1 is determined directly:

$$x_1(t) = \frac{C}{\rho(t)\,[\, u(t) + \alpha\, u^p(t)\,] - u(t)} = \frac{C}{\alpha(1-p)}\, \frac{1}{\rho(t)\, u^p(t)} \tag{23}$$

The constant C can be expressed in terms of $\rho(0)$ by evaluating Equation 19 at $t = 0$:

$$C = U_L\, [\rho(0)\,(1 + \alpha\, U_L^{p-1}) - 1\,] \tag{24}$$

Next, x_2 will be determined in terms of ρ and x_1. To this end, first note that Equation 19 can be written as

$$C = \rho'(t)\, x_1(t) = -\rho\, x_1' - x_2'$$

from this it follows that

$$x_2'(t) = -\frac{d(\rho\, x_1)}{dt}$$

This equation is valid on the entire interval $[0, 1]$. Thus, given $\rho(t)$ and $x_1(t)$, and the appropriate boundary condition, $x_2(t)$ can be obtained from this equation directly. Here, one gets

$$x_2(t) = x_2(\tau_1) \;+\; [\, \rho(\tau_1) x_1(\tau_1) - \rho(t)\, x_1(t)\,]$$

Given τ_1, the values $x_1(\tau_1)$ and $x_2(\tau_1)$ are fixed by the expressions for x_1, x_2 on $[0, \tau_1]$. If, in addition, $\rho(0)$ is fixed, one can similarly obtain $\rho(\tau_1)$.

The difficult part of this problem is the determination of the control function $u(t)$ on (τ_1, τ_2). Once $u(t)$ is determined, x_1, x_2, and ρ will all be fixed by the expressions derived

above. $u(t)$ will be determined here by solving an ordinary differential equation. To this end, first Equation 19 gives [2]

$$\rho x_1 = \frac{C}{\alpha(1-p)}\, u^{-p}(t) \quad \Longrightarrow \quad \frac{d(\rho\, x_1)}{dt} = -\frac{pC}{\alpha(1-p)}\frac{u'}{u^{p+1}}$$

combining this with $ux_1 = x_2' = -d(\rho x_1)/dt$, and Equation 23, one obtains

$$\frac{du}{dt} = u^2(1 + p\alpha u^{p-1})p^{-1} \tag{25}$$

with the boundary conditions

$$u(\tau_1) = U_L \quad \text{and} \quad u(\tau_2) = U_U$$

If this differential equation can be integrated analytically, all that remains is carrying out a few details. In particular, it will be shown that τ_1 and τ_2 are fixed once U_L, U_U, α, and p are given. Combining $\rho(1) = 0$ with some continuity conditions, one can then obtain $\rho(0)$.

Right Subinterval $[\tau_2, 1]$

Here $u(t) = U_U$, and by direct integration of Equations 3 and 17, the following can be obtained:

$$x_1(t) = x_1(\tau_2)\, e^{-(U_U + \alpha U_U^p)(t-\tau_2)}$$

$$\rho(t) = \frac{1 - e^{(U_U + \alpha U_U^p)(t-1)}}{1 + \alpha U_U^{p-1}}$$

Having fixed $u(t), \rho(t)$, and $x_1(t)$, Equation 18 can be solved for $\mu(t)$, and $x_2(t)$ can be obtained from Equation 4.

Continuity of ρ at τ_2 will first be used to find τ_2:

$$\rho(\tau_2^-) = \rho(\tau_2^+) \quad \Longrightarrow \quad \frac{1}{1 + p\alpha U_U^{p-1}} = \frac{1 - e^{(U_U + \alpha U_U^p)(\tau_2 - 1)}}{1 + \alpha U_U^{p-1}}$$

After some algebraic manipulation, the following is obtained:

$$\tau_2 = 1 + \frac{1}{U_U + \alpha U_U^p} \ln\left[\frac{\alpha(p-1)U_U^{p-1}}{1 + p\alpha U_U^{p-1}}\right] \tag{26}$$

It should be remembered that all these results were derived assuming $p > 1$. From Equation 26, it is clear that the second corner point τ_2 depends only on α, p, and U_U, and not on U_L. Furthermore, $\tau_2 < 1$ for all $p > 1, \alpha > 0, U_U > 0$. This indicates the existence of a corner at τ_2 and a constant segment $u = U_U$ on $[\tau_2, 1]$ whatever the values of these parameters.

[2] Differentiability of $u(t)$ on (τ_1, τ_2) is assumed here.

The constant $\rho(0)$ can be found in terms of τ_1 by combining Equation 24, with the following expression which is obtained by evaluating Equation 19 at τ_1^+:

$$C = x_1(\tau_1)\frac{\alpha U_L^p(1-p)}{1+p\alpha U_L^{p-1}}$$

From this together with Equation 24, it follows that

$$\rho(0) = \frac{1}{1+\alpha U_L^{p-1}}\left[1+\frac{\alpha U_L^{p-1}(1-p)x_1(\tau_1)}{1+p\alpha U_L^{p-1}}\right]$$

where

$$x_1(\tau_1) = e^{-(U_L+\alpha U_L^p)\tau_1}$$

Thus, given τ_1, one can determine $\rho(0)$.

EXAMPLES

- $p = 2$: Ray [2] suggested the use of the numerical values

$$p = 2 \qquad \alpha = \frac{1}{2}$$

For these particular values, it is possible to integrate Equation 25 to obtain an analytical expression giving $u(t)$ on (τ_1, τ_2):

$$\frac{2}{U_L} - \frac{2}{u(t)} + 2\ln\left[\frac{(1+u(t))U_L}{u(t)(1+U_L)}\right] = t - \tau_1$$

Furthermore, since $u(\tau_2) = U_U$, this equation allows the determination of τ_1:

$$\tau_1 = \tau_2 - \frac{2}{U_L} + \frac{2}{U_U} - 2\ln\left[\frac{(1+U_U)U_L}{U_U(1+U_L)}\right]$$

Thus, given U_U and U_L, one can obtain τ_2 from Equation 26, and then use this value here to obtain τ_1. If U_L is small enough, one will have $\tau_1 < 0$. In this case, all the equations obtained above remain valid simply by setting $\tau_1 = 0$, and replacing U_L by $u(0)$ (for example, in Eqn. 24). On the other hand, for all $U_U > U_L > 0$, one has $0 < \tau_2 < 1$.

The optimal control function obtained with $U_U = 2.0$ and $U_L = 1.0$ is shown in Figure 1. Also shown in this figure are the optimal control functions for two other values of α. These were obtained using the following general expression for $p = 2$:

$$\frac{2}{U_L} - \frac{2}{u(t)} + 4\alpha\ln\left[\frac{(1+2\alpha u(t))U_L}{u(t)(1+2\alpha U_L)}\right] = t - \tau_1 \qquad (27)$$

- $p = 3.0$ The optimal control policy for $p = 3.0$ can also be obtained by integrating Equation 25 analytically:

$$\frac{3}{U_L} - \frac{3}{u(t)} + 3\sqrt{3\alpha}\left[\text{arctg}\left(\sqrt{3\alpha}\,U_L\right) - \text{arctg}\left(\sqrt{3\alpha}\,u(t)\right)\right] = t - \tau_1 \qquad (28)$$

  The optimal control functions for three values of α are shown in Figure 2.

- Arbitrary values of p : For other values of p and α, the optimal control function was determined by numerically integrating Equation 25. The strategy used here is as follows. Equation 25 is integrated backwards starting from $t = \tau_2$ and $u(\tau_2) = U_U$. Integration is continued until $u(t) = U_L$. The value of t at that point is accepted as τ_1.

  The results obtained in this manner for several values of p for fixed α are illustrated in Figure 3. The effect of α on the optimal control policy (for fixed p) can be seen in Figures 1 and 2.

  The optimal control policies for $p = 2.0$ and several values of α were also obtained using a numerical procedure [5], and the numerical results obtained are in excellent agreement with the exact solution (i.e. Equation 27) obtained here.

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References

[1] Levenspiel, O. , *Chemical Reaction Engineering*, John Wiley & Sons, New York (1972).

[2] Ray, W.H., *Advanced Process Control*, McGraw-Hill (1981).

[3] Troutman, J.L., *Variational Calculus with Elementary Convexity*, Springer-Verlag, New York (1983).

[4] Troutman, J.L., *Optimal Control with Elementary Convexity*, Supplement to [3], Syracuse University, New York (1986).

[5] Akgiray, O., *Process Design Using Optimal Control Methodology*, Ph. D. Thesis, Syracuse University (1990).

FIGURE 1

Optimal Control Function (p = 2.0)

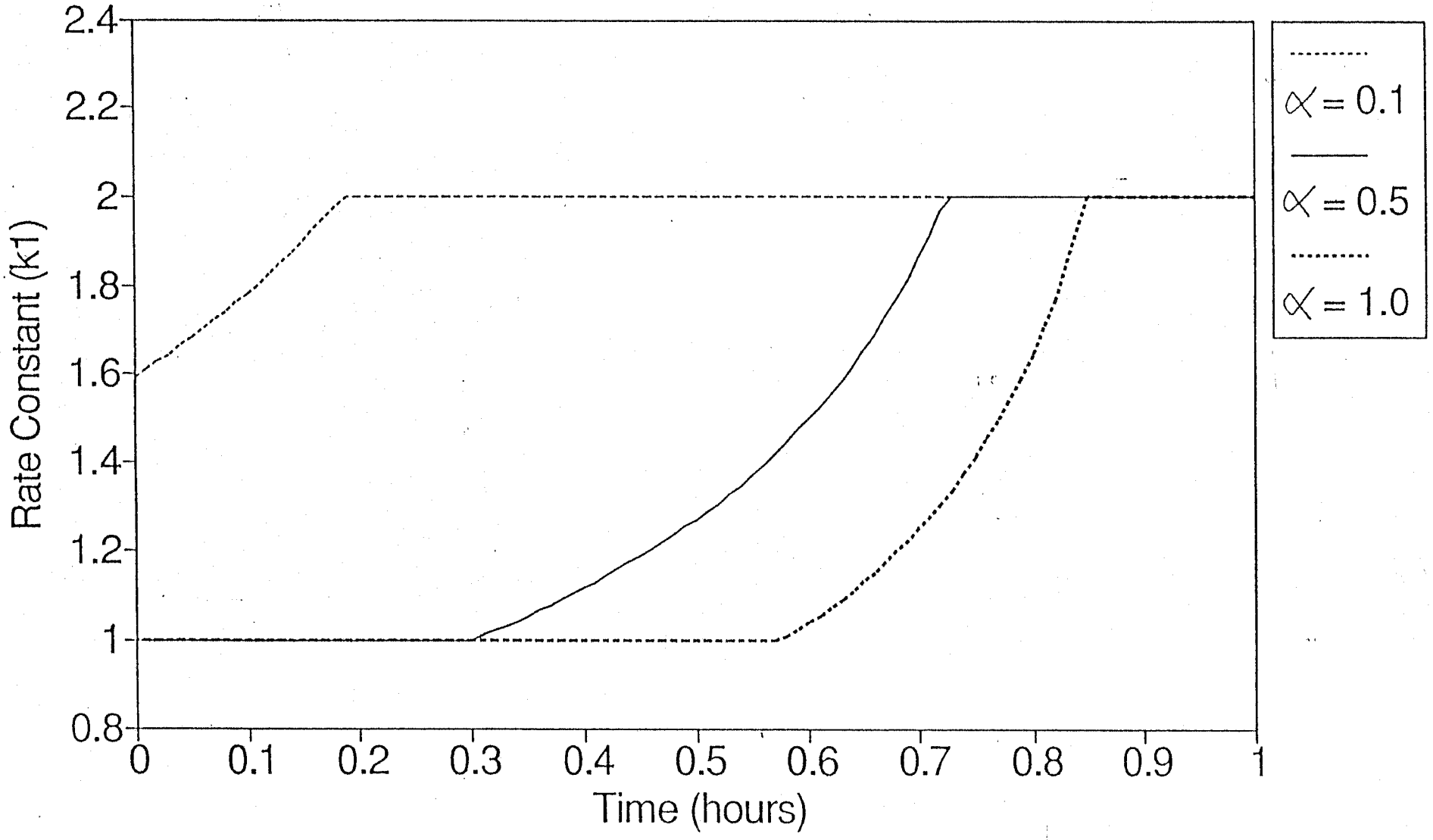


FIGURE 2

Optimal Control Function (p = 3.0)

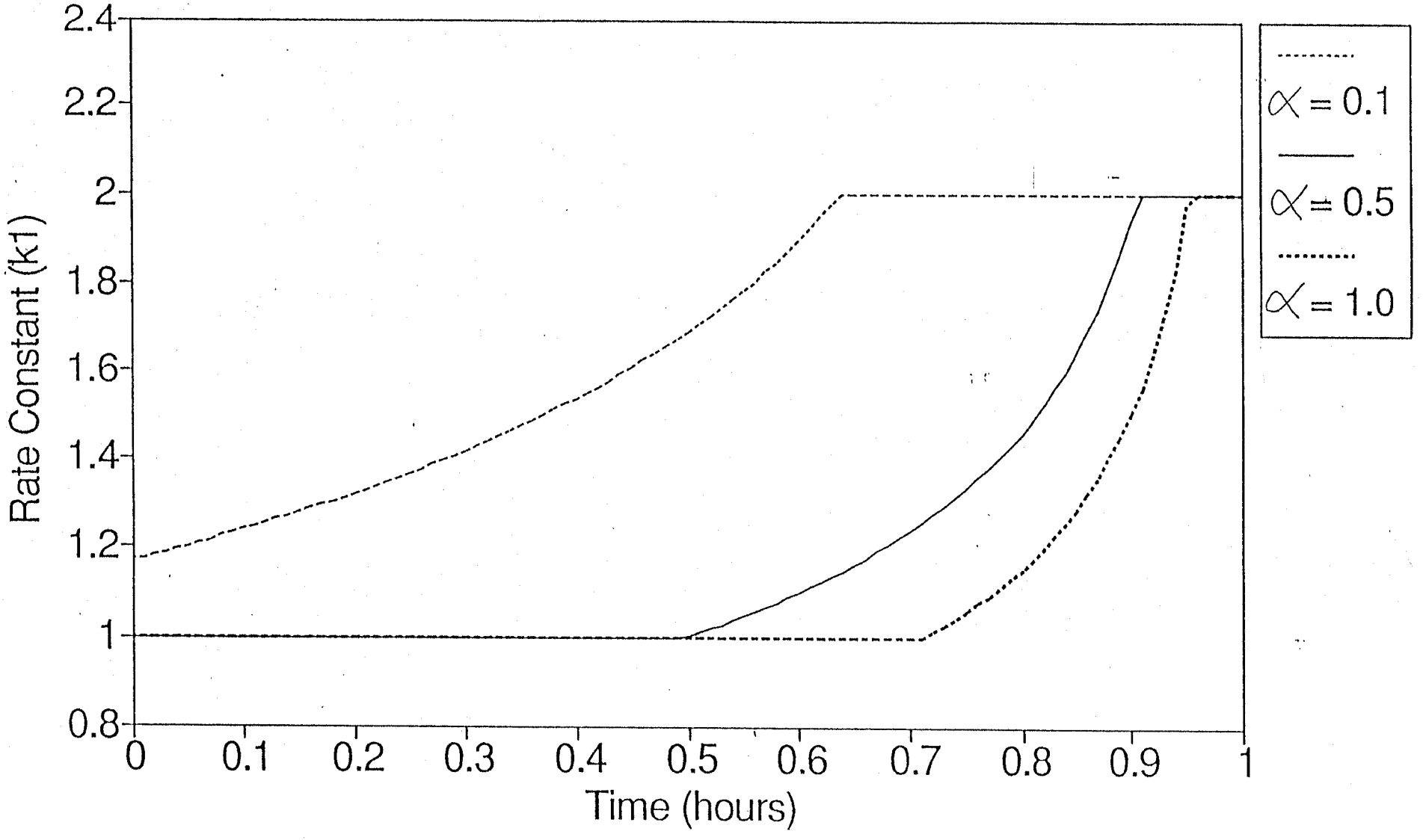


FIGURE 3

Optimal Control Function ($\propto = 0.5$)

