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DEVELOPMENT OF NONSINGULAR OPTIMIZATION ALGORITHM AND ITS APPLICATION TO CHEMICAL ENGINEERING SYSTEMS

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Abstract – A nonsingular transformation algorithm which converts a singular control problem to nonsingular one was developed to calculate optimal control profiles. Several chemical engineering problems for applying a nonsingular transformation algorithm were presented and optimal profiles were calculated in this paper. The singular control algorithm and nonsingular transformation algorithm were compared. The efficiency of the transformation algorithm was displayed in this article. The switching points from bang-bang to singular control were calculated by imposed initial conditions and control variable constraints.

Key words : Optimization, Singular Control, Nonsingular Transformation

INTRODUCTION

Various numerical methods [Fraser-Andrews, 1989; Maurer, 1976; Modak et al., 1989; Seinfeld and Lapidus, 1968; Soliman and Ray, 1972] for calculating singular optimal profiles have been developed. Since the optimal singular profiles are a function of state and adjoint variables, and the initial conditions of state variables and final conditions of adjoint variables are known, some researchers [Maurer, 1976; Oberle, 1979] have used a multiple shooting method to solve this two-point boundary problem. The computational difficulties related to a singular control problem are often serious in calculating optimal control profiles. Although the gradient method [Ko and Stevens, 1971] was sometimes applied to a singular control system, acceptable convergence was not obtained. Some researchers [Ko and Stevens, 1971; Soliman and Ray, 1972] have produced suboptimal but not actual optimal solutions.

Transformation methods were developed to circumvent the difficulty in solving the singular control problem. Kelley [1966] developed a transformation method which decreases the order of the system and converts the singular problem to a nonsingular control problem. The first order partial differential equations have to be solved to use Kelley's transformation. Therefore, this method is rarely used for the optimal control solution. Soliman and Ray [1972] used transformation of the control variables, $\delta u = \delta u_1 + \delta u_2$, proposed by Goh [1966], to calculate the optimal control profile. They compared solutions of each method. However, they could not produce actual optimal control profiles using their transformation technique. Modak et al. [1989] proposed unidirectional computational algorithms for the calculation of singular control profiles of single cell protein production [Chen et al., 1977; Dibiasio et al., 1981] with switching time estimation.

The basic idea of this paper is to convert a singular control problem to a nonsingular one by using transformed control variables with gradient and initial condition constraints. If the initial conditions of the system are not at the singular state, bang-bang control actions are imposed to make the system in a singular state quickly and efficiently. Optimal trajectories for a system that has the bang-bang and singular control regions should have the maximal time for the singular control period.

In this article, the optimal control profiles of the chemical engineering system are calculated with a developed nonsingular optimization approach.

SINGULAR CONTROL

Singular control problems are formulated when the control variables appear linearly in the system equations. If we calculate the Hamiltonian of this system, it appears as a linear function of control variables, and the optimal control profiles are very difficult to calculate for higher order and complex systems. The systems are given as the following differential equations :

$$\dot{\mathbf{x}} = \mathbf{f}(\mathbf{x}) + \mathbf{b}F \tag{1}$$

$$\mathbf{x}(t_0) = \mathbf{x}_0 \tag{2}$$

where **f** is a nonlinear function of a state vector, **b** is a constant vector, F is a control variable, and $\mathbf{x}_0$ is the initial condition of the state vector.

The objective of the optimal problem is to calculate the optimal control profile, F(t), which minimizes a performance index that is a function of state variables. Constraints are typically imposed on control variables :

$$\underset{F}{\text{Maximize}}\,[PI = G(t_f, \mathbf{x}(t_f))] \tag{3}$$

$$F_{min} \le F \le F_{max} \tag{4}$$

In the above equations, PI represents the performance index, G is the functional form of the performance index, $\mathbf{x}(t_f)$ is the

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state vector **x** at the final time t_f, F_{min} is minimum control allowed, F_{max} is the maximum control allowed, and t is the elapsed fermentation time. This performance index may be a function of the final time, which may either be fixed *a priori* or left unspecified. This problem is a standard calculus of variations problem. This system is considered a singular control problem because the Hamiltonian is a linear function of the control variable, F. The optimal control variable profile can be determined by the minimum principles of Pontryagin [1962].

MINIMUM PRINCIPLES OF PONTRYAGIN

The control, which minimizes the Hamiltonian, minimizes the performance index. The Hamiltonian is a scalar function of the state and adjoint variables. It has a constant value during optimal operations and is represented by multiplication of adjoint variables and state equations as defined by the following equations:

$$H = \lambda^T[\mathbf{f}(\mathbf{x}) + \mathbf{b}F] = \psi(\lambda, \mathbf{x}) + \phi(\lambda)F \tag{5}$$

where

$$\psi = \lambda^T \mathbf{f} \tag{6}$$

$$\phi = \lambda^T \mathbf{b} \tag{7}$$

where the adjoint vector, $\lambda(t)$, must satisfy the following ordinary differential equations:

$$\lambda = -\frac{\partial H}{\partial \mathbf{x}} = -\left(\frac{\partial \mathbf{f}}{\partial \mathbf{x}}\right)^T \lambda = -\mathbf{f}_x^T \lambda \tag{8}$$

and the transversality conditions are defined below.

$$\lambda(t_f) = \frac{\partial(PI)}{\partial \mathbf{x}(t_f)} \tag{9}$$

The minimum principle is to find the control trajectory which makes the Hamiltonian constant. Time derivatives of the Hamiltonian are represented by the following equation and are equal to zero. Due to the relation between state variables and adjoint variables, the first two parts cancel.

$$\frac{dH}{dt} = \frac{\partial H}{\partial X}\frac{\partial X}{\partial t} + \frac{\partial H}{\partial \lambda}\frac{\partial \lambda}{\partial t} + \frac{\partial H}{\partial F}\frac{\partial F}{\partial t} + \frac{\partial H}{\partial t} = \frac{\partial H}{\partial F}\frac{\partial F}{\partial t} = 0 \tag{10}$$

Since the Hamiltonian is linear in the control variable, F(t), the feeding policy depends on the coefficient, ϕ, which is called the switching function. When ϕ is positive, the optimal control is F_{max}, and the optimal control is F_{min} when ϕ is negative. If ϕ is identically zero over a finite time interval, $t_i \leq t \leq t_j$, then the maximum principle cannot specify the optimal control during the interval. This is called the singular interval and the optimal feed rate is called the singular feed rate, $F_s(t)$. The trajectory followed by the state variables during the singular interval is called the singular arc. The optimal feeding policy is defined below:

$$F(t) = \begin{Bmatrix} F_{min} & \phi > 0 \\ F_s(t) & \phi = 0 \\ F_{max} & \phi < 0 \end{Bmatrix} \tag{11}$$

The optimal feed rate sequence is a combination of bang-bang and singular intervals.

1. Singular Optimal Control Strategy

During the singular interval, the minimum principle cannot provide sufficient information to determine the singular feed rate and we have to use singular control theory. Since the switching function, ϕ, is identically zero during the singular interval, all of its time derivatives must also be zero:

$$\frac{d^k\phi}{dt^k} = \phi^{(k)} = 0 \quad k = 1, 2, \cdots, \infty \tag{12}$$

where $\phi^{(k)}$ represents the k-th order time derivative of ϕ. A general expression for the singular feed rate is obtained by differentiating twice the switching function, ϕ, so that F appears linearly. The first order dimensionless time derivative of ϕ yields:

$$\dot{\phi} = \dot{\lambda}^T \mathbf{b} = -\lambda^T \mathbf{a}_x \mathbf{b} = \lambda^T \mathbf{c} = 0. \tag{13}$$

The switching function is once again differentiated:

$$\ddot{\phi} = \dot{\lambda}^T \mathbf{c} + \lambda^T \mathbf{c}_x \dot{\mathbf{x}} = -\lambda^T \mathbf{f}_x \mathbf{b} + \lambda^T \mathbf{c}_x(\mathbf{f} + \mathbf{b}F_s) = 0 \tag{14}$$

which can be rearranged to obtain the general expression for $F_s(t)$:

$$F_{min} \leq F_s(t) = \frac{\lambda^T(\mathbf{f}_x \mathbf{c} - \mathbf{c}_x \mathbf{f})}{\lambda^T \mathbf{c}_x \mathbf{b}} \leq F_{max} \tag{15}$$

which is a nonlinear function of state and adjoint variables where the subscript s denotes evaluation during the singular period. Without elimination of λ from this equation, obtaining the optimal feed profile requires solution of this two-point boundary value problem, in which the initial condition of the state variables and the final conditions of the adjoint variables are known. Kelley et al. [1989] showed that the following (Generalized Legendre-Clebsch) condition is necessary for the optimality of a singular arc.

$$(-1)^q \frac{\partial}{\partial F}\left[\frac{d^{2q}}{dt^{2q}} H_F(\mathbf{x}, \lambda, t)\right] \geq 0 \tag{16}$$

where, q denotes the order of singularity.

DEVELOPMENT OF NONSINGULAR TRANSFORMATION ALGORITHM

For higher order systems having complex dynamic models, it is very difficult to derive the explicit or simple functional expression for the singular control profile. Several mathematical and numerical approaches have been adopted to solve a singular system by using minimum principles. Conjugate gradient methods have been used for the entire reaction time; however, they cannot determine the exact switching points.

Some trials had been done to transform the original singular control problem to a simple form of a nonsingular optimization problem. Kelley [1966] proposed a transformation method which converts a singular control system to a nonsingular control one by decreasing the order of the system. This method needs a first order partial differential equation solving procedure and is difficult to implement for complex nonlinear differential equations. Other researchers have used different transformation methods and modifications to calculate suboptimal control profiles.

A modified nonsingular control strategy was developed to determine optimal control profiles by transforming control variables and their constraints.

N-th order fed-batch fermentation systems are described by the following differential equations.

$$\dot{x}_i = f_{oi}(x_i, \ldots\ldots, x_n, t), \qquad i = 1, \ldots, n-2 \tag{17}$$

$$\dot{x}_{n-1} = F \tag{18}$$

$$\dot{x}_n = f_{oi}(x, \ldots\ldots, x_n, t) + S_f F, \qquad i = 1, \ldots, n$$

$$F_{min} \le F(t) \le F_{max} \tag{19}$$

$$\underset{F}{\text{Minimize}}[PI = G(t_f, \mathbf{x}(t_f))] \tag{20}$$

where, F is the control variable (feed rate) and PI is the performance index of the system to be minimized. We introduce transformation variables (z_i) to change the singular control system to a nonsingular control system and define a new control variable.

$$z_i = x_i, \qquad i = 1, 2, \ldots., n-1. \tag{21}$$

$$z_n = x_n/x_{n-1} \quad \text{[New control variable]} \tag{22}$$

Since x_n and x_{n-1} are functions of the control variable F, and F is limited by F_{min} and F_{max}, the time derivative of the new control variable is also constrained.

$$z_n^{min} \le z_n \le z_n^{max} \tag{23}$$

where, z_n^{min} and z_n^{max} are calculated by state variables and limitation of control variables, F_{min} and F_{max}.

The time derivative of x_n is replaced by a function of z_n and z_{n-1}.

$$\dot{x}_n = f_{on} + f_{1n}F = z_n F + \dot{z}_n z_{n-1} \tag{24}$$

The original control variable F is represented by a function of the modified control variable z_n and its time derivative.

$$F = \frac{\dot{z}_n z_{n-1} - f_{on}}{f_{1n} - z_n} = g(\mathbf{z}, \dot{z}_n) \tag{25}$$

where, z_n is calculated by the transformed control variable z_n and is a function of time. Rewriting system equations by using the transformed variables given:

$$\dot{z}_n = f_{oi}(z_i, \ldots\ldots, z_n, t), \quad i = 1, \ldots, n-2 \tag{26}$$

$$\dot{z}_p = f_{o,n-1}(z_i, \ldots\ldots, z_n, t),$$

$$z_{n-1} = \frac{1}{S_f - z_n}\{z_p + (S_f - z_n(0))z_{n-1}(0)\} \tag{27}$$

with constraint by the final condition of volume.

$$g = \frac{1}{S_f - z_z}\{z_p + (S_f - z_n(0))\, z_{n-1}(0)\} - x_{n-1,f} \le 0 \tag{28}$$

The Hamiltonian of this system is represented as a function of state and adjoint variables.

$$H = \lambda^T \dot{z} + \alpha g = h + \alpha g \tag{29}$$

$$\text{where } \alpha \begin{cases} \ge 0 & \text{when } g = 0 \\ = 0 & \text{when } g < 0 \end{cases}$$

The gradient of substrate concentration is a function of time and determined by substrate concentration profile. The adjoint equations must satisfy the following ordinary differential equations:

$$-\dot{\lambda} = \frac{\partial H}{\partial X} = \begin{cases} \dfrac{\partial h}{\partial X} - \alpha \dfrac{\partial g}{\partial X} & g = 0 \\[2ex] \dfrac{\partial h}{\partial X} & g < 0 \end{cases} \tag{30}$$

Since the Hamiltonian is a nonlinear function of S, a control vector iteration technique can therefore be used to solve this problem. The derivative of the Hamiltonian with respect to substrate concentration is:

$$\frac{\partial H}{\partial z_n} = \frac{\partial h}{\partial z_n} + \alpha \frac{\partial g}{\partial z_n} = 0 \tag{31}$$

For $g < 0$, $\alpha = 0$ and Eq. (31) determines the optimal control profile S^*. For $g=0$, Eqs. (26 and 31) determine the S^* and α which is needed in Eq. (30).

From the derivation, we can prove that optimal profiles of singular control variables in a singular control system are the same as nonsingular optimal profiles of transformed control variables. Therefore, the calculation of optimal profiles using a nonsingular technique can be applied for higher order and complex dynamic systems.

NUMERICAL PROCEDURES

1. Guess the initial optimal control profiles (this automatically determines its derivative).
2. Calculate the control variable constraints (the initial substrate concentration and the derivatives of the substrate concentration at the maximum and minimum feed rates).
3. Calculate the state variables by forward integration of Eq. (1).
4. Calculate the adjoint variables by backward integration of Eq. (8).
5. Calculate the derivatives of the Hamiltonian from Eq. (5).
6. Update the substrate concentration profile using the following control vector iteration with the substrate concentration constraint [Eq. (28)].

$$z_n^{i+1}(t) = z_n^i(t) - \varepsilon \left[\frac{dH}{dz_n}\right]^i \tag{32}$$

7. Iterate until the increase in the performance index is less than a specified tolerance (Steps 3 through 6).
8. There may be one or more switching time(s) if the gradient of the control variable is greater or less than the maximum or minimum control allowed. In this case, set the control action (s), pick the switching time(s), and calculate the optimal substrate concentration profile using Steps 1 through 7.
9. Compare the performance index for different switching time(s), if applicable, and select the best performance index, switching time(s), and optimal substrate profiles.

1. Example I. Exothermic Chemical Reaction in PFR

The first example was adopted from Ko and Stevens [1971a, b]. The first-order reversible, exothermic chemical reaction taking place in a plug-flow tubular reactor is considered the model

Table 1. The parameters which were used for this system

Parameter	Value
k_{10}	175360
k_{20}	2.4885×10^{10}
E_1	−11374
E_2	−22748
J	300
T_c	290
l	5
X_{A0}	0.27
T_o	415

system.

$$A \xleftrightarrow[k_2]{k_1} B$$

The mass and energy balance equations of this reaction are given by

$$\dot{x}_A = k_1(1-x_A) - k_2x_A = R \tag{33}$$

$$\dot{T} = JR - U(T-T_c) \tag{34}$$

The reaction constants are a function of temperature and exhibit Arrhenius kinetics.

$$k_1 = k_{10}e^{-E_1/R_oT} \text{ and } k_2 = k_{20}e^{-E_2/R_oT} \tag{35}$$

The reaction parameters of this system are given in Table 1.

This problem has been solved by several researchers [Ko and Stevens, 1971b; Modak et al., 1989; Siebenthal and Aris, 1964] using various optimal control methods. Ko and Stevens [1971b] have used the combined gradient method to calculate the optimal control profile of the heat transfer rate. Optimal profiles for several cases of control variable constraints are obtained using the combined method. Modak et al. [1989] used a unidirectional algorithm to calculate the optimal control profile. The optimal switching points were estimated and the flow rate was calculated explicitly.

1-1. Singular Control Approach

The control variable U is the heat transfer rate of the tubular reactor and is dependent upon fluid mixing or other coolant properties. The minimal control action of this system is zero, denoting the reactor at the adiabatic state. The maximal value of the control action is determined by the coolant properties and flow rate.

The object of this control problem is to maximize the concentration of B. It is the same as minimizing the concentration of A.

$$J = \underset{U}{\text{Min}}[x_A(t_f)] \tag{36}$$

The Hamiltonian of the system is given as

$$H = \lambda_1[k_1(1-x_A)-k_2x_A] + \lambda_2[J(k_1(1-x_A)k_2x_A) - U(T-T_C)] \tag{37}$$

The adjoint variables are calculated by taking the derivative of the Hamiltonian with respect to the state variables with final conditions calculated by the derivative of Eq. (36) with respect to the state variables.

$$-\dot{\lambda}_1 = -(\lambda_1+\lambda_2J)(k_1+k_2) \qquad \lambda_1(t_f)=1 \tag{38}$$

$$-\dot{\lambda}_2 = -(\lambda_1+\lambda_2J)[k_1'(1-x_A)-k_2'x_A] - \lambda_2U \qquad \lambda_2(t_f)=0 \tag{39}$$

Since the Hamiltonian is a linear function of the heat transfer rate, this system is classified as a singular control problem and it may have bang-bang and singular control. The switching function of this system is calculated from the derivative of the Hamiltonian with respect to the control variable.

$$\phi = \frac{\partial H}{\partial U} = -\lambda_2(T-T_C) \tag{40}$$

Control action U is determined by the sign of the switching function. If the switching function is greater than zero, the control action should be minimal. If it is less than zero, the control action should be maximized to minimize the Hamiltonian. When the switching function is equal to zero, a singular control algorithm should be applied to calculate the optimal control action. By application of singular control theory, the optimal control profiles are calculated. Singular control appears only when $\lambda_2=0$. The optimal control during the singular interval is calculated by setting the switching function equal to zero. The singular control profile of this system has been calculated by Ko and Stevens [1971b].

$$U_s(t) = \frac{JR}{T-T_c} + \frac{R_{21}R}{R_{22}(T-T_c)} \tag{41}$$

where, $R_{21} = \dfrac{\partial^2R}{\partial T\partial x_A}$ and $R_{22} = \dfrac{\partial^2R}{\partial T^2}$.

From Eq. (39) and $\lambda_2=0$, the relation between temperature and conversion during the singular control period is derived as follows:

$$T = \frac{E_1-E_2}{\ln\left[\dfrac{k_{10}E_1(1-x_A)}{k_{20}E_2x_A}\right]} \tag{42}$$

The relation between temperature and conversion during the singular period is derived by Eq. (42). After calculation of conversion and temperature, the singular control profile of U is calculated by using the derivative of the conversion and Eq. (34).

1-2. Nonsingular Control Approach

As shown in the example, it is difficult to derive the explicit functional form of singular control profiles using singular control technique and to determine switching times for some cases.

Therefore, we transformed the singular control problem to a nonsingular control one with temperature as a control variable. The objective for this system is to maintain the reactor temperature at its optimal state by manipulating the heat transfer rate. Since the reaction starts at the initial temperature (not singular temperature) and the cooling rate is limited by coolant properties, the control of the temperature profile is constrained. Therefore, the optimal control of this system is composed of a constrained period and an optimal period. During the first period of reaction, the control variable U has limited control action in attaining a singular state as soon as possible. The first constrained control action is used to increase the reactant temperature for this problem. The switching time is the point at which the control action changes from bang-bang to singular control. If the maximum control action is infinite, impulse control action can be used to change a reaction environment to a singular state. The control variable U controls the temperature at its optimal state. Therefore, the coolant flow rate control problem can be converted into a simple temperature control problem. The control action U is determined by converting Eq. (34) into U as a function of temperature and conversion.

$$U=\frac{\dot{T}+JR}{T-T_c} \tag{43}$$

By applying the maximum principle to Eq. (33), we can calculate the optimal trajectories of temperature and concentration by the following procedure. To minimize the final concentration of X_1, the reaction rate should be maintained at the maximal rate during the reaction period. The objective is achieved by making the derivative of Eq. (33) with respect to temperature equal to zero.

$$\frac{dR}{dT}=0=\frac{k_{10}E_1}{R_oT^2}\,e^{-E_1/R_oT}\cdot(1-x_A)-\frac{k_{20}E_2}{R_oT^2}\,e^{-E_2/R_oT}\cdot x_A \tag{44}$$

The temperature is calculated from Eq. (44) and the solution is the same as Eq. (42).

$$T=\frac{E_1-E_2}{\ln\left[\dfrac{k_{10}E_1(1-x_A)}{k_{20}E_2x_A}\right]} \tag{45}$$

Some problems can be solved by nonsingular algorithm with a mathematical approach, in which a switching time does not need to be estimated. From Eq. (44), the phase plane temperature and concentration are calculated. Fig. 1(a) shows the optimal trajectory (singular trajectory) phase plane. If the initial concentration of C_A is pure, the initial optimal operating temperature for this system is infinity. Since the reactant feed temperature is 415 K, the first control action for this system is the minimal value of the control variable to increase the reactor temperature up to the singular temperature. Therefore, the singular control action should be followed by minimal control action. By applying minimal control action to Eq. (34), the trajectories of temperature and conversion are calculated by the following procedure. Let U=0 and divide Eq. (34) by Eq. (33). The slope of initial control action on the phase plane is 300, as shown by the dotted line in Fig. 1(a).

$$\frac{\dot{T}}{\dot{x}_A}=\frac{dT}{dx_A}=J=300 \tag{46}$$

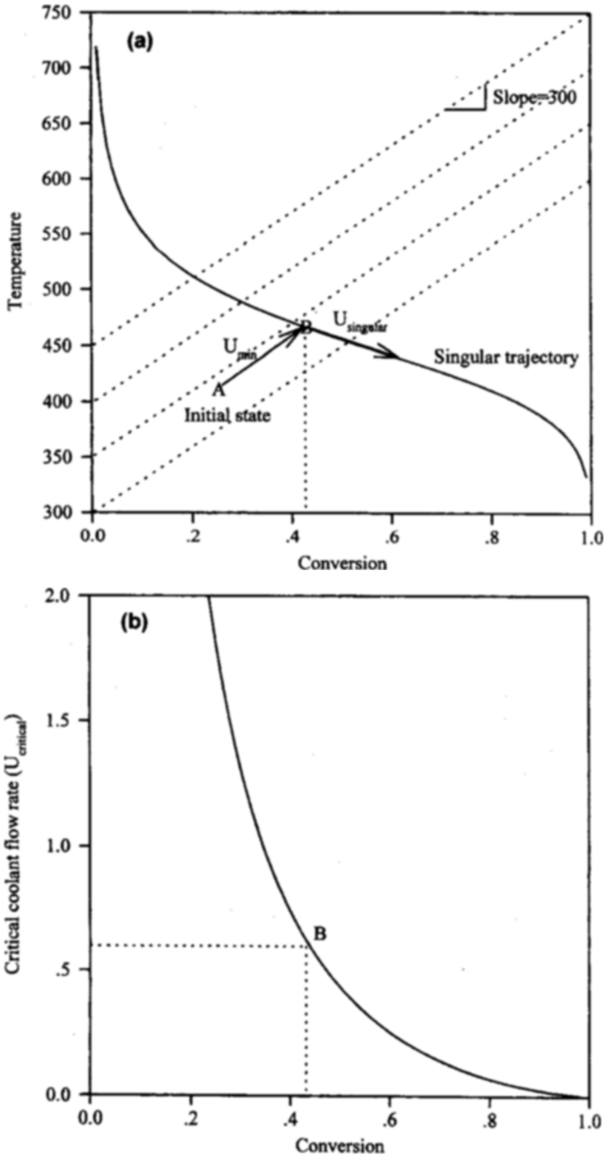

Fig. 1. (a) Phase plane of possible optimal trajectory. (b) Critical heat flow rate.

From the initial state, the reactor operation follows the straight dotted line, the slope of which is 300. The straight line intersects with the singular arc (optimal trajectory) and follows it until the reaction stops or final conditions are reached. There can be another limited control action, the existence of which is determined by the maximal heat transfer rate. If the maximal control variable is smaller than the critical control variable, there should be another limited control action (U_{max}). The critical control variable is calculated at the intersection between the dotted line and singular arc. The critical flow rate is determined by Eqs. (33) and (34), and the result is shown in Fig. 1(b). The gradient, dx_2/dx_1, is calculated by numerical methods using the results in Fig. 1(a). The following equations were developed from the relation between Eq. (33) and Eq. (34):

$$\frac{\dot{T}}{\dot{x}_A}=\frac{dT}{dx_A}=\frac{JR-U_c(T-T_c)}{\dot{x}_A} \tag{47}$$

The critical coolant flow rate is calculated from Eq. (47).

$$U_c=\frac{\left(J-\dfrac{dX_2}{dX_1}\right)R}{X_2-T_c} \tag{48}$$

As shown in Fig. 1(b), the critical control variable of this system is 0.6. At the intersection, the conversion and temperature are 0.42 and 470 K, respectively. When the conversion is 0.42, the critical control variable is 0.6. It is represented by the dotted line in Fig. 1(b). Fig. 2(a) shows the optimal control profile

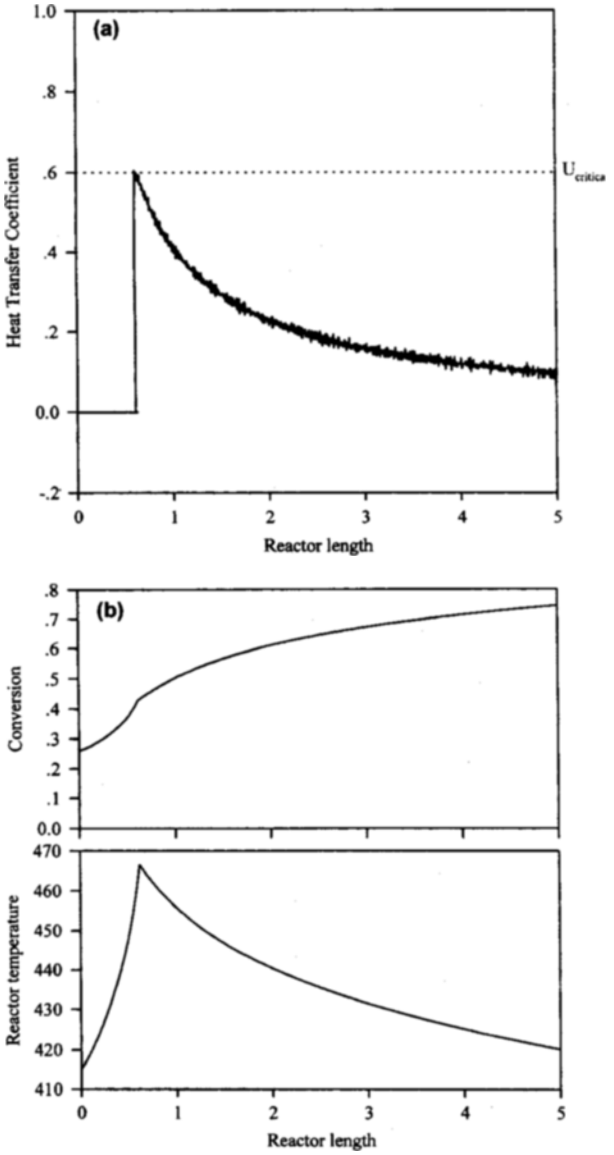

Fig. 2. (a) Optimal heat transfer rate profile calculated by nonsingular transformation algorithm (control action changes from minimum to singular). (b) Optimal temperature and conversion profiles initial temperature is 415 K and conversion is 0.25.

of the tubular reactor calculated from the optimal temperature profile shown in Fig. 2(b). As explained before, the control action changed from minimal to singular. Since the critical control variable is the same as the maximal control variable, only one switching point exists in this problem. Point A in Fig. 1(a) is the reaction starting point of this system. At point B, the control action changes from minimum to singular. The optimal conversions and temperature profiles are shown in Fig. 2(b). The temperature rapidly increases when the control variable is a minimum. After the minimal control action, the temperature profile follows the singular control profile. Ko and Stevens [1971b] have drawn the phase diagram after solving the problem using a combined gradient method and analyzing the optimal trajectories.

2. Example II. Series Reaction in a Plug Flow Reactor

To review the number of switching time changes with changes in the control variable constraints, a series reaction in a plug flow reactor is used as a model system [Douglas and Denn, 1965]. The change in number of switching points is calculated by the nonsingular control technique. There are three components in the reactor. The reaction constants follow Arrhenius kinetics.

$$A \xrightarrow{k_1} B \xrightarrow{k_2} C$$

The reaction kinetics and the initial concentration of each component are calculated as follows:

$$\frac{dX_A}{dt} = -k_1 C_{Ao} x_A^2 \qquad x_A(0) = 1 \tag{49}$$

$$\frac{dx_B}{dt} = k_1 C_{Ao} x_A^2 - k_2 x_B \qquad x_B(0) = 0 \tag{50}$$

$$x_C = 1 - x_A + x_B \tag{51}$$

where, $k_1 = k_{10} e^{-E_1/R_o T}$ and $k_2 = k_{10} e^{-E_2/R_o T}$.

The material and heat balance equations for a plug flow reactor have the following form.

$$v \frac{dx_A}{dz} = -k_1 C_{Ao} x_A^2 \tag{52}$$

$$v \frac{dx_B}{dz} = k_1 C_{Ao} x_A^2 - k_2 x_B \tag{53}$$

$$\frac{dT}{dz} = \frac{2\pi r h(T_c - T) - ((-\Delta H_1) k_1 C_{Ao}^2 x_A^2 + (-\Delta H_2) k_2 C_{Ao} x_B) \pi r^2}{\pi r^2 v \rho C_p} \tag{54}$$

where, $\rho = \rho_A x_A + \rho_B x_B + \rho_C x_C$, and $C_p = C_{P_A} x_A + C_{P_B} x_B + C_{P_C} X_C$.

The object of this control problem is to maximize the concentration of B at the end of the reaction by manipulating the coolant temperature. The performance index of this problem is

$$PI = \underset{T_c}{\mathrm{Min}}[-x_B(z=1)] \tag{55}$$

We have used several dimensionless variables to simplify the equations.

2-1. Singular Control Approach

Eqs. (52) to (54) are changed to a simple dimensionless form and are given as the following equations:

$$\frac{dx_A}{dy} = -\alpha x_A^2 e^{-\kappa_1/\theta} \tag{56}$$

$$\frac{dx_B}{dy} = -\alpha x_A^2 e^{-\kappa_1/\theta} - \beta x_B e^{-\kappa_2/\theta} \tag{57}$$

$$\frac{d\theta}{dy} = \gamma(\theta_c - \theta) - [\alpha \delta_1 x_A^2 e^{-\kappa_1/\theta} + \beta \delta_2 x_B e^{-\kappa_2/\theta}] \tag{58}$$

Table 2. Dimensionless variables used in the series reaction

$$y = \frac{z}{l},\ \alpha = \frac{k_{10} C_{Ao} l}{V},\ \beta = \frac{k_{20} l}{v},\ \gamma = \frac{2hl}{rv\rho C_p},\ \delta_1 = \frac{(-\Delta H_1) C_{Ao}}{\rho C_p T_R},$$

$$\delta_2 = \frac{(-\Delta H_2) C_{Ao}}{\rho C_p T_R},\ \theta = \frac{T}{T_R},\ \kappa_1 = \frac{E_1}{R_o T_R},\ \text{and}\ \kappa_2 = \frac{E_2}{R_o T_R}.$$

For the minimization of the performance index, Pontryagin's maximum principle is used. The Hamiltonian of this system is given as

$$\begin{aligned}H=&\lambda_1(-\alpha x_A^2 e^{-\kappa_1/\theta})+\lambda_2(\alpha x_x^2 e^{-\kappa_1/\theta}-\beta x_B e^{-\kappa_2/\theta})\\&+\lambda_3[\gamma(\theta_c-\theta)-(\alpha\delta_1 x_A^2 e^{-\kappa_1/\theta}+\beta\delta_2 x_B e^{-\kappa_2/\theta})]\end{aligned} \tag{59}$$

To maximize the Hamiltonian of this system, the switching function is employed and the singular control technique applied for this system. The switching function of this system is

$$\phi=\frac{\partial H}{\partial\theta_c}=\lambda_3\gamma \tag{60}$$

To maximize the Hamiltonian of this system, the following control scheme should be applied:

$$\begin{array}{ll}\phi>0 & \theta_c=\theta_{cmax}\\ \phi<0 & \theta_c=\theta_{cmin}\\ \phi=0 & \theta_c=\text{singular control}\end{array} \tag{61}$$

When $\phi=0$, a singular control strategy is needed for this system. A singular control region exists for the case where $\lambda_3=0$ and $\dot{\lambda}_3=0$. The adjoint variables of this system are calculated as follows:

$$-\frac{d\lambda_1}{dy}=\frac{\partial H}{\partial x_A}=2\alpha(\lambda_2-\lambda_1-\lambda_3\delta_1)x_A e^{-\kappa_1/\theta} \tag{62}$$

$$-\frac{d\lambda_2}{dy}=\frac{\partial H}{\partial x_B}=-(\lambda_2+\lambda_3\delta_2)\beta e^{-\kappa_2/\theta} \tag{63}$$

$$\begin{aligned}-\frac{d\lambda_3}{dy}=\frac{\partial H}{\partial\theta}=&[\lambda_1(-\alpha x_A^2 e^{-\kappa_1/\theta})+\lambda_2\alpha x_A^2 e^{-\kappa_1/\theta}\\&+\lambda_3\alpha\delta_1 x_A^2 e^{-\kappa_1/\theta}](\kappa_1/\theta^2)\\&-(\lambda_2\beta x_B e^{-\kappa_2/\theta}+\lambda_3\beta\delta_2 x_B e^{-\kappa_2/\theta})(\kappa_2/\theta^2)-\lambda_3\gamma\end{aligned} \tag{64}$$

The final conditions of the adjoint variables are calculated by differentiating the performance index with respect to its state variables.

$$\lambda_1(t_f)=0.0,\ \lambda_2(t_f)=-1,\ \text{and}\ \lambda_3(t_f)=0.0 \tag{65}$$

The singular control technique is applied to the system. However, the explicit form of θ_C is not calculated during the derivation of switching functions. The derivation of $\dot{\lambda}_3$ is too complex to calculate as an explicit function of θ_C. Since the final condition of λ_3 is equal to zero and its derivative is also equal to zero, this system should be simplified to the optimal temperature profile calculation problem. The simplified adjoint variable equations are as follows:

$$-\frac{d\lambda_1}{dy}=\frac{\partial H}{\partial x_A}=2\alpha(\lambda_2-\lambda_1)x_A e^{-\kappa_1/\theta} \tag{66}$$

$$-\frac{d\lambda_2}{dy}=\frac{\partial H}{\partial x_B}=-\lambda_2\beta e^{-\kappa_2/\theta} \tag{67}$$

$$\begin{aligned}-\frac{d\lambda_3}{dy}=\frac{\partial H}{\partial\theta}=&(-\lambda_1\alpha x_A^2 e^{-\kappa_1/\theta})+\lambda_2\alpha x_A^2 e^{-\kappa_1/\theta})(\kappa_1/\theta^2)\\&-(\lambda_2\beta x_B e^{-\kappa_2/\theta})(\kappa_2/\theta^2)=0\end{aligned} \tag{68}$$

Since $\lambda_3=0$, the temperature profile during the singular region is calculated by Eq. (68). The optimal temperature profile for the singular region of a tubular reactor is

$$\theta=\frac{\kappa_1-\kappa_2}{\ln\left[\dfrac{\kappa_1(\lambda_2-\lambda_1)\alpha x_A^2}{\kappa_2\lambda_2\beta x_B}\right]} \tag{69}$$

The optimal control profile of θ_c is calculated from the optimal temperature profile calculated by Eq. (69).

2-2. Nonsingular Control Approach

When the initial reactant temperature is not at the singular state, the first control action should be bang-bang (maximum or minimum) to bring the reactor to the singular state. If the initial temperature of reactant is at the singular state, this problem can be solved by a simple gradient search algorithm. Otherwise, bang-bang control is applied. This bang-bang control is converted to a temperature gradient limiting control. For the singular control region, this problem can be converted to a temperature optimal control problem. The temperature of the bang-bang control region is constrained by the initial reactant temperature and maximal (or minimal) temperature variations due to coolant temperature limitation. Therefore, Eqs. (44) to (46) are rewritten as below with the control variable, θ. The optimal profile of the dimensionless temperature θ can be calculated by control vector iteration. After the optimal temperature is calculated, the optimal coolant temperature is calculated from state variables and the derivative of the reactor temperature. The performance index is the same as that of the singular control system.

$$\frac{dx_A}{dy}=-\alpha x_A^2 e^{-\kappa_1/\theta} \tag{56}$$

$$\frac{dx_B}{dy}=\alpha x_A^2 e^{-\kappa_1/\theta}-\beta x_B e^{-\kappa_2/\theta} \tag{57}$$

$$P=\underset{\theta}{\mathrm{Max}}[x_B(z=l)] \tag{58}$$

The relation between coolant and reactant temperatures is calculated by Eq. (46).

$$\theta_c=\theta+\frac{\left[\dfrac{d\theta}{dy}+(\alpha\delta_1 x_A^2 e^{-\kappa_1/\theta}+\beta\delta_2 x_B e^{-\kappa_2/\theta})\right]}{\gamma} \tag{70}$$

The Hamiltonian of this system is described as follows:

$$H=\lambda_1(-\alpha x_A^2 e^{-\kappa_1/\theta})+\lambda_2(\alpha x_A^2 e^{-\kappa_1/\theta}-\beta x_B e^{-\kappa_2/\theta}) \tag{71}$$

Adjoint variables of nonsingular control variables are defined as follows:

$$-\frac{d\lambda_1}{dy}=\frac{\partial H}{\partial x_A}=2\alpha(\lambda_2-\lambda_1)x_A e^{-\kappa_1/\theta} \tag{72}$$

$$-\frac{d\lambda_2}{dy}=\frac{\partial H}{\partial x_B}=-\lambda_2\beta e^{-\kappa_2/\theta} \tag{73}$$

For the optimal operation of the system, the derivative of the Hamiltonian with respect to the dimensionless temperature should be zero.

$$\begin{aligned}\frac{\partial H}{\partial\theta}=&(-\lambda_1\alpha x_A^2 e^{-\kappa_1/\theta}+\lambda_2\alpha x_A^2 e^{-\kappa_1/\theta})(\kappa_1/\theta^2)\\&-(\lambda_2\beta x_B e^{-\kappa_2/\theta})(\kappa_2/\theta^2)=0\end{aligned} \tag{74}$$

Dimensionless temperature constraints of this system are $\theta(y=0)=\theta_o$ and $\dot{\theta}_{min} \leq \dot{\theta} \leq \dot{\theta}_{max}$. The control variable constraints, θ_{min} and θ_{max}, are determined by the coolant or steam temperature. The reactor temperature is changed with the coolant temperature changes. The optimal control profile for the overall reaction period is calculated from the adjoint variable and other state variables. The optimal temperature profile for the singular region is obtained from a simple analytical technique. The dimensionless temperature profile is calculated from Eq. (74). The result is the same as Eq. (69).

$$\theta=\frac{\kappa_1-\kappa_2}{\ln\left[\frac{\kappa_1(\lambda_2-\lambda_1)\alpha x_A^2}{\kappa_2\lambda_2\beta x_B}\right]} \tag{69}$$

The reaction parameters used for the simulation are listed in Table 3.

Using the kinetic parameters in Table 3, we calculated the optimal temperature by Eq. (74). The optimal temperature profile for this reaction is shown in Fig. 3. The optimal starting temperature is 370 °K. The reactant temperature should be increased up to 370 °K to increase the reaction rate from component A to component B. If the temperature is higher than the optimal, B converts to C rapidly. For an initial reaction temperature of 280 °K, the optimal operation of this system is shown in Fig. 4.

Since the initial temperature is lower than the optimal temperature, the reactant should be heated up to optimal reaction temperature. When the dimensionless reactor length is 0.08, the reaction condition reaches the singular arc and this arc is maintained to the end of the reactor. Since the initial temperature is low, the reaction rate is slow and there is small change in conversion.

If there is a state variable constraint in this system due to the properties of the reactor, the optimal results can also be calculated by the nonsingular method. Since the reactor temperature is taken as the control variable, the optimal operating temperature is calculated by a simple gradient technique with control variable constraint. After the calculation of the optimal reactor temperature, the coolant temperature is calculated from Eq. (70). When the dimensionless reactor temperature does not exceed 0.35, the optimal reactor and coolant temperature profiles are shown in Fig. 5.

Table 3. Kinetic parameters which used for this system [Douglas and Denn, 1965]

Parameter	Values
k_{10}	5×10^{10}
k_{20}	3.33×10^{17}
E_1	18000
E_2	30000
C_{P_A}	1
C_{P_B}	1
C_{P_C}	1
ρ_A	1
ρ_B	1
ρ_C	1
$-\Delta H_1$	3000
$-\Delta H_2$	3000
h	10
r	1
l	100
v	10

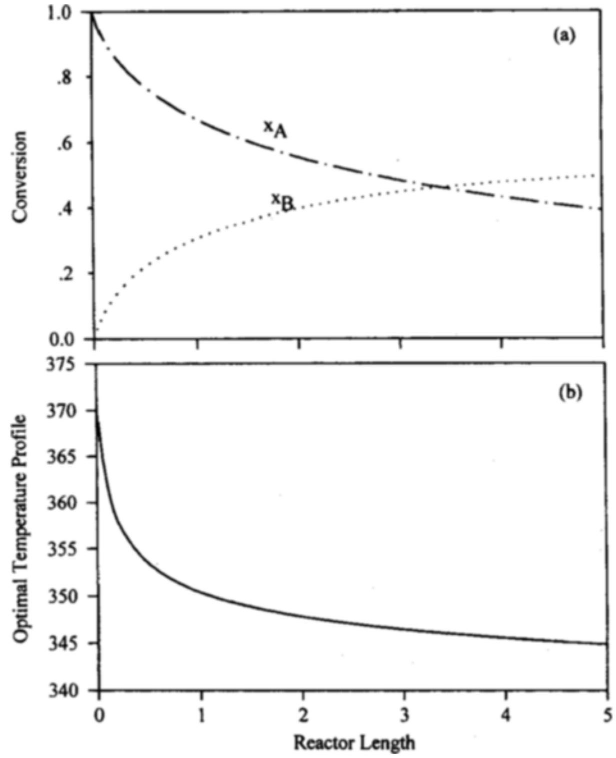

Fig. 3. Optimal temperature and conversion profiles without coolant and steam temperature constraints.
(a) Conversion and (b) Temperature profile

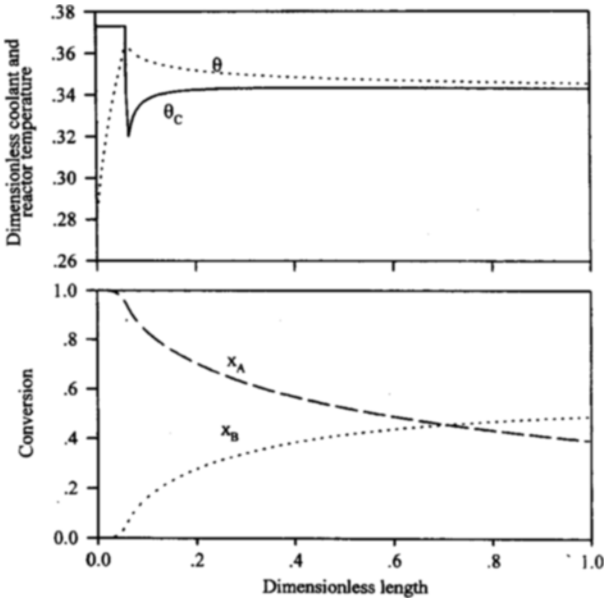

Fig. 4. The optimal profiles of state and control variables with steam temperature constraints (θ_o=0.28, θ_{cmax}=0.373).

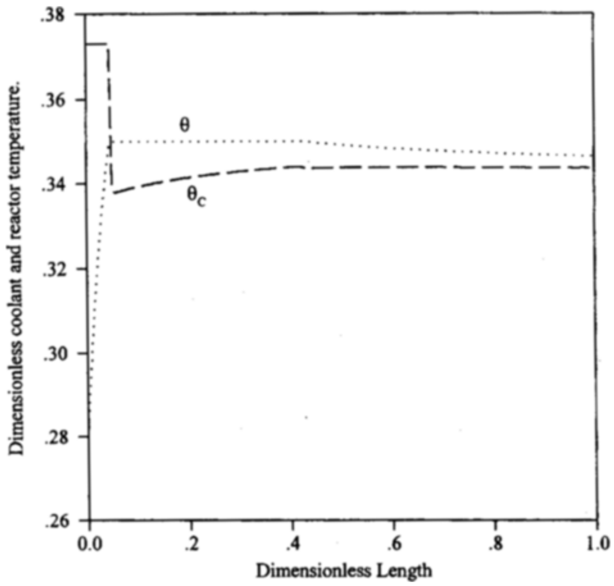

Fig. 5. Optimal coolant and reactor temperature profiles reactor temperature cannot be greater than 0.35 (θ_o=0.28, θ_{max}= 0.353, and θ_{cmax}=0.372).

This system can have one more control action change, depending on the minimal coolant temperature particularly, when the heat transfer rate is decreased to a half of the original transfer rate. Coolant temperature should be lower than that indicated in Fig. 4. Fig. 6(a) shows the sharp decrease in coolant temperature due to the low heat transfer rate. The minimal cooling dimensionless temperature of Fig. 6(a) is 0.269. It is lower than the freezing point of water. Because iced water cannot be used as a coolant, the minimal coolant temperature should be higher than the chilled water temperature 0.283. Considering coolant temperature limitations, the optimal control profile of this system is shown in Fig. 6(b). The number of switching times is increased from 1 to 2 due to the control action constraints.

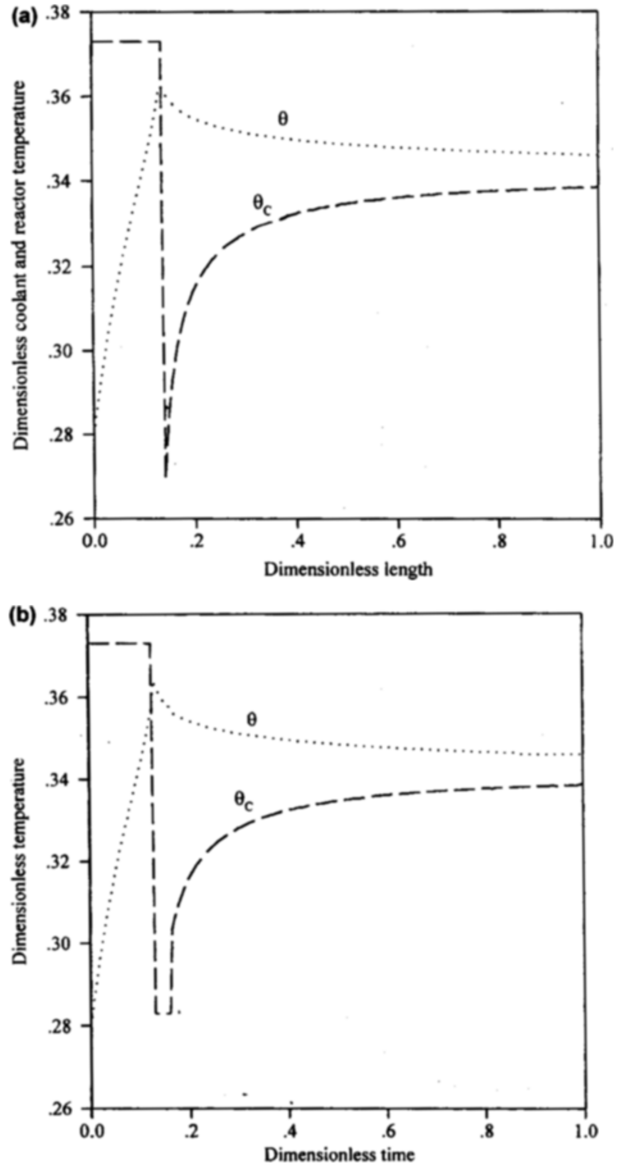

Fig. 6. (a) Heat transfer rate is decreased to half of original value without coolant temperature constraint. (b) Heat transfer is decreased to half of original value with coolant temperature constraint (θ_{cmin}=0.283).

CONCLUSION

A nonsingular transformation algorithm was proposed in this paper. The equivalency between the singular control algorithm and nonsingular transformation algorithm was verified with mathematical derivation.

Two chemical engineering problems, exothermic chemical reaction and series reaction, were proposed as examples. The numerical optimization algorithm was proposed for the calculation of optimal control profiles. The calculated optimal control profiles with a nonsingular transformation algorithm were shown to be same as those calculated with a singular control algorithm.

For the system with one or more switching points in the optimal control profiles, it was difficult to determine the optimal profiles with the singular control algorithm. However, the optimal control profiles were easily calculated with the developed nonsingular transformation algorithm.

NOMENCLATURE

A, B : chemical species
b : constant vector
c : function vector (f_ib)
C_{Ao} : initial concentration of component A [gmol/cm^3]
C_p : heat capacity [cal/g/K]
E : activation energy [cal/gmol]
E_1 : activation energy corresponding forward reaction [cal/mole]
E_2 : activation energy corresponding backward reaction [cal/mole]
F : control variable
f : nonlinear vector function of state vector
F_{max} : maximum allowed control
F_{min} : minimum allowed control

F_s : singular control
g : functional form of control variable
G(g) : functional form of performance index
ΔH : heat of reaction [cal/gmol]
h : heat transfer coefficient [cal/cm^2/K/min]
H : Hamiltonian
J : $(-\Delta H)/C_p\rho$ [K mL/mole]
k_1 : reaction constant [gmol/cm^3/min]
k_2 : reaction constant [1/min]
l : the reactor length
PI : performance index
q : order of singularity
r : radius of reactor [cm]
R : reaction rate [min^{-1}]
R_o : gas constant [cal/gmol/K]
T : reactant temperature [K]
t : reaction time [hr]
T_c : cooling jacket temperature [K]
t_f : final time [hr]
T_o : initial reactant temperature [K]
U : control variable, proportional to heat transfer coefficient
v : flow velocity [cm/min]
V : volume [L]
V_{max} : maximum reactor volume [L]
$\mathbf{x}$: state vector
x_A : mole fraction of A
$\mathbf{x}_o$: initial condition of state vector
y : dimensionless variable [z/l]
z : reactant position [cm]
$\mathbf{z}$: transformed state vector

Greek Letters

α : dimensionless variable $\left(\alpha=\frac{k_{10}C_{Ao}l}{v}\right)$
β : dimensionless variable $\left(\beta=\frac{k_{20}l}{v}\right)$
δ : dimensionless variable $\left(\delta_i=\frac{(-\Delta H_i)C_{Ao}}{\rho C_p T_R}\right)$
ϕ : switching function
γ : dimensionless variable $\left(\gamma=\frac{2hl}{rv\rho C_p}\right)$
κ : dimensionless variable $\left(\kappa_i=\frac{E_i}{RT_R}\right)$
λ : vector of adjoint variables
θ : dimensionless temperature [T/T_R]
ρ : density of fluid [g/cm^3]

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