

## Modeling and Adaptive Pole-Placement Control of LDPE Autoclave Reactor

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**ABSTRACT** - A two-compartment four-cell model is developed for the adiabatic autoclave slim type reactor for free radical polymerization of low density polyethylene(LDPE). The mass and energy balances give rise to a set of ordinary differential equations, and by analyzing the system it is possible to predict properly not only the reactor performance but also the properties of polymer product. The steady state multiplicity is found to exist and examined by constructing the bifurcation diagram. The effects of various operation parameters on the reactor performance and polymer properties are investigated systematically to show that the temperature distribution plays the central role for the properties of polymer product. Therefore, it is essential to establish a good control strategy for the temperature in each compartment. In this study it is shown that the reactor system can be adaptively controlled by pole-placement algorithm with conventional PID controller. To accomplish a satisfactory control, the estimator and controller are initialized during the period of start-up.

### Introduction

Low density polyethylene(LDPE) was first produced by reaction experiment at high pressures in ICI, 1933. The processes for high pressure polyethylene followed two basic routes; autoclave and tubular reactors.

Ethylene is polymerized in the stirred autoclave via free radical mechanism using organic peroxides as initiators at temperatures ranging from 150 ~ 300°C and pressures from 1000 atm to 3000 atm. The set of reaction temperature and pressure determines the phase condition within the reactor; i.e., single or two phases[Fig. 1]. Since the reaction is highly exothermic, and the reactors are usually operated under adiabatic conditions, the heat of reaction is absorbed by the incoming feed stream as sensible heat. Hence, the monomer conversion in the reactor is dependent on the temperature difference between the reactor inlet and outlet. Conversion ranges normally between 10 and 20% and the reactor residence time varies from 10 to 60 seconds. Especially, it is a usual practice that commercial high pressure polyethylene reactors are operated near the edge of stability for the sake of greater productivity.

There are two basic types of the autoclave reactors : (1) highly backmixed reactor, such as Du Pont compact reactor and (2) less

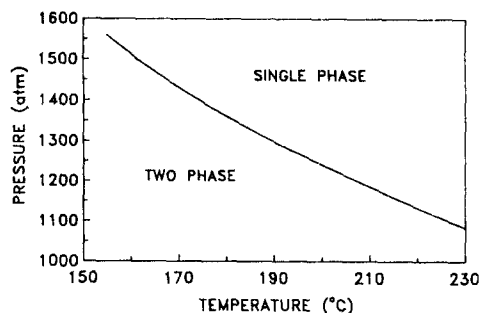


Fig.1. Ethylene/LDPE phase diagram.

highly mixed one, such as ICI slim type reactor and National Distillers Autoclave. Figure 2 shows a slim type reactor. This type of reactor has an internally mounted stirrer motor and the length to diameter ratio falls in the range of 8~20. Reactors of this type can be compartmented into two or more zones and operated with appropriate temperature distribution in the reactor. It is used to produce narrow MWD resins which are well suited for film and injection molding application.

In the area of the polymerization reactor control, the research interest has been focused on the MMA and styrene polymerization reactors under low pressure condition. Continuous polymerization reactor control for the MMA solution polymerization has been studied by Kwalik and Schork[9], Tanner et al.[17], Adebekun and Schork[1] and Mendoza-Bustos et al.[11], while batch polymerization reactor control has been treated by Ponnuswamy et al.[14], Tzouanas and Shah[18] and Kiparissides et al.[8]. Ponnuswamy et al.[14] used the Pontryagin's maximum principle to control the molecular weight by manipulating the temperature and the initiator concentration in MMA solution polymerization reactor. Kwalik and Schork[9] applied the adaptive pole-placement algorithms to control the reactor temperature by manipulating the jacket temperature and the initiator feed concentration. The advanced adaptive pole-assignment control has been applied by Tzouanas and Shah[18] to control the monomer conversion and the polymer properties such as weight average molecular weight. Control of styrene polymerization reactor has been investigated by Henderson and Cornejo[7], and Takamatsu et al.[16].

In most cases of the polymerization reactor control, the target

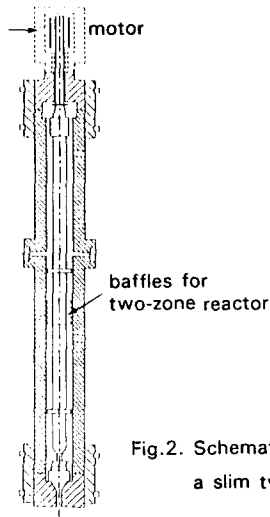


Fig.2. Schematic diagram of a slim type reactor.

is the control of the reactor temperature, the molecular weight distribution or the copolymer composition, whereas the manipulated variables are the jacket temperature and the feed concentration of the monomer, initiator or solvent.

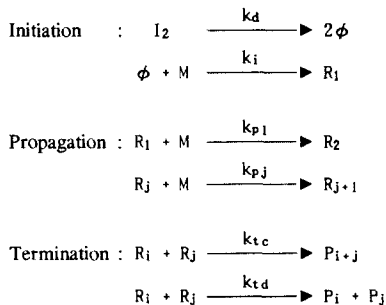
Among various objectives of the polymerization reactor control, the most important one is the control of the polymer properties. For this purpose it is prerequisite to develop some method of on-line measurement and to estimate the parameters.

Ponnuswamy et al.[13] examined a variety of problems related to the on-line measurement and the control of batch polymerization reactor. A Kalman filter has been applied by Schuler and Suzhen[15] to estimate the state of free radical polymerization reactor.

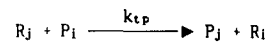
The present study aims for the application of pole-placement adaptive controller to a highly nonlinear process related to the LDPE autoclave reactor. The principal objective is to establish the feasibility of implementing this adaptive control structure and to elucidate the practical techniques required for a successful closed-loop operation.

### Kinetic Mechanism

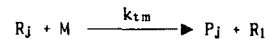
A deterministic description of the highly complicated physical and chemical processes in a polymerization reactor is based on the kinetic analysis of the polymerization mechanism. Here we shall adopt the free radical polymerization reaction mechanism and thus the elementary reactions to be considered are as follows :



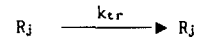
Chain transfer to polymer :



Chain transfer to monomer :



Short chain branching :



The symbols,  $I$ ,  $\phi$ ,  $M$ ,  $R_j$  and  $P_j$  represent the initiator, free radical, monomer, living polymer, and dead polymer, respectively. The reaction rate constants except for  $k_{tp}$  are assumed independent of the chain length  $j$  and hence are determined at any temperature and pressure by the Arrhenius expression. According to Feucht et al.[5], the rate of chain transfer to polymer is dependent on the chain length of the polymer. In addition, the density of the polymer can be calculated from the number of short chain branches by means of the empirical relationship[6]

$$\rho_{\text{polymer}} = 0.9723 - 0.018 \ln(V_{zk})$$

where  $V_{zk}$  is  $500 k_{tr}/(k_p M)$  [ $\text{CH}_3/10^3 \text{ C}$ ].

The parameters for the reaction rate constants are taken from literature[2,3,5]. The initiators considered in this study are acetyl cyclohexanesulphonyl peroxide, ter-butyl peroxy-pivalate and ter-butyl peroxyacetate.

### Model Development

The adiabatic reactor for ethylene polymerization may be appropriately described by mass balances of the monomer, the initiator, the total living polymer and the total dead polymer, the moment equations for the concentrations of total living polymer and total dead polymer, and the energy balance for the reaction mixture. The slim type reactor to be treated here shall be represented by the two-compartment four-cell model with backflow in each compartment. For the four-cell model as shown schematically in Fig.3, the mass and energy balances yield a set of ordinary differential equations which may be put in the form as follows :

$$\begin{aligned}
 V_i dC_{ji}/dt &= q_i C_{rji} + q_i' C_{j,i+1} - \left( \sum_{k=1}^i q_k + q_i' + q_{i-1}' \right) C_{ji} \\
 &\quad + \left( \sum_{k=1}^{i-1} q_k + q_{i-1}' \right) C_{j,i-1} + V_i r_{ji} \\
 \rho C_p V_i dT_i/dt &= \rho C_p [q_i T_{rj} + q_i' T_{i+1} - \left( \sum_{k=1}^i q_k + q_i' + q_{i-1}' \right) T_i \\
 &\quad + \left( \sum_{k=1}^{i-1} q_k + q_{i-1}' \right) T_{i-1}] + V_i r_{pi} (-\Delta H_p) + P_i
 \end{aligned} \tag{1}$$

The first subscript  $j$  in  $C_{ji}$  and  $r_{ji}$  represents the species in the order of  $I, M, G, G', G'', F, F'$  and  $F''$ , where  $G$  and  $F$  are the total concentrations of living and dead polymers, respectively, and the prime and the double prime denote the first and second moments. The expressions for  $r_{ji}$  ( $j=1,2,\dots,9$ ) are given in Table 1. The second subscript  $i$  ( $i=1,2,3,4$ ) denotes the cell number,  $r_p$  is the polymerization rate given in Table 1, and  $P$

represents the rate of heat generation by mixing[12].

The numerical data used for simulation of the slim type reactor are given in Table 2. The parameters are the initiator feed concentrations, the feed temperature and the feed flow rates.

To examine the steady state, it is often convenient to analyze the dependence of the solution upon distinguished parameters which are called the bifurcation parameters. Here we use the software

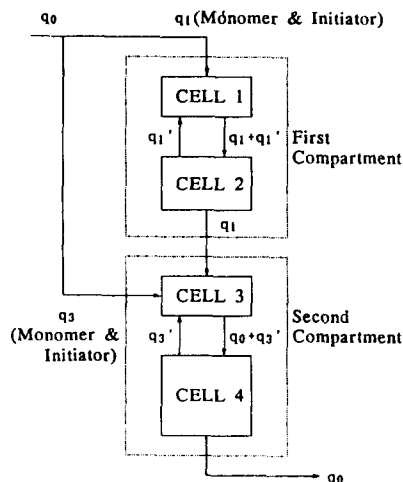


Fig.3. Schematic diagram of the two-compartment four-cell model for the slim type reactor.

Table 1. Expressions for the reaction rates.

$r_I = -k_d \cdot I$
$r_M = -2 \cdot f \cdot k_d \cdot I - k_p \cdot M \cdot G - k_{tm} \cdot M \cdot G$
$r_G = 2 \cdot f \cdot k_d \cdot I - (k_{td} + k_{tc}) \cdot G^2$
$r_{G'} = 2 \cdot f \cdot k_d \cdot I + k_p \cdot M \cdot G - (k_{td} + k_{tc}) \cdot G \cdot G' + k_{tm} \cdot M \cdot (G - G') + k_{tp} \cdot (G \cdot F'' - G' \cdot F')$
$r_{G''} = 2 \cdot f \cdot k_d \cdot I + k_p \cdot M \cdot (2G' + G) - (k_{td} + k_{tc}) \cdot G \cdot G'' + k_{tm} \cdot M \cdot (G - G'') + k_{tp} \cdot (G \cdot F''' - G'' \cdot F')$
$r_F = k_{td} \cdot G^2 + \frac{1}{2} k_{tc} \cdot G^2 + k_{tm} \cdot M \cdot G$
$r_{F'} = (k_{td} + k_{tc}) \cdot G \cdot G' + k_{tm} \cdot M \cdot G' + k_{tp} \cdot (G' \cdot F' - G \cdot F'')$
$r_{F''} = k_{td} \cdot G \cdot G'' + k_{tc} \cdot (G \cdot G'' + G'^2) + k_{tm} \cdot M \cdot G'' + k_{tp} \cdot (G'' \cdot F' - G \cdot F''')$
$r_P = k_p \cdot M \cdot G$

Table 2. Thermodynamic data and conditions for the reactor.

$\rho$	413	g/l
$C_p$	0.518	cal/g·K
$(-\Delta H_p)$	21400	cal/gmol
$f$	1	-
$P$	1200	atm
$T_f$	293.15	K
$q_0$	18	l/s
$V_1$	64	l
$V_2$	97	l
$V_3$	65	l
$V_4$	215	l

package "AUTO" written by Doedel[4], whereas for the transient behavior we use the semi-implicit fourth order Runge-Kutta method.

### Parameter Estimation

On-line determination of process parameters is central in adaptive control and it is an important part of a self-tuning regulator. In adaptive systems, the parameter estimation is used in the larger context of system identification.

In this study, we use the recursive least squares method which is frequently applied in conjunction with adaptive control because of its rapid convergence and excellent model tracking.

It is particularly simple for a mathematical model that can be represented by equations of the form

$$Y(t) = \phi_1(t)\theta_1 + \phi_2(t)\theta_2 + \dots + \phi_n(t)\theta_n = \phi^T(t)\theta \quad (2)$$

where  $Y$  is the observed variable,  $\theta_1, \theta_2, \dots$ , and  $\theta_n$  are unknown parameters, and  $\phi_1, \phi_2, \dots$ , and  $\phi_n$  are known functions that may depend on other known variables. The least-squares estimate of  $\theta$  then satisfies the recursive equations

$$\begin{aligned} \hat{\theta}(t) &= \hat{\theta}(t-1) + K(t)[y(t) - \phi^T(t)\hat{\theta}(t-1)] \\ K(t) &= P(t-1)\phi(t)[\lambda + \phi^T(t)P(t-1)\phi(t)]^{-1} \\ P(t) &= (I - K(t)\phi^T(t))P(t-1)/\lambda \end{aligned} \quad (3)$$

where  $\lambda$  denotes the forgetting factor for which we use 0.98 in this study.

### Pole-Placement Control

An R-S-T digital controller can be designed by employing this control strategy for both stable and unstable systems. The structure of the closed-loop system is given in Fig. 4. The plant to be controlled is characterized by the pulse transfer function :

$$H(q^{-1}) = \frac{q^{-d} B(q^{-1})}{A(q^{-1})} \quad (4)$$

in which  $d$  is the integer number of sampling periods contained in the time delay, and

$$\begin{aligned} A(q^{-1}) &= 1 + a_1 q^{-1} + \dots + a_n q^{-n} \\ B(q^{-1}) &= b_1 q^{-1} + b_2 q^{-2} + \dots + b_m q^{-m} \end{aligned} \quad (5)$$

The closed-loop transfer function is given by

$$\begin{aligned} H_{CL}(q^{-1}) &= \frac{q^{-d} T(q^{-1}) B(q^{-1})}{A(q^{-1}) S(q^{-1}) + q^{-d} B(q^{-1}) R(q^{-1})} \\ &= \frac{q^{-d} T(q^{-1}) B(q^{-1})}{P(q^{-1})} \end{aligned} \quad (6)$$

in which

$$\begin{aligned} P(q^{-1}) &= A(q^{-1}) S(q^{-1}) + B(q^{-1}) R(q^{-1}) \\ &= 1 + p_1 q^{-1} + p_2 q^{-2} + \dots \end{aligned} \quad (7)$$

This function defines the closed-loop pole and represents the regulation behavior.

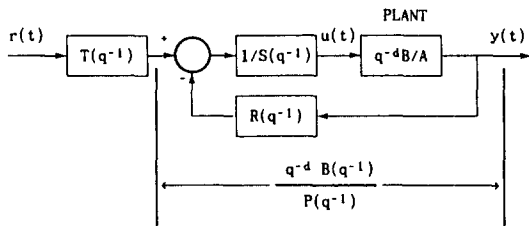


Fig.4. Pole-placement with R-S-T controller.

## Results and Discussion

Bifurcation analysis may be applied to a nonlinear lumped parameter system for the purpose of investigating the effects of various process parameters on the steady state. Once the values of the parameters are given, the steady state solution can be continuously computed as one of the parameters changes. For the values listed in Table 2, the steady state of the present model is analyzed and discussed.

In Fig.5 the feed temperature is chosen as the bifurcation parameter and the diagram is drawn for the exit temperatures of the first and second compartments. The steady state multiplicity exists and the reactor system may have up to nine different steady states for the particular range of the feed temperature. The molecular weight distribution curve (Molecular weight=12676, Polydispersity=4.5,  $\rho_p=0.9158$ ) for the conditions in Table 2 is presented in Fig. 6.

For the systematic analysis of the reaction system, we use the two parameter continuation method. Figure 7 shows the results of the two parameter continuation on the plane of the feed temperature and the initiator feed rate, and on the plane of the feed temperature and the residence time  $\tau$  which is defined by

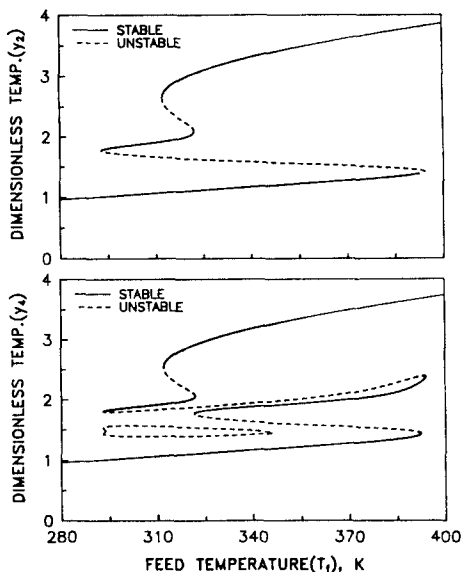


Fig.5. Bifurcation diagrams for the exit temperature of each compartment showing the effect of the feed temperature.

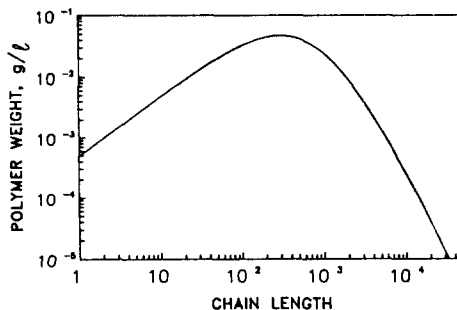


Fig.6. Molecular weight distribution curve obtained under the conditions specified in Table 2.

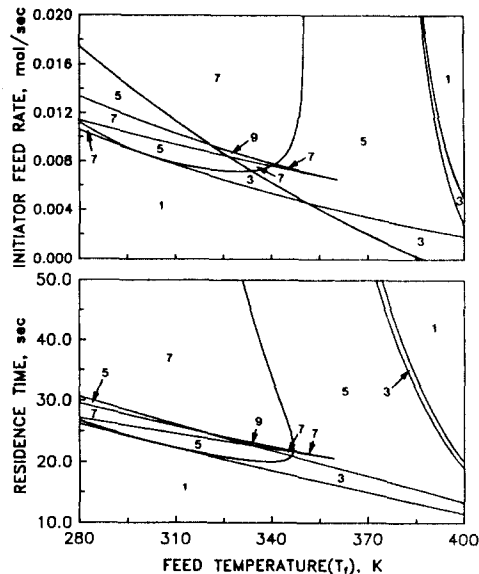


Fig.7. Multiplicity regions of the steady states.

the total reactor volume  $V_t$  divided by the total flow rate  $q_0$ . Here the number in the diagram indicates the number of the steady states. Using these results, we can determine appropriate ranges of control parameters.

In the polymerization reactor, it is important to predict the physical properties (molecular weight, density, etc.) of the polymer product. In Fig. 8 variations of polymer properties with the feed temperature are compared between the cases of temperature control and open-loop operation. When the reactor temperature is controlled, it can be expected that the properties of the polymer are also controlled to some extent.

Figure 9 illustrates the reactor and trajectory temperature profiles during the start-up procedure along with the required variations of the initiator feed rates  $U_2$  and  $U_4$ . In this step both the pressure and the mass flow rate are increased and, as mentioned above, the initiators of three different kinds are used at the low, middle and high temperature ranges, respectively, and both the estimator and the controller are initialized. By regulating the tracking parameters, the reactor temperature overshoot and the time required for the start-up can be reduced.

The pole-placement controller is implemented to control the reaction temperature of the LDPE slim type reactor. In this case the closed-loop characteristic equation is specified as

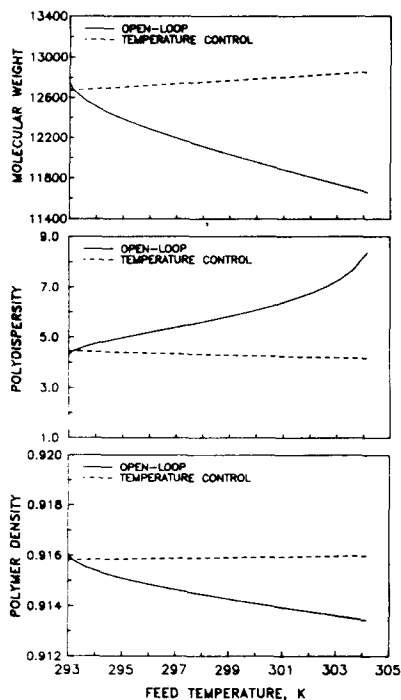


Fig.8. Variations of polymer properties with the feed temperature for the cases of the reactor temperature control and the open-loop operation.

second order. The performance of this controller is investigated for changes in the set-point as well as for disturbances mounted on the load. Presented in Fig. 10 are the responses of the exit temperature from and the initiator feed rate to each compartment under the control with the pole-placement regulator and the curves are compared with those obtained by using the conventional PID controller.

At point S, five percent step increase in the set-point is introduced. If compared with the performance of the PID controller, the overshoot and the time to reach the new steady-state are reduced. Since, however, the initiator feed rates change abruptly, the performance of control valve must be taken into account in actual application. At point L, one percent step increase in the feed temperature is introduced as a disturbance in the load. In this case it is noticed that, after a short transient period, the regulator stabilizes the controlled variables.

### Conclusions

This study is concerned with the evaluation of adaptive pole-placement control strategy and by implementation on a realistic model for the LDPE autoclave reactor. A simulation model is derived describing the bulk polymerization of low density polyethylene in the slim type reactor. This system exhibits strong non-linearities and thus steady state multiplicity. For the systematic analysis of the reaction system, two parameter continuation method was used and by this analysis the appropriate ranges of control parameters can be determined.

The adaptive pole-placement controller performs adequately when applied to the highly nonlinear LDPE polymerization reactor.

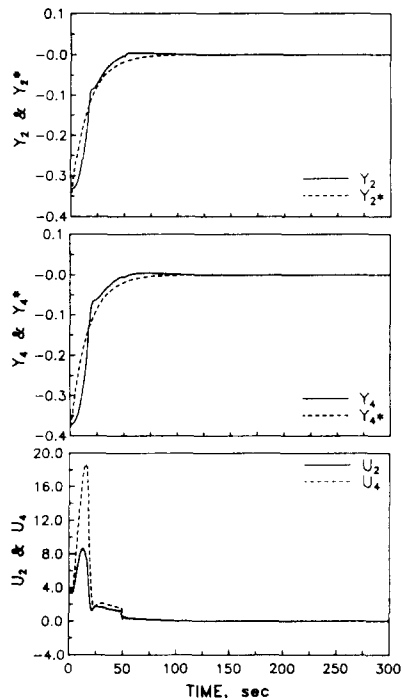


Fig.9. Start-up procedure with the reactor temperature control.

The closed-loop results demonstrate a satisfactory performance with this control strategy. An inherent difficulty encountered in the application of this control scheme is the implementation of the estimator and controller. The key to the adaptability of the control algorithm rests with the performance of the parameter estimator. Since the initiator decomposition rate is very fast, the reaction temperature changes very rapidly during the course of determining the model parameters. Therefore, it is important to estimate the parameters properly. In this study, estimator and controller are initialized during the start-up procedure.

### Notation

$C_p$	: Heat capacity of the reaction mixture	[cal/g. $^{\circ}$ C]
$f$	: Initiator efficiency	
$G$	: Living polymer concentration	[mol/l]
$F$	: Dead polymer concentration	[mol/l]
$\Delta H_p$	: Heat of polymerization	[cal/mol]
$I$	: Initiator concentration	[mol/l]
$k$	: Reaction rate constant	[s $^{-1}$ or l/mol.s]
$M$	: Monomer concentration	[mol/l]
$P$	: Reactor pressure	[atm]
$R_j$	: Living polymer species of chain length $j$	
$P_j$	: Dead polymer species of chain length $j$	
$q$	: Flow rate	[l/s]
$q^{-1}$	: Backward shift operator	
$T$	: Temperature	[K]
$U$	: Dimensionless initiator feed rate	
$Y$	: Dimensionless observed temperature	
$Y^*$	: Dimensionless trajectory temperature	

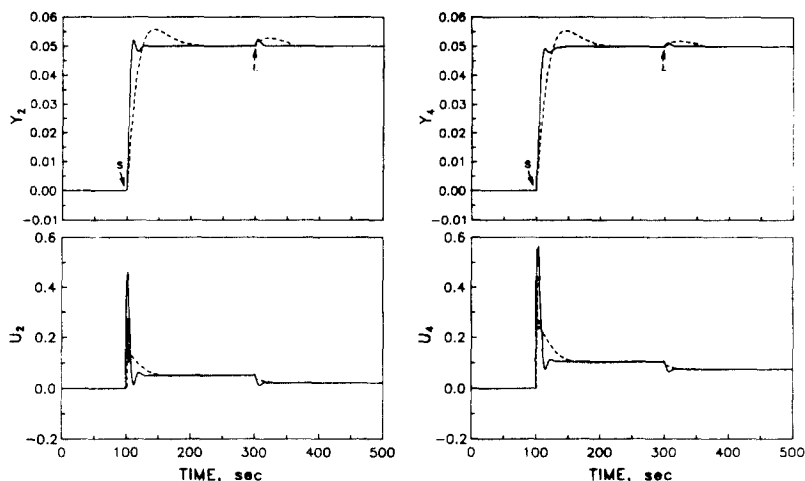


Fig.10. Performances of the adaptive pole-placement controller(—) and the PID controller(----) in response to a set point change and to a load disturbance.

$y$  : Dimensionless temperature

$V$  : volume [L]

#### Greek letters

$\lambda$  : Exponential forgetting factor

$\theta_i$  : Model parameter

$\rho$  : Density of the reaction mixture [g/L]

$\tau$  : Reactor residence time [s]

#### Subscripts

$f$  : Feed condition

$i$  :  $i$ -th cell or chain length

$j$  : Chain length

1, 2, 3, 4 : Cell number

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