Korean J. Chem. Eng., **26**(5), 1201-1207 (2009)
DOI: 10.2478/s11814-009-0193-8

Application of experimental non-linear control based on generic algorithm to a polymerization reactor

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(*Received 18 September 2008 • accepted 1 February 2009*)

Abstract−The dynamics of free radical polymerization of styrene and on-line control of temperature in a cooling jacketed batch polymerization reactor is investigated. The benzoyl peroxide initiator is introduced into the reactor once at the beginning of the reaction to obtain the desired monomer conversion and the desired average chain length in a minimum reaction time. The optimal constant set temperature, which is generally realized in industrial applications, and the set profile are used as two different optimal operating conditions. The temperature control of the polymerization reactor is achieved experimentally and theoretically. The control of nonlinear systems has progressed considerably, and various nonlinear process model based control techniques have appeared in the literature. The problem is how to tune the controller in order to obtain comparable closed loop responses. Generic model control (GMC) is applied and the performance of the control results are compared with the previously published control results.

Key words: Polystyrene, Non-linear Control, Generic Model Control

INTRODUCTION

Polymerization reactors are widely used in the petrochemical industry. The optimization and the optimal control of these reactors are of great importance in obtaining a desired quality of product [1]. Model-based algorithms are similar in the sense that they rely on models to predict the behavior of the process over some future time interval, and the control calculations are based on these model predictions. The models used for these predictions have been linear approximations of the process which are obtained experimentally from step response data. Unfortunately, strong nonlinear behavior is often found in real chemical processes such as highly exothermic batch polymerization reactors [2-4]. There are also some approaches in which linear control is used, combined with non-linear models for setpoint updating [5]. There is a strong desire to develop a generic control structure that directly imbeds nonlinear process models [6-10]. Generic model control (GMC) assumes a desirable process output trajectory and derives a nonlinear control law [11]. Suzuki et al. [12] investigated theoretically a design method for a temperature control system using generic model control (GMC). Ali et al. [13] developed a modified generic model controller theoretically and tested it through a simulation study.

This paper presents a control strategy that is applied on batch reactor both theoretically and experimentally. This strategy is based on the principle of most suitable process model in order to improve the control algorithm. The process model has emerged from mass and energy preservation correlations. The heat emerging from the reaction has been estimated on-line, by utilizing artifical neural network and kinetic information. An on-line controlled cooling jacketed batch reactor has been used in experimental study. First, dynamic studies have been achieved experimentally. During these experiments, the change of reactor temperature, monomer conversion, viscosity and molecular weight values was obtained. Experiments were done at different optimum conditions. Temperature control was realized by using a GMC algorithm. For this reason, the algorithm was written in Visual Basic. The control performance was determined by tracking the set point very closely.

MATHEMATICAL MODEL OF THE POLYMER REACTOR

1. Kinetic Model

The free radical kinetic model given below was determined from the literature and experimental data. The mass balance equation for the initiator was:

$$\frac{dI}{dt} = -k_d I \tag{1}$$

The mass balance equation for the monomer was:

$$\frac{dM}{dt} = -k_p\left(\frac{2fk_d I}{k_t}\right)^{1/2} M = R_M \tag{2}$$

Table 1. Kinetic and physical parameters

Constant	Values	
A_d	2.6×10^{16}	s^{-1}
A_p	1.051×10^{7}	1/mol·s
A_t	1.255×10^{9}	1/mol·s
E_d	34200	cal/mol·K
E_p	7060	cal/mol·K
E_t	1680	cal/mol·K
M_o	6,6985	mol/*l*
f	0.5	-
G	1	-
M^*	0.5-0.3	-
X_n^*	500-1000	-

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By making long-chain assumption, the overall rate of heat production by the reactions is given by:

$$R_H=(-\Delta H)R_M \tag{3}$$

The parameters of the kinetic model were determined from experimental reaction curves and the computer data bank. For the identification of kinetic model, a least square method was used. The kinetic parameters obtained for various temperatures are given in Table 1.

2. Reactor Model

The following assumptions were made in the mathematical modelling of a cooling jacketed batch polymerization reactor:

1. Temperature, specific heat and conversion of monomer do not vary with position in the reactor which is well mixed.

2. The mean heat capacity of medium in the reactor is independent of the composition and temperature.

The heats of initiation and termination are negligible compared to the heat of propagation.

4. The inlet temperature of the cooling water is constant.

5. The decrease in volume due to increase in density was neglected. Therefore, the change of monomer concentration was accepted as due only to changes occurring during the reaction.

6. The change of viscosity has no effect upon the reaction kinetics.

The overall heat transfer coefficient in the energy balance is assumed to be a function of viscosity. The energy balance for the reactor can be given as follows:

$$V\rho C_p\frac{dT}{dt}=Q+(-\Delta H)R_m V-UA(T-\bar{T}_c) \tag{4}$$

Where:

$$U=\frac{1}{S\mu_r^{0.33}+F}$$

Here, S and F are constants [4]. Energy balance for the cooling jacket is as follows:

$$V_c\rho_c C_{pc}\frac{dT_{co}}{dt}=mC_{pc}(T_{ci}+T_{co})+UA(T-T_c) \tag{5}$$

$$\bar{T}_c=\frac{T_{ci}+T_{co}}{2}$$

The empirical viscosity equation containing the initial viscosity (μ_0), monomer (M), initiator concentrations (I_o, M_o) and average molecular weight (M_w) was found from the dynamic studies as follows [4]:

$$\mu_r=\mu_o+0.9615\frac{[(M_o-M)M_w]^{0.654}}{(I_o-I)^{-0.588}} \tag{6}$$

$$\mu_o=3.727\times10^{-3}T_o+3.009\times10^{-3}M_o^2-327.6I_o^3$$

The mathematical model developed with the help of Eqs. (1)-(6) was solved with the Runge-Kutta integration method. The results were compared with the experimental values and good agreement was obtained.

CONTROLLER DESIGN

Generic model control has strong advantages for developing nonlinear controllers. Application of GMC algorithm to a styrene poymerization reactor is explained in detail as follows:

The transfer function between the output y and the setpoint y_d is given below:

$$y(s)=\frac{Gp*Gc}{1+Gp*Gc}yd(s) \qquad q(s)=\frac{Gp*Gc}{1+Gp*Gc} \tag{7}$$

Where, q(s) presents the reference model. The controller acts in such a way as to keep the output y close to the references model output. The transfer function of the controller is given in Eq. (8).

$$u=Gc*e \qquad Gc=\frac{q}{Gp(1-q)} \tag{8}$$

The generic model controller tries to eliminate the difference between the output, y and the references model output y_r ($y=y_r$).

$$\frac{dy_r}{dt}=K_1(y_{set}-y)+K_2*\int(y_{set}-y)dt \tag{9}$$

by using the Laplace transforms of the above equation:

$$yr(s)=\frac{K_1s+K_2}{s^2+K_1s+K_2}=\frac{2\xi\tau s+1}{\tau^2s^2+2\xi\tau s+1}$$

Where:

$$\xi=\frac{K_1}{2\sqrt{K_2}} \qquad \tau=\frac{1}{\sqrt{K_2}} \tag{10}$$

Transfer function of controller in Laplace domain:

$$Gc(s)=\frac{K_1s+K_2}{Gp(s)*s^2}=\frac{1}{s}\left[\frac{K_1}{Gp(s)}+\frac{K_2}{Gp(s)s}\right] \tag{11}$$

Without using Laplace domain, the generic model controller evaluates the manipulated variable, u.

Consider a process described by a first order differential equation:

$$\frac{dy}{dt}=f(y,u,d)$$

The algorithm will yield a control law that tracks the desired temperature. To find values of the manipulated input variable which force the controller output to follow a referans model output, the following equation is used:

$$f(y,u,d)=K_1*(yd-y)+K_2*\int_0^t(yd-y)dt \tag{12}$$

A real process generally behaves in a quite nonlinear manner and changes behavior over a period of time. That is why the process model is unclear. This process is described by the following equation:

$$f(y,u,d)=\frac{dT}{dt}=\frac{Q+(-\Delta H)R_mV-UA(T-T_c)}{VgC_p} \tag{13}$$

The manipulated variable is evaluated from the following mathematical description:

$$\begin{aligned}Q=&-(-\Delta H)R_mV+UA(T-T_c)\\&-V\rho C_p\left[K_1(T_{set}-T)+K_2\sum_{i=1}^{k}(T_{set}(i)-T(i))\Delta t\right]\end{aligned} \tag{14}$$

EXPERIMENTAL PROCEDURE

The free radical polymerization of styrene was carried out in a 1.1 L cooling jacketed glass reactor that has four inlets. A rotating

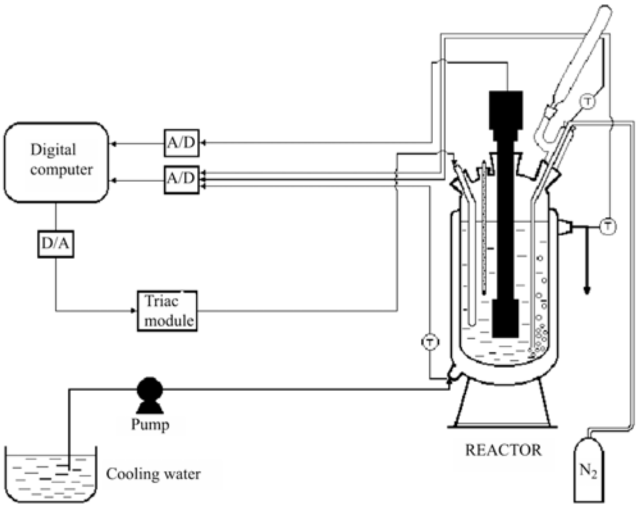

Fig. 1. Experimental system.

viscometer was inserted through the middle inlet and the heater, thermocouple, and reflux condenser were placed in the others. The temperature of the reactor and the inlet and outlet temperatures of the cooling water were determined by thermocouples and transferred into a computer though A/D converters. The viscosity changes were sent by the help of another A/D converter. A thyristor, which is a switching instrument and control power module inside the reactor, was connected to the computer through a D/A converter. The experimental system is given in Fig. 1. The rotating viscometer used in the viscosity measurements sends numerical signals to this digital computer. The power and the agitation speed of this viscometer change due to variations in medium viscosity while concentration of polymer increases. The viscometer is capable of correcting for the smallest changes in viscosity due to changes in temperature. It also works as a stirrer. The circulation flow from the peristaltic pump which regulates the circulation rate of cooling water can be both manually and digitally controlled.

The system was purged with nitrogen, sent from the bottom of the reactor, in order to facilitate the stirring of the system under an inert atmosphere. A reflux condenser was used in the system to regain the vaporized toluene. Styrene was vacuum distilled and benzoyl peroxide initiator was dissolved in chloroform and recrystallized in methanol before use.

The dynamic and control studies were carried out by loading 1.1 L of 70% solution of styrene in toluene, keeping the flow rate of cooling water at a fixed value, and heating the reaction mixture with an electrical heater with the help of a variac/thyristor. Benzoyl peroxide was fed into the system in an instant manner after the system reached a steady-state condition. The experiments were carried out at different isothermal temperature for optimum monomer and initiator concentrations based on the desired conversion and chain length values. During the reaction the viscosity and temperature changes were observed with a digital computer. The temperature was controlled by using GMC methods during the experiments. It is well known that the temperature control of the reactor depends mainly on the heating and cooling systems of the reactor and it is acceptable in industry [14] A jacketed reactor is equipped with an inside heating coil, for which the temperature control is ensured by acting on both variables: the flow rate of cooling water flowing in the cooling jacked and heat input through immersedcoil. Most often, one of these two input rates is fixed and the other is changed as the manipulated variable. In this work, the cold fluid flow rate was set to a mean value and only the heat input rate was adjusted. Very similar type of choice is available in the industrial and laboratory size semi-batch jacketed styrene polymerization reactors'control. The viscosity, amount of polymer, and average molecular weight of the samples were determined by falling ball viscometer, precipitation method and ubbelohde viscometer, respectively. Comparison of the experimental and theoretical results showed good agreement.

All the experimental optimization data are listed in Tables 3 and 4. The following experiments were performed to verify the sensitivity of the control systems and calculate the control parameters. The cooling jacketed reactor was taken as a heat exchanger and the heat given by the immersed heater was observed from the temperature change of the cooling water, and the energy of reactor was kept constant. The mixing chamber was considered to be continuous as regards to energy. When such a mixing chamber was used as a polymer reactor, the heat formed during the reaction was accepted as a disturbance for the heat exchanger. All the control and system parameters were calculated, and control designs were made based on this approach. As well, this mixing chamber was subjected to pseudo effects similar to those in polymer reactors, and the effectiveness of the control systems was checked without consuming of any compound.

OPTIMIZATION

Two optimization methods were used. The first one is to deter-

Table 2. Experimental and theoretical results at optimal operating conditions (using Lagrange)

Optimum parameters					Dynamic (no control)		Experimental		Theoretical	
M_0 (mol/L)	T (°C)	I_0 (mol/L)	X_n	m^*	X_n	m^*	X_n^*	m^*	X_n^*	m^*
6,699	103	0.0126	500	0.5	310	22	415	0.49	429	0.44
6,699	105	0.00378	1000	0.3	699	32	-	-	849	0.26

Table 3. Experimental and theoretical result at optimum operating conditions (using Hamiltonian)

Optimum parameters					Dynamic (no control)		Experimental		Theoretical	
M_0 (mol/L)	T (°C)	I_0 (mol/L)	X_n	m^*	X_n	m^*	X_n^*	m^*	X_n^*	M^*
6,699	92	0.0150	500	0.5	301	35	522	0.55	414	0.48

Table 4. The values used for calculation of (−ΔH R_m) and heat transfer coefficient

Variac (Volt)	90
Cooling water flow rate (ml/min)	40
Inlet temperature of cooling water (°C)	24.5
Temperature of cooling water (°C)	55
Outlet temperature of cooling water (°C)	40
Reactor volume (ml)	1200
Cooling jacket volume (ml)	455
Average temperature of cooling water (°C)	32.25
Area, A (m^2)	0.0611

mine minimum initiator concentration (I_0), optimum reaction temperature (T^*) and minimum polymerization time (t_f^*) for a given monomer conversion (m^*) and average chain length (X_n^*). Experimental verification of these experimental conditions was carried out for one charge of initiator at the beginning of polymerization. The optimum operating parameters for the case of one charge of initiator were calculated from the mass balance equations and Lagrange multipliers. Optimal operation condition is given in Table 2. The second method is to reach the required final monomer conversion and numerical average value of molecular weight in a minimum reaction time; this optimization problem, which includes calculation of optimal temperature profile, was realized. In this calculation, the optimization algorithm is used to solve both algebraic and differential equations concerning polymerization model. For the optimal temperature profile in minimum time, the performance index is determined and the problem is solved by Hamiltonian. Optimal operation condition is given in Table 3.

APPLICATION OF GMC TO A BATCH REACTOR

The GMC algorithm is applied to a batch reactor which is described by the energy balances. This structure derived below allows the incorporation of a process model directly in the control algorithm.

$$F(T,Q,T_j)=\frac{dT}{dt}=\frac{Q+(-\Delta H)R_m V-UA(T-T_c)}{VgC_p}$$

In general, the exact process model is rarely known, and the model is introduced such that:

$$F(T,Q,T_j)-K_1(T_{set}-T)-K_2\int(T_{set}-T)dt=0 \tag{15}$$

The value of the manipulated variable is evaluated from Eq. (16).

$$\begin{aligned}Q=&-(-\Delta H)R_m V+UA(T-T_c)\\&+V\rho C_p\left[-K_1(T_{set}-T)-K_2\sum_{i=1}^{k}(T_{set}(i)-T(i))\Delta t\right]\end{aligned} \tag{16}$$

Using Eq. (16) may cause an applicability problem. Therefore, Eq. (16) is rewritten as Eq. (17). Q(t+1) is evaluated by rearranging Eq. (16) as Eq. (17)

$$\begin{aligned}Q(t+1)=&-(-\Delta H)R_m V+UA(T(t)-T_c(t))\\&+V\rho\rho_p\left[-K_1(T_{set}(t)-T(t))-K_2\sum_{i=1}^{k}(T_{set(i)}(t)-T_{(i)}(t))\Delta t\right]\end{aligned} \tag{17}$$

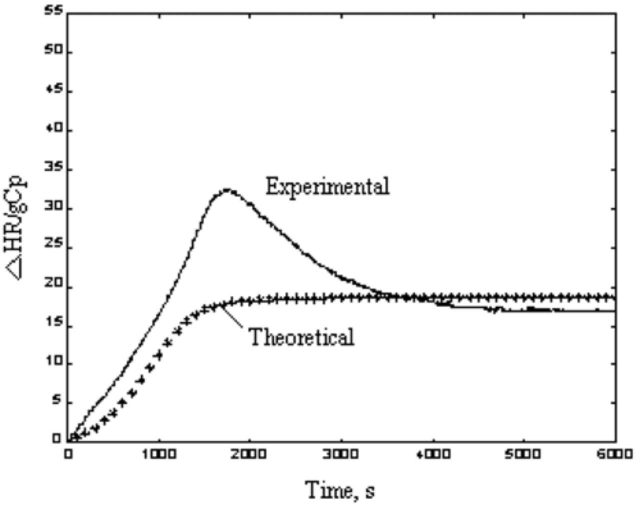

Fig. 2. The change of the term ($\Delta HR_m/\rho C_p$) with time at the condition (T=92 °C, I_0=0.015 mol/*l*).

The value of the reaction heat in Eq. (17) is obtained from kinetic knowledge in theoretical work. It is also calculated on-line in experimental work. The change of the term ($\Delta HRr/\rho C_p$) with time is given in Fig. 2. Also, the values used for calculation of (−ΔH R_m) and heat transfer coefficient are shown in Table 4.

1. Evaluation of the Term, (−ΔH R_m)

The energy balance equation written for reactor is rearranged as follows:

$$\frac{(-\Delta HR_m)}{\rho C_p}=(T_2-T_1)-\frac{Q}{V\rho C_p}-\frac{UA(T-T_c)}{V\rho C_p} \tag{18}$$

Eq. (18) is used in the computer program and the term ($-\Delta HR_m$) is evaluated on-line. In this program, the reaction heat changes with time are given in Fig. 2 at the optimal operating condition (T=92 °C, I_0=0.015 mol/L). Reaction heat profile obtained from kinetic data and neural network by using Eq. (3) is also given in the same figure. Reaction heats were calculated by using artifical neural networks and reaction heats follow the theoretical heat changes very closely. In this calculation, the value of −ΔH, which equals 11,500 cal/molK, is obtained from literature for kinetic data. The rate of reaction, R_m, is evaluated from Eq. (2)

2. Calculation of GMC Constants

Process time constant (τ) is evaluated as 30.5 min by using a process reaction curve [4]. Eq. (10) is used to obtain the constants, K_1 and K_2. By using the time constant value calculated, the various K_2 values obtained from various ξ values are given in Table 5 for calculation of generic model coefficients. By using these coefficients, the control program is executed for 105 °C set point and the integral

Table 5. K_2 and ISE values obtained from various ξ values

ξ	K_2	ISE (T=105 °C)
2	0.131	2.01
3	0.196	1.78
4	0.262	1.97
7	0.459	Control program is out of run

of square of the error (ISE) values are obtained for each run. In Table 5, the suitable ISE value is found by using $K_2=0.196$ and $K_1=0.00107$ values at 105 °C constant set point. In the same table, it is shown that the chosen constant K_2 value is used theoretically to obtain the suitable ISE value. K_1 is also found experimentally by changing the set point temperature, and then a data bank is obtained. During servo control problems K_1 is changed adaptively by using the data bank. In the experimental control studies, these K_1 values are used, and then K_2 is obtained by a similar procedure given in Table 5.

The system closed loop transfer function is given below:

$$\frac{T(s)}{Td(s)}=\frac{183s+1}{30.5^2s^2+183s+1}$$

The relative order of the system is calculated as follows:

$$[\text{The relative order}]=[\text{the order of the numerator}]-[\text{the order of the denominator}]$$

In the present work, relative order is determined as a unit.

THEORETICAL RESULTS OF GENERIC MODEL CONTROL

GMC system is applied to keep the temperature of a free radical solution polymerization of styrene in a jacketed batch reactor. By applying quasi steady-state and long chain approximation for live radicals, mass balances for monomer, initiator and moments of the molecular weight distribution are written for a batch reactor, and in addition, reactor dynamics is expressed by energy balances of the reactor and cooling jacket. The overall heat transfer coefficient for the batch jacketed polymerization reactor is modeled as a function of the viscosity of the reaction mixture. These mass and energy balances are used for simulation study. This simulation program was tested to predict the dynamics of the polymerization reactor theoretically, and in addition it was used to design the GMC system because the temperature variations have a great effect on molecular weight distribution, which determines the quality of polymer product.

The simulation results at optimal conditions are given in Figs. 3 and 4. Dynamic optimal temperature profiles and theoretical results of GMC reactor temperature control are also given in the same figures. The open-loop simulation results deviate from real profiles. Deviations in conversion and molecular weight are about 45% and 60%. In closed-loop experimental temperature profiles, there is not any deviation. Deviation in molecular weight is less than deviation shown in open-loop.

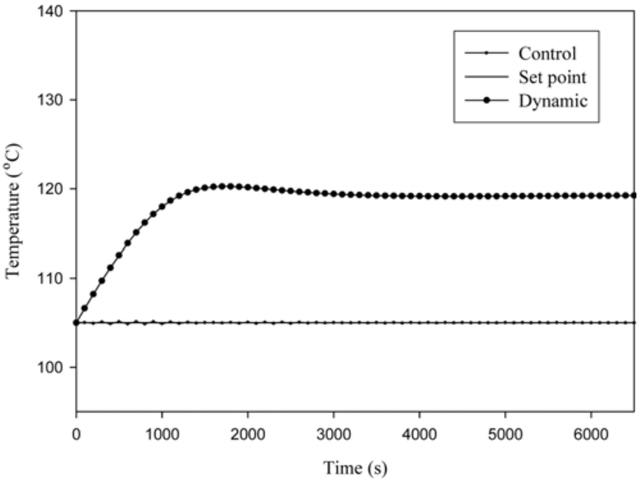

Fig. 3. Dynamic reactor simulation and GMC control results at constant set temperature (T=105 °C, I_o=0.00378 mol\l).

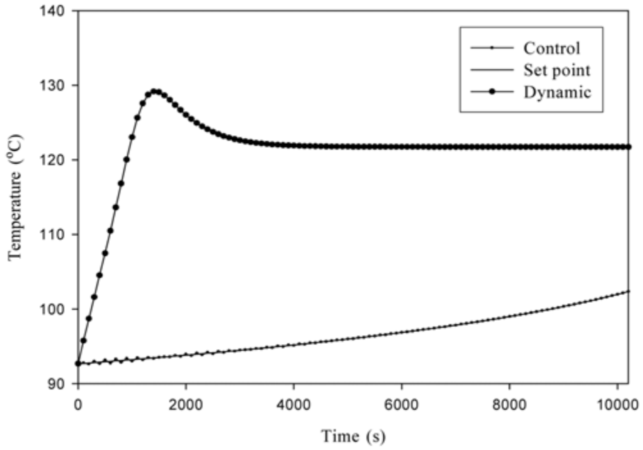

Fig. 4. Dynamic reactor simulation and GMC control results at optimal set temperature profile (T=92 °C, I_o=0.0150 mol\l).

EXPERIMENTAL RESULTS OF TEMPERATURE CONTROL OF POLYMERIZATION REACTOR

In this section, temperature control of styrene and toluene polymerisation with benzoyl peroxide initiator as achieved by using the GMC algorithm. Experiments were achieved at two different optimal conditions which are given in Tables 2 and 3. The aim of the control is to achieve reaction at optimal temperature profile, which is essential for the required optimal conditions. In the polymerisation reactor, 1.1 L mixture is utilized. During the experiment, to determine conversion and viscosity, 5 ml and 15 ml sampling mixtures were taken. These samples also affect as a disturbance. After the viscosities were measured, the sampling mixtures were added to the reactor to keep mixture amount constant. This also makes a second

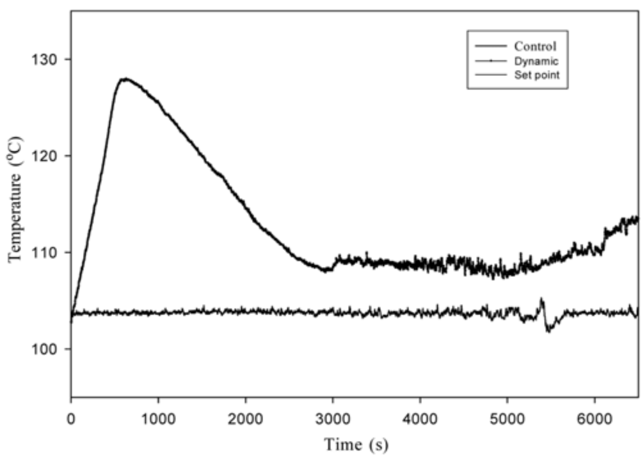

Fig. 5. Experimental GMC control of reactor temperature and dynamics (T=103 °C, I_o=0.0126 mol/l).

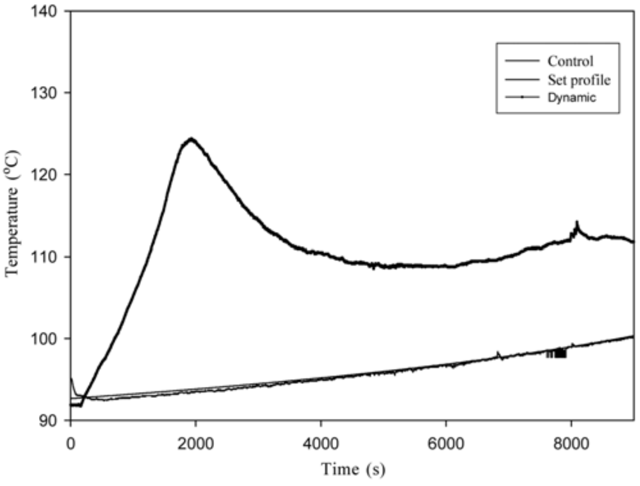

Fig. 6. Experimental GMC control of reactor temperature and dynamics (T= 92 °C, I_o=0.0150 mol/*l*).

load effect. Although all these load effects exist in the experiments, the reaction must be continued at the required temperature profile. Therefore, cooling water was fed to the jacketed pilot-plant reactor at a constant flow rate, and to obtain optimal initial reaction temperature, an immersion heater was utilized. To keep the reaction at the optimal temperature profile, the signals as BYT (2.17 cal/s) values, which are sent to immersion heater, are manipulated by using the GMC algorithm. Experimental results are shown in Figs. 5-6. Although various load effects exist, temperature control was achieved successfully by using GMC. As it is shown in Figs. 5-6 and Tables 2 and 3, required conversion and molecular weight values are obtained with a little error.

CONCLUSION

Non-linear control based on the GMC algorithm is applied theoretically and experimentally to a jacketed batch polymerization reactor. Reaction heat is evaluated on-line by using kinetic data. Optimization is utilized to obtain the final monomer conversion and average number of chain lengths by using mathematical models related with styrene solution polymerization. In the optimal condition, nonlinear and adaptive control work is achieved experimentally. Online performance of GMC control is observed. It is compared with theoretical performance. Methods of identification of control parameters are examined. The GMC is applied to styrene polymerization reaction for on-line temperature control at desired temperature values. Theoretical and experimental polymer conversion and molecular weight values are compared with optimal values to observe success of GMC control application.

The nonlinear process model is utilized in the GMC control algorithm. The GMC algorithm includes change ratio of control variable and feedforward relation. Therefore, nonlinear control structure allows the GMC algorithm to improve the ability of heat and temperature protection without additional important criteria.

There are some previous application papers using GMC. Our control work using set profile is compared with the previous work [15] in Table 6 by using ISE and IAE (the integral of absolute values of error) criteria. As it is clearly seen from using a very similar set profile, our control result is considerably good. In the second part of the study, industrial radical polymerization processes are generally realized at constant temperatures. In radical polymerization the organic initiator (benzoyl peroxide) degradation rate increases when the temperature of the reaction mixture rises. As a result, the molecular weight of the polymer decreases.

Table 6. The present control work using set profile is compared with the previously published work [15]

Controller type	IAE	ISE
FLC (Fazzy LogicControl) (previous work, [14])	7.2	3.88
GMC (Generic Model Control) (previous work, [14])	8.4	9.76
PID (Conventional Control) (previous work, [14])	13.1	11.65
GMC (Generic Model Control) (The present work)	5.5	4.87

In polymerization processes with increasing set temperature, polymer molecular weight changes with time. The dispersion range of product molecular weight increases. The polymerization rate is also affected by changes in temperature.

During radical polymerization it is important to use a constant set temperature for control of the polymerization rate and for obtaining constant quality of the polymer with a certain range of molecular weight. In the present work, using a constant set temperature, very good control result is obtained and shown in Fig. 5.

NOMENCLATURE

A : heat transfer area [m^2]
A_d, A_p, A_t : frequency factor for initiator decomposition, propagation and termination, respectively
C_p, C_{pc} : specific heat of reactor content and coolant [J/k mole °C]
E_d, E_p, E_t : activation energies for initiator decomposition, propagation and termination, respectively
($-\Delta$H): heat of the reaction [J/k mole]
Gp : transfer function of proces
Gc : transfer function of controller
I_o I : initial initiator and initiator concentrations [mole/*l*]
k_d, k_i, k_p : kinetic constants for initiator decomposition, initiator and propagation, respectively
k_{tc}, k_{td} : kinetic constants for termination by combination and disproportionation, respectively (k_t=k_{tc}+k_{td})
K_1, K_2 : constants of GMC
M_o, M: initial monomer and monomer concentrations, respectively [mole/*l*]
m : rate of cooling water [ml/min]
Q : heat input [W]
ρ : density of the mixture [kg/m^3]
ρ_c : density of the cooling water [kg/m^3]
μ_r : viscosity of reactor content [cp]
T, T_c : reactor and cooling water temperatures [°C]
T_{ci}, T_{co} : inlet and outlet cooling water temperatures [°C]
t : time [s]
U : overall heat transfer coefficient [kcal/m^2 K sec.]
V : volume of the mixture [m^3]
V_c : volume of the cooling water jacket [m^3]
X : conversion
y(t), u(t) : output, input variable at time t

d : load effect
f : jel effect
f(…) : nonlinear function

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