IChemE

0263–8762/99/$10.00+0.00
© Institution of Chemical Engineers
Trans IChemE, Vol 77, Part A, July 1999

TEMPERATURE CONTROL OF A BATCH POLYMERIZATION REACTOR

S. YÜCE, A. HASALTUN*, S. ERDOĞAN* and M. ALPBAZ**

Department of Chemical Engineering, Cumhuriyet University, Sivas, Turkey
**Department of Chemical Engineering, Gazi University, Ankara, Turkey*
***Department of Chemical Engineering, Ankara University, Ankara, Turkey*

In this work, DMC (Dynamic Matrix Control) of a batch solution polymerization reactor has been investigated experimentally and by simulation to achieve a specific constant number average chain length and conversion in a minimum time. The process variables are reaction temperature and initiator initiation concentration. The performance of the DMC controller was compared with that of the IMC (Internal Model Control) controller. It was seen that DMC and IMC controllers yielded a good performance in maintaining the reactor temperature at its set point at the isothermal conditions.

Keywords: polymerization; temperature; dynamic matrix control

INTRODUCTION

During the last ten years, researchers have had an increasing interest in controlling polymerization reactors. Indeed, these processes are very complex and show nonlinear and time varying dynamic behaviour. The polymerization reaction generates a large amount of heat, which must be removed from the reactor in order to have the reactor temperature follow the required profile, and be stable. These processes require modeling so as to control and optimize the experimental conditions. Recently, several control algorithms based on predictions derived from an input-output model have been developed for the control of these reactors. One of the algorithms used extensively for the control of complex systems is DMC.

There exists a considerable literature on the control of polymerization reactors. However, application of control techniques in this area is mostly restricted to numerical simulation. Cutler and Ramaker[1] initiated the dynamic matrix control algorithm, and then a few experimental studies with various chemical systems were reported in the literature (Garcia *et al.*[2], Peterson *et al.*[3]).

In this work, experimental applications of DMC and IMC controllers beside simulation studies were accomplished in a batch polymerization reactor. A free radical polymerization of styrene was chosen as a process for the investigation, because polystyrene is an important product in today's industrial polymers and it has a very wide range of applications. In this process, temperature control is the most important control problem. Sacks, Lee and Biesenberger[4] have found that temperature variations have a great effect on MWD (Molecular Weight Distribution) which determines the quality of polymer product. In this work, the control objective is taken as a desired number average molecular weight and a desired conversion in a minimum time. To determine the optimal temperature and optimal initiator initiation concentration, Lagrange's multiplier method (Chen and Jeng[5]) was used. A DMC algorithm was developed for this system and implemented on the experimental reactor system to control the temperature of the polymerization reactor under the optimal conditions. The results obtained from the DMC experiments were compared with the result of IMC experiments performed under the same conditions. These algorithms were also tested in the simulation.

Experimental System

A schematic diagram of the experimental apparatus is provided in Figure 1. The reactor was a 1.1 l glass cylindrical vessel. The reactor content was mixed by a rotating viscometer which also sends on-line the viscosity of the content to the computer. A condenser is attached to one of the head port to provide an exit for the nitrogen and to condense toluene and monomer, because solvent evaporation takes place in the temperature range under consideration. Nitrogen is bubbled through the reaction mixture to keep oxygen out of the system. The temperature of the reactor and the temperatures of the jacket inlet and outlet were measured by thermocouples and sent to the computer. The viscosity of the reacting mixture and temperatures were monitored and observed on the computer during the polymerization.

An immersed heater is provided inside the reactor for liberating the appropriate amount of heat. A thyristor unit regulates the voltage to the immersed heater depending upon the signals from the computer. The flow rate of the coolant is measured by a flowmeter. A microcomputer with A/D and D/A converters is employed for data acquisition and control of the polymerization reactor.

In the experimental work, commercial styrene was firstly vacuum-distilled to remove the inhibitor. Benzoyl peroxide which was dissolved in chloroform and then recrystallized in methanol was used as initiator. Toluene was chosen as a solvent. At the start of the experiment, the

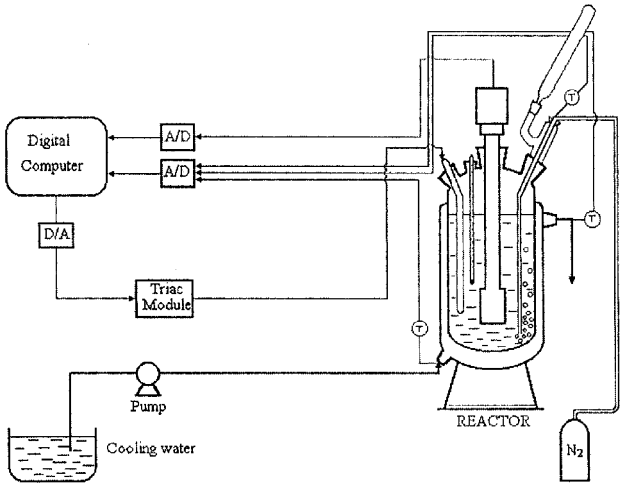


Figure 1. Experimental equipment of polymerization reactor.

reactor content which was 70% styrene and 30% toluene was heated to the desired temperature. Then, the appropriate amount of benzoyl peroxide was added to the reactor. Next, the control methods were applied in the control system of the reactor in order to keep the reactor temperature at the desired value. Polymerization continued at the desired temperature over a calculated length of time. Polymer samples were withdrawn from the reactor at certain intervals for conversion and molecular weight analysis during the polymerization.

MODELLING AND OPTIMIZATION

A basic free radical polymerization mechanism has three essential reaction steps: initiation, propagation and termination. Considering the standard free radical polymerization (Ray[6]; Karagöz *et al.*[7]) and assuming constant density, no chain transfer and no gel effects, and applying quasi steady-state and long chain approximations for live radials, mass balances for monomer, initiator, and moments of the molecular weight distributions in a batch reactor can be written as follows:

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

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

$$\frac{d\mu_0}{dt} = 2f\left(1-\frac{v}{2}\right)k_d I \tag{3}$$

$$\frac{d\mu_2}{dt} = (2+v)\frac{k_p^2}{k_t}M^2 \tag{4}$$

with initial conditions $M(0) = M_0$, $I(0) = I_0$, $\mu_0(0) = \mu_2(0) = 0$.

The monomer conversion and the number chain lengths can be given as

$$m = 1-\frac{M}{M_0}, \qquad X_n = \frac{mM_0}{\mu_0} \tag{5}$$

Assuming perfect mixing, constant reacting heat capacity and density, the reactor dynamic can be expressed by energy balances for the reactor and the jacket as follows:

$$\frac{dT}{dt} = \frac{Q}{M_r c_p} + \frac{(-\Delta H)R_m V}{M_r c_p} - \frac{UA(T-T_c)}{M_r c_p} \tag{6}$$

$$\frac{dT_c}{dt} = \frac{m_c(T_{ci}-T_{co})}{V_c\rho_c} + \frac{UA(T-T_c)}{V_c c_{pc}\rho_c} \tag{7}$$

Here

$$R_m = dM/dt \quad \text{and} \quad T_c = (T_{ci}+T_{co})/2 \tag{8}$$

The overall heat transfer coefficient for the batch jacketed polymerization reactor was modelled as a function of the viscosity of the reacting mixture as

$$U = \frac{1}{S\mu_r^{0.33}+F} \tag{9}$$

Where S and F are constants which depend on the reactor sizes and physical properties, μ_r is the viscosity of the reacting mixture.

The mathematical model equations of this process (equations (1)–(4)) were solved using the 4th order Runge-Kutta integration method.

The control variables in the isothermal batch jacketed reactor are taken as reactor temperature and initial concentration. In order to obtain optimal operating conditions to achieve a predetermined conversion and number average chain length in a minimum time, the method of Lagrange's Multiplier is used. According to this method, the equations for optimal T and I_0 can be obtained by optimizing the following function:

$$W = t_f(T, I_0) + \lambda g(T, I_0) \tag{10}$$

Where λ is the Lagrange's multiplier. The equations for this system are:

$$\frac{\partial t_f}{\partial I_0} + \frac{\partial g}{\partial I_0} = 0 \tag{11}$$

$$\frac{\partial t_f}{\partial T} + \lambda\frac{\partial g}{\partial T} = 0 \tag{12}$$

$$g(T, I_0) = 0 \tag{13}$$

where

$$g(T, I_0) = -\ln(1-m^{\cdot}) + 2A_p\left(\frac{2f}{A_d A_t}\right)^{0.5} \times \exp\left[-\frac{1}{2RT}(2E_p - E_d E_t)\right] \times \left[\left(1-\frac{M_0 m^{\cdot}}{fI_0 X_n^{\cdot}}\right)^{0.5} - 1\right] \tag{14}$$

$$t_f(T, I_0) = -\frac{1}{k_d}\ln\left[1-\frac{M_0^{\cdot}}{fI_0}\right] = -\frac{1}{A_d e^{-E_d/RT}}\ln\left[1-\frac{M_0 m^{\cdot}}{fI_0 X_n^{\cdot}}\right] \tag{15}$$

Here, $m^{\cdot}$ and $X_n^{\cdot}$ are desired conversion and desired number average chain length respectively.

Optimal I_0 and T can be found from the following

equations obtained from the solution of equations (11)–(15).

$$\frac{E_d}{(3E_d + E_t) - 2E_p}\ln\left[1 - \frac{M_0\dot{m}}{fI_0\dot{X}_n}\right] + \frac{M_0\dot{m}}{f\dot{X}_nI_0\left(1 - \frac{M_0\dot{m}}{fI_0\dot{X}_n}\right)^{0.5}} = 0 \quad (16)$$

$$T = \frac{3E_d + E_t - 2E_p}{2R\ln\frac{[-\ln(1-\dot{m})]}{A_p\left(\frac{8f}{A_dA_t}\right)^{0.5}I_0^{0.5}\left[1 - \left(1 - \frac{M_0\dot{m}}{f\dot{X}_n}\right)^{0.5}\right]}} \quad (17)$$

The kinetic constants used in this work are given in the nomenclature section.

DYNAMIC MATRIX CONTROL

DMC is a powerful control algorithm that has been used for the design of industrial controllers. In DMC, the manipulated variable has been adjusted previously, and the controlled variable has been influenced by these adjustments as well as by disturbances. The task of the control algorithm is to determine future adjustments to the manipulated variable so that the controlled variable can return quickly to the set point.

The difference between the predicted values of the controlled variable and the set point is defined as the performance index J (Luyben[8]; Marlin[9]). To minimize J, the sum of the errors squared is calculated

$$J = \sum_{i=1}^{NP}[X^{set}(k+i) - X_{new}(k+i)]^2 + f_t^2\sum_{i=1}^{NC}[\Delta u(k+i)]^2 \quad (18)$$

Where X_{new} is the closed-loop real response for the value at the ith step into the future and it is the sum of past response (X_{past}) and the future changes in the manipulated variable are:

$$X_{new} = X_{past} + \sum_{i=1}^{NC} a_{ik}(\Delta u)^{new} + \underline{d} \quad (19)$$

or

$$X_{new} = X_{past} + \underline{A}\underline{\Delta u} + \underline{d} \quad (20)$$

Here NP denotes the future time over which the control performance is evaluated and termed the output horizon. NC is the number of adjustments and is given by the input horizon. NC must be less than NP.

A is the dynamic matrix and it is composed of the step response coefficients as:

$$A = \begin{bmatrix} a_{11} & a_{12} & \ldots & a_{1,NC} \\ a_{21} & . & & . \\ . & . & & . \\ a_{NP,1} & a_{NP,2} & & a_{NP,NC} \end{bmatrix} \quad (21)$$

To prevent large swings in the manipulated variables, f_t weighting factor is added into the performance index. Increasing f_t increases the damping coefficient of the closed loop system.

X^{set} is the set point and can remain constant at its current value in the future. The goal of perfect controlled-variable performance would be to have zero error for all samples in the future. The solution to this problem is a least-square solution in the form of the following control increments:

$$\underline{\Delta u} = \lfloor\underline{A}^T\underline{A} + f_t^2\underline{I}\rfloor\underline{A}^T(X^{set} - X_{past} - \underline{d}) \quad (22)$$

Where I is the identity matrix, the vector Δu is the NC value of the future changes in the manipulated variables that minimizes the performance index. NP, NC and f_t are used as tuning parameters by the designer.

In the present work, the steps used in the application of the DMC algorithm may be summarized as:

(i) Calculate the NP values of X_{past} from the equation below:

$$X_{past} = X_0^{meas} + \sum_{k=0}^{1-NP}[b_{i+1-k} - b_{i-k}](\Delta u)^{past} \quad (23)$$

(ii) Calculate the NC values of the future changes in the manipulated variables from equation (22) using the dynamic matrix A.

(iii) Implement the first change $(\Delta u)^{new}$.

(iv) Repeat these calculations at the next sampling time to account for changing disturbances and to incorporate feedback.

INTERNAL MODEL CONTROL

The basic idea of IMC is to use a model of the process open-loop $G_m(s)$ transfer function. This model is factored into the product of the two factors

$$G_m(s) = G_m^+(s)G_m^-(s) \quad (24)$$

Here $G_m^+(s)$ is the noninvertible part and $G_m^-(s)$ is the invertible part and has an inverse that is causal and stable, leading to a realizable stable controller. The IMC controller eliminates all elements in the process model $G_m(s)$ that lead to an unrealizable controller by taking the inverse of only the invertible factor to give

$$G_c(s) = [G_m^-(s)]^{-1}f_1 \quad (25)$$

To achieve a good control performance, a filter (f_1) of the feedback signal can be placed before the controller to modulate the manipulated variable behaviour and to increase the robustness of the system. The IMC filter f_1 is described as

$$f_1 = \frac{1}{(\tau_c s + 1)^r} \quad (26)$$

Where τ_c is the desired closed-loop time constant and r is a positive integer. One of the filter forms for plant/model mismatch is

$$f(z) = \frac{1 - a_F}{1 + a_Fz^{-1}} \quad (27)$$

Where $a_F = e^{-\tau}/\tau_f$, τ_f being the time filter constant.

In this study, the dynamic model was assumed to be first order with dead time. The IMC system with a single filter

$$G_m(s) = \frac{K_me^{-d_ms}}{\tau_m s + 1} \quad (28)$$

Table 1. Optimal operating conditions used in experimental studies and in simulation.

m^* (%)	X_n^*	M_0 (mole l^{-1})	I_0 (mole l^{-1})	T_R (°C)	t_f (min)	T_{ci} (°C)	m_c (kg s^{-1})
50	500	6.0898	1.26	103.8	52	21	5×10^{-4}
30	500	6.0898	0.75	114.8	42	21	5×10^{-4}
30	1000	6.0898	0.38	105.5	124	21	5×10^{-4}

$$G_m^-(s) = \frac{K_m}{\tau_m s + 1} \quad \text{and} \quad G_m^+(s) = e^{-d_m s} \tag{29}$$

Here the invertible part is employed in deriving the controller as:

$$G_c = [G_m^-(s)]^{-1} f_1 = \frac{\tau_m s + 1}{K_m} \frac{1}{\tau_c s + 1} \tag{30}$$

In summary, the IMC system execution involves the following steps with three difference equations

1. Calculate the predicted controlled variable, that is, the output from the model $G_m(s)$. The dynamic model can be simulated in discrete form as

$$y_m(k) = K_m(1 - e^{-\tau/\tau_m})u(k - N - 1) + e^{-\tau/\tau_m} y(k - 1) \tag{31}$$

Here τ is the sampling time.

2. Calculate the difference between the measured and model-predicted controlled variables, then find the error signal as

$$e(k) = y_s(k) - [y(k) - y_m(k)] \tag{32}$$

3. Calculate the IMC controller output as

$$\begin{aligned} u(k) = {} & (1 - e^{-t/\tau_f})e(k - 1) \\ & + (e^{-\tau/\tau_m} e^{-\tau/\tau_f} - e^{-t/\tau_m})e(k - 2) \\ & + K_m(1 - e^{-t/\tau_m})e^{-t/\tau_f} u(k - 1) \end{aligned} \tag{33}$$

RESULT AND DISCUSSION

Some theoretical and experimental studies have been carried out to observe the dynamic behaviour of the system and, in addition, model and control parameters were obtained. In the first part of the study, an experimental reaction curve was found by performing the open-loop step test. For this, a styrene-toluene mixture was placed in the reactor. After the reactor was heated by means of an immersed heater, cooling water was passed through the reactor cooling jacket. In this case, the reactor filled with the styrene-toluene mixture was considered as a heat exchanger. The heat given by the heater was absorbed by the cooling water. If this is taken into consideration, the mixing chamber can be considered to be continuous in terms of energy. When such a mixing chamber was used as a polymer reactor, with defined values of heat input and cooling flow rate, the system can reach the steady state condition (Table 1). Therefore, DMC and IMC formulations which are basically developed for processes operating in the vicinity of a steady state can be applied to a batch process. The heat released during the reaction was accepted as a disturbance for the heat exchanger. To obtain the reaction curve, a step test was used. For this, when the system is at the steady state conditions as given in Table 1, a step change is given to the heat input ($\Delta Q = 12.97$ W). The temperature response to this effect is presented in Figure 2. Necessary parameters and a Dynamic Matrix A for DMC, and the representation of the transfer function of the system in equation (28) were obtained from this figure. For all control works, heat input from the immersed heater was selected as a manipulated variable.

In the case of IMC, the first order dead time model was obtained to represent the system behaviour as:

$$G_p = \frac{0.6\, e^{-2.5s}}{30\, s + 1} \tag{34}$$

For this transfer function of the system, the unit of the time constant and dead time are minute. Sample time T was taken as 35.5 sec. The inverse of the Laplace transform for this transfer function was evaluated and then, time variation of the reactor temperature was given in Figure 2.

In the third part of the study, the theoretical simulation work was carried out prior to experimentation. Therefore, the polymerization reactor model was used in optimization and control studies. This work provides considerable experimental economy. In the same way, theoretical step test was evaluated using the reactor model with energy balance and the time response of the theoretical reactor temperature is represented in Figure 2. As can be easily seen from Figure 2, the agreement among the responses of the reactor temperature obtained from the computer simulation program, experimental data and inverse of the transfer function for the polymerization reactor is sufficiently good.

The total simulation programs having monomer, and initiator material balances and equations for the Lagrange's multipliers, were used to calculate the optimum reactor temperature. Desired and optimal values for product quality and operating conditions are given in Table 1.

In the fourth part of the study, experimental and theoretical control systems were evaluated. The best values of control parameters were determined using the total simulation program. Several theoretical systems have been tried to obtain these control parameters using a trial and error method. Then these calculated parameters were used in the

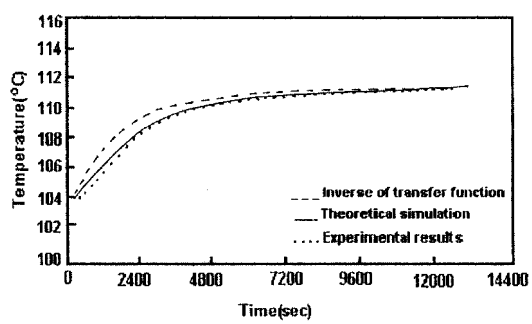


Figure 2. Process reaction curve ($\Delta Q = +12.97W$).

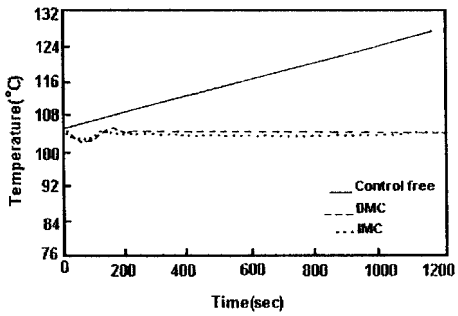


Figure 3. Predicted theoretical temperature response for DMC, IMC and control free (I_0 = 0.0038 mole l^{-1}, T = 105.5°C).

experimental control method. As a result, for a DMC system NP = 4, NC = 1 and f = 0 were found, similarly for an IMC system the IMC filter time constant was selected as 1 minute. Using the optimal values of the control parameters for DMC and IMC, theoretical control results for this process are presented in Figure 3. As can be seen, simulation results show that DMC and IMC techniques are effective ways to control the temperature at a constant value. From these simulation results, product quality which are X_{theo} and m_{theo} were found to be similar to optimal values (X^* = X_{theo} = 500; m^* = m_{theo} = 50).

In the experimental control method, firstly no chemical reaction was carried out in the reactor to provide a convenient basis for the comparison of control performance of the polymerization reactor. For this, the reactor was filled with a styrene and toluene mixture and the system was controlled using DMC and IMC algorithms. Figure 4 shows DMC and IMC control of the reactor temperature for this case. Although there are some small fluctuations at the start of the run, later the controller fully compensates them.

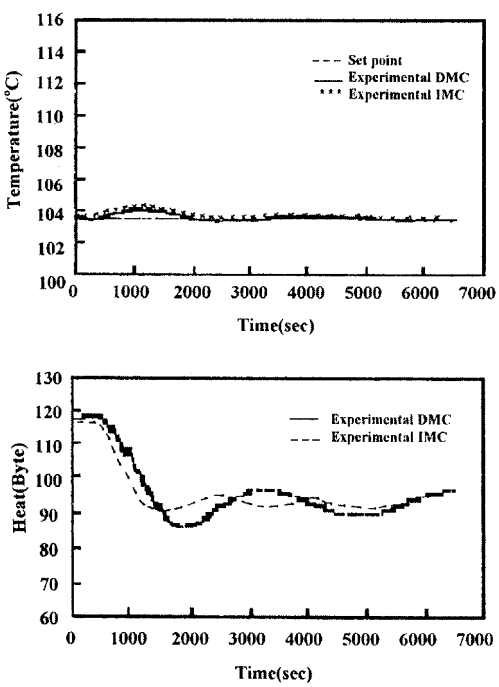


Figure 4. DMC and IMC of temperature of reactor which is filled with styrene and toluene mixture (I_0 = 0.0126 mole l^{-1}, T = 103.8°C).

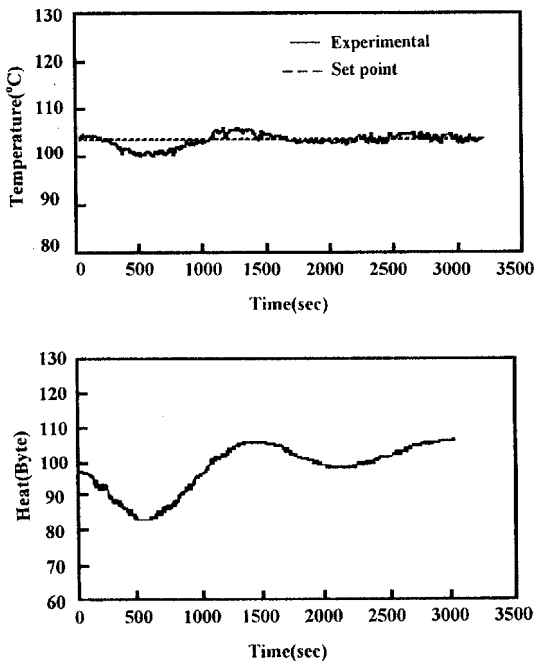


Figure 5. Temperature response and the change of manipulated variable (heat-input) with time under DMC (I_0 = 0.0126 mole l^{-1}, T = 103.8°C).

DMC results of the polymerization reactor at isothermal optimal conditions are given in Figures 5, 6 and 7. These figures represent the temperature and heat (manipulated variable) variations with time at the optimal conditions calculated for three different initiator concentration and different desired properties. There are some fluctuations at the beginning of the reaction for all cases and these fluctuations become more significant as optimal temperature

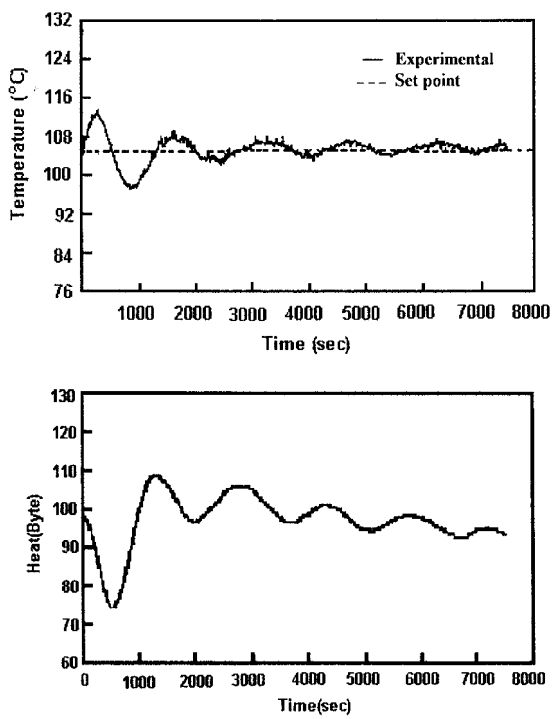


Figure 6. Temperature response and the change of manipulated variable (heat-input) with time under DMC (I_0 = 0.0038 mole l^{-1}, T = 10.5°C).

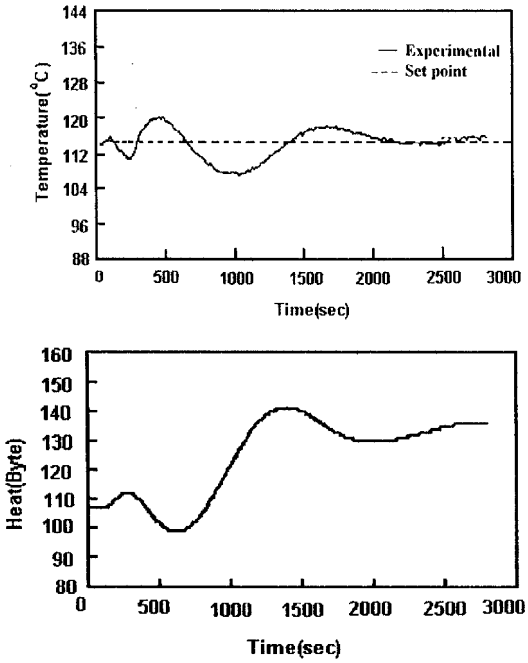


Figure 7. Temperature response and the change of manipulated variable (heat-input) with time under DMC (I_0 = 0.0075 mole l^{-1}, T = 114.8°C).

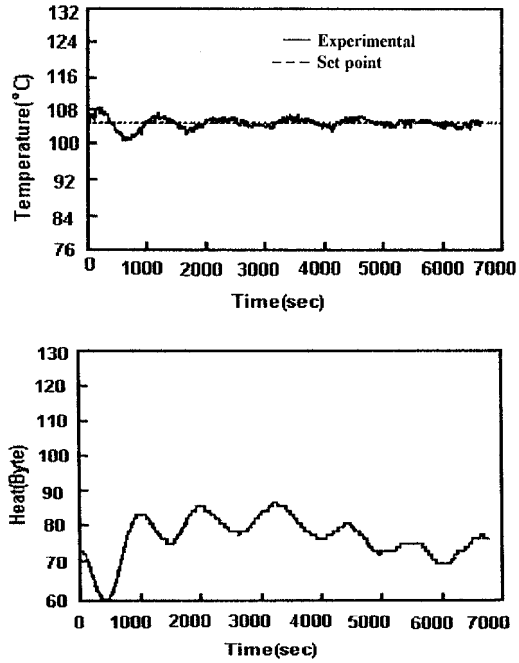


Figure 9. Temperature response and the change of manipulated variable (heat-input) with time under IMC (I_0 = 0.0038 mole l^{-1}, T = 105.5°C).

increases. Heat (Q) variations are given as byte and Q is manipulated in an oscillatory but acceptable fashion, because, the rate of reaction and the rate of heat generation strongly depend on the reaction temperature and initiator concentration. Also, thermal polymerization, which has not been considered in modelling, begins at a high temperature. After DMC experiments were completed, the IMC algorithm was also tested with the same conditions in the same experimental set-up. IMC results for three isothermal temperatures are given in Figures 8, 9, 10. As may be seen, IMC provided as good a performance as the DMC controller. It is effective in damping oscillations but the fluctuations occurred in a higher frequency.

The samples for conversion and molecular weight measurements were taken from the reactor at certain intervals (10 min). Taking samples also created a load effect. After the measurements, the addition of the sample back to the reactor to keep the mixture quantity constant created

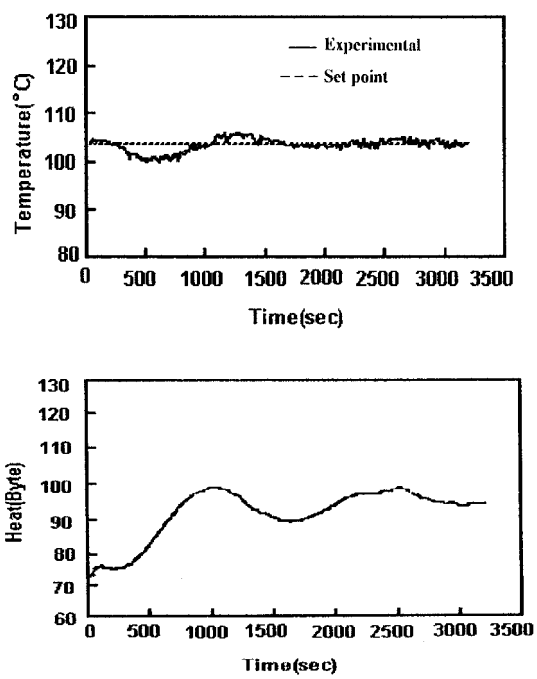


Figure 8. Temperature response and the change of manipulated variable (heat-input) with time under IMC (I_0 = 0.0126 mole l^{-1}, T = 103.8°C).

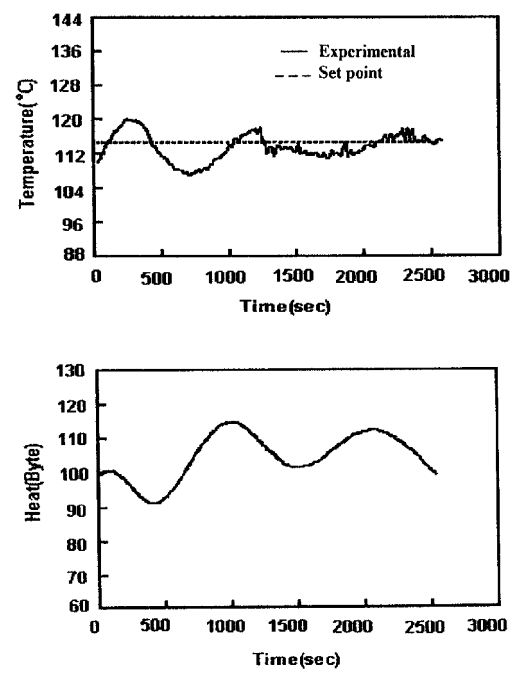


Figure 10. Temperature response and the change of manipulated variable (heat-input) with time under IMC (I_0 = 0.0075 mole l^{-1}, T = 114.8°C).

Table 2. Comparison of experimental results with target values.

T (°C)	103.8	105.5	114.8
I_0 (mole/l)	0.0126	0.0038	0.0075
Desired Conversion, m* (%)	50	30	30
Experimental Conversion DMC, m (%)	52	40	41.3
Experimental Conversion, IMC, m (%)	52.5	42	40.7
Desired Chain Length, X_n^*	*500*	*1000*	*500*
Experimental Chain Length DMC, X_n	488	920	548
Experimental Chain Length IMC, X_n	480	935	539

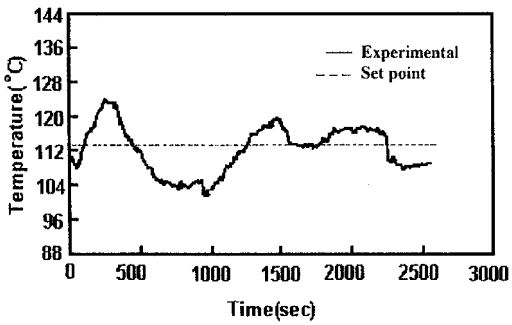


Figure 11. Effect of withdrawing from the reactor (DMC algorithm for

a second load effect. Figure 11 depicts the effect of withdrawing samples from the reactor. Temperature fluctuation around the set point is higher for this case. This shows the high sensitivity of the controlled system. As a result, sample withdrawing affected the control of the polymerization reactor.

In order to see whether the IMC method is better than the DMC method, the experimental conversion and number average chain length values measured at the end of the experiments have also been compared with target values (Table 2). The experimental results are about the same for the IMC and DMC methods. The agreement between target values and experimental results is very good for a 50% conversion ($I_0 = 1.26\times 10^{-2}$ mole l^{-1}, $T = 103.8$°C). This is due to the fact that the temperature variations for both of controllers are about the same. But, the experimental conversion values are higher than the desired values for 30% conversion. Also, X_n values are different from the desired values. These results can be attributed to the long polymerization time (124 min for $I_0 = 0.0038$ mole l^{-1}) and to the thermal polymerization and high reaction rate because of the higher reaction temperature (114.8°C for $I_0 = 0.0075$ mole l^{-1}). Thermal polymerization begins at about 80°C and certain polymerization takes place up to the operation temperature before the initiator was added to the reactor. Solvent evaporation for the polymerization having a long end time could not be avoided during the experiments. As a result, the total volume decreased about 10%. Solvent evaporation may also affect the kinetic and the rate of polymerization. On the other hand, constant volume assumption has been made in the derivation of the optimal policy. This leads to errors in modelling. To compensate the volume effect, monomer, solvent and initiator mixture may be sent continuously to the reactor or a correction factor for the volume should be added to the modelling.

CONCLUSIONS

The DMC system has been proposed and applied to the problem of controlling the temperature in batch polymerization reactors. The performance of the DMC controller was also compared with that of an IMC controller. It was seen that simulation results show excellent performance for DMC and IMC controllers. Because of continuously changing conditions, gel effects and strong nonlinearities, some oscillations in the temperature around the set point in the experiments were seen, but the DMC controller was effective in damping these oscillations. The results of experiments have also shown that DMC and IMC can control temperature well in a polymerization reactor. For both controls, the desired final conversions and the number average chain length values were achieved. Based on these experiments at the laboratory scale, the implementation of a DMC controller at the industrial scale for temperature control of batch polymerization reactors is promising.

NOMENCLATURE

A	heat transfer area, m^2
$\underline{A}$	dynamic matrix vector
A_d, A_p, A_t	frequency factor for initiator decomposition, propagation and termination, sec^{-1}, sec^{-1} $mole^{-1}$
a_1, a_2	variables
c_p, c_{pc}	specific heats for reactor content and coolant respectively, kJ kg^{-1} K^{-1}
d	disturbance effect
d_m	dead time, min
E_d, E_p, E_t	activation energies for initiator decomposition, propagation and termination, cal $mole^{-1}$ K^{-1}
f	initiator efficiency
f_t	detuning parameter for DMC
I, I_0	initiator concentration, initial initiator concentration, mole l^{-1}
J	performance index
G_m	transfer function of process
K_c, K_m	controller gain, model gain
k_d	dissociation rate constant $2.6\times 10^{16}\exp(-34200/RT)$, s^{-1}
k_p	propagation rate constant $1.051\times 10^{7}\exp(-7060/RT)$, l $mole^{-1}$ s^{-1}
k_t	termination rate constant $1.255\times 10^{9}\exp(-1680/RT)$, l $mole^{-1}$ s^{-1}
k_{tc}	termination by combination rate constant, l $mole^{-1}$ s^{-1}
k_{td}	termination by disproportionation rate constant, l $mole^{-1}$ s^{-1}
$-\Delta H$	heat of reaction, kcal $mole^{-1}$
M, M_0	monomer concentration, initial monomer concentration, mole/l
M_r	mass of reactor content, kg
m, $m^{\cdot}$	conversion, desired conversion
m_c	coolant flow rate, kg sec^{-1}
Q	heat given from the heater, W
R_m	reaction rate, mole l^{-1} s^{-1}
T	reactor temperature, °C
T_c, T_{ci}, T_{co}	average, inlet and outlet coolant temperatures, °C
t, t_f	time, polymerization time, min
U	overall heat transfer coefficient, W m^{-2} K^{-1}
u	IMC controller output

V_c	volume of jacket, m^3
v	k_{tc}/k_{td}, constant
y	output variable
y_m	model output
y_s	set point
X_n, X_n^*	number average chain length, desired number average chain length
X_i	measured variable

Greek symbols

λ	Lagrange's multiplier
ρ	coolant density, kg m^{-3}
μ_i	ith moment of dead polymer distribution
μ_r	the viscosity of reacting mixture, cp
τ	sampling time, min
τ_f	filter time constant, min
τ_m	process time constants, min

REFERENCES

1. Cutler, C. R. and Ramaker, B. L., 1980, Dynamic matrix control—a computer control algorithm, *Proc Joint Automatic Control Conf, San Francisco*, paper WP5.
2. Garcia, C. E., Prett, D. M. and Morari, M., 1989, Model predictive control theory and practice—a survey, *Automatica*, 25: 335–348.
3. Peterson, T., Hernandez, Y., Arkun, Y. and Schork, F. J., 1992, A nonlinear DMC algorithm and its application to a semibatch polymerization reactor. *Chem Eng Sci*, 47: 737–753.
4. Sacks, M. E., Lee, S. and Biesenberger, J. A., 1973, Effect of temperature variation on molecular weight distribution: batch, chain addition polymerization, *Chem Eng Sci*, 28: 241–257.
5. Chen, S. and Jeng, W., 1978, Minimum end time policies for batch radical chain polymerization. *Chem Eng Sci*, 33: 735–743.
6. Ray, W. H., 1972, On the mathematical modeling of polymerization reactors, *J-Macromolec Sci Revs Macromol Chem*, 08(1).
7. Karagöz, A. R., Özkan, G., Erdoğan, S. and Alpbaz, M., 1997, Modelling, optimization and control of batch polymerization reactor of styrene, *Control and Computer, IASTED*, 25(1): 21.
8. Luyben, W. L., 1990, *Process Modeling, Simulation and Control for Chemical Engineers*, 2nd Edition, (McGraw Hill, New York).
9. Marlin, T. E., 1995, *Process Control: Design Processes and Control System for Dynamic Performance* (McGraw-Hill, New York).
10. Seborg, D. E., Edgar, T. F. and Mellichamp, D. A., 1989, *Process Dynamics and Control*, (John Wiley and Sons, New York).

ADDRESS

Correspondence concerning this paper should be addressed to Dr S. Yüce, Department of Chemical Engineering, Ankara University, 06100 Tandoğan, Ankara, Turkey. (E-mail: scetinka@scienc e.ankara.edn.tr).

The manuscript was received 22 July 1997 and accepted for publication after revision 29 March 1999.