

Application of Generalized Minimum Variance Algorithm to Polymerization Reactor at a Constant Set Point

Sevil ÇGV PMC¹, A.Rıza KCTCI², Sebahat ETUQ³, Hale HCRQ², Mustafa ANRDC²

¹ Cumhuriyet University, Department of Chemical Engineering, 58140 Sivas, Turkey

² Ankara University, Department of Chemical Engineering, 06100, Ankara, Turkey

³ Gazi University, Department of Chemical Engineering, 06570, Ankara, Turkey

*Corresponding Author

e-mail: cetinkaya.sevil@gmail.com

Received:

Accepted:

ABSTRACT

Generalized Minimum Variance Control (GMV) has been applied to a jacketed batch reactor to control the temperature of a free radical solution polymerization of styrene and its performance was examined. As controller, an on-line digital computer was used to keep the reactor temperature at pre-calculated optimal set point. A Pseudo Random Binary Sequence (PRBS) signal and Bierman algorithm were used to estimate the Auto Regressive Integrated Moving Average (CARMA) model parameters for GMV control system. Heat input to the polymerization reactor was chosen as a manipulated variable. Performance results of GMV were compared experimentally and theoretically and with the results obtained by using conventional deadbeat algorithm. GMV control system shows satisfactory behavior to track the reactor temperature.

KEYWORDS: Generalized Minimum Variance (GMV), mathematical modeling, temperature control, polymerization reactor

INTRODUCTION

Manufacturing a polymer and acquirement of desired polymer product play a key role in a polymer industry. In practice, batch processes are often used to produce high quality polymer. In a polymer process, one of the most encounter drawback when producing desired polymer is heat releasing during process and it must be removed from reactor in order to allow the reactor temperature to catch be stable and to reach desired monomer conversion, chain length and average viscosity molecular weight [1,2].

In recent years, there has been a growing emphasis on control of batch chemical and polymerization reactors. Many industrial applications and academic studies have appeared in the literature [3,4]. Two example of industrial application has been illustrated by Seki et al. (2001). One of them is a slurry phase polypropylene semi-batch reactor control and the other is a slurry phase high density polyethylene continuous stirred tank reactor control. Either application nonlinear model predictive control algorithm (NMPC) was used. In the first application, the NMPC successfully prevented thermal runaway of the reactor temperature control and in the second application; the NMPC improved the closed loop performance during the grade change over operation.

The control of batch chemical reactors is important for the quality of the product and yield. Conventional controller such as deadbeat is applied to this type of systems [6-10]. Here, another important factor is the performance of the controllers. Product quality and profitability should be satisfied. Two quantitative methods for evaluating closed-loop control performance are used mostly [11]. The first one is frequency response, which determines the response of important variables in the control system to sine function of either the disturbance or set point. The second method is simulation, which is effective in giving the entire transient response to important changes in the forcing functions.

During the last ten years, model predictive control is used to improve the control performance of batch polymerization reactor. The general strategy of predictive controllers is to utilize a discrete model to predict the future output and then compute the control action [12-14]. In this way, the difference between the set point and the predicted output is minimized.

The Generalized Minimum Variance (GMV) controller was formed basically as a modification of the Minimum Variance (MV) technique of Aström and Wittenmark [15, 16]. This is known as a one-step-ahead optimal control law. The technique is generally to hold weighting coefficient as small as possible in order to keep it as close as required to the objective of sustaining a minimum output variance while still preserving closed loop stability.

In this work, experimental and theoretical applications of GMV controller were accomplished in a polymerization reactor. In the first part of the work, theoretical simulation program having mass and energy balance was realized to observe the dynamic behavior of the reactor. This program was performed to calculate the model and control parameters. In the second part, second order CARMA model for GMV system was developed between the heat input and the reactor temperature. Parameters of this model were calculated by using PRBS signal and Bierman algorithm. For this calculation, total simulation program was used. GMV control system was tried to keep the reactor temperature at optimal values. Tuning GMV control parameters were optimized by applying total simulation program and then control parameters were used in experimental control work. The performance of this control system was tested and compared with theoretical GMV and Deadbeat results.

MATERIALS and METHODS

Modeling and Optimization

Free radical polymerization reactors can be modeled in terms of a set of differential equations. Many modeling studies on the polymerization of styrene in a batch reactor have been reported in the literature [17]. To simplify the mathematical description of the system, constant density, no-chain transfer, quasi steady state and long-chain approximations for live radicals have been assumed. Based on these assumptions, the modeling equations can be

written as follows

$$\frac{dI}{dt} = -k_d I \quad (1)$$

$$\frac{dM}{dt} = -k_p \left(\frac{2fk_d}{k_t} \right)^{0.5} M I^{1/2} \quad (2)$$

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

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

$$M(0) = M_0, \quad I(0) = I_0, \quad \mu_0(0) = \mu_2(0) = 0$$

with initial conditions,

The following energy balances for the reactor and cooling jacket can express the reactor dynamic

$$\frac{dT}{dt} = \frac{Q_w}{V\rho C_p} + \frac{(-\Delta H)R_m}{\rho C_p} - \frac{UA(T - \bar{T}_c)}{V\rho C_p} \quad (5)$$

$$\frac{dT_c}{dt} = \frac{m_c(T_{ci} - T_{co})}{V_c \rho_c} + \frac{UA(T - \bar{T}_c)}{V_c \rho_c C_{pc}} \quad (6)$$

The overall heat transfer coefficient for the polymerization reactor was modeled as a function of the viscosity of the reacting mixture [18].

$$U = \frac{1}{\mu_r^{0.33} S + F} \quad (7)$$

Here, S and F are constants. Viscosity was measured on-line during the experiments. From the solutions of equation (1-3), the following equations were obtained

$$I = I_0 \exp(-k_d t) \quad (8)$$

$$\ln \frac{M}{M_0} = \frac{2k_p}{k_d} \left(\frac{2fk_d I_0}{k_t} \right)^{0.5} \left[\exp\left(-\frac{k_d t}{2}\right) - 1 \right] \quad (9)$$

$$\mu_0 = 2f \left(1 - \frac{\nu}{2} \right) I_0 [1 - \exp(-k_d t)] \quad (10)$$

The time required to reach the predetermined conversion m^* and number average chain length X_n^* can be obtained by solving eqn.(10) for $t = t_f$.

$$t_f(T, I_0) = -\frac{1}{A_d e^{-E_d/RT}} \ln \left[1 - \frac{M_0 m^*}{f I_0 X_n^*} \right] \quad (11)$$

Substituting eqn. (11) into eqn. (9) at $t = t_f$ gives the following equation:

$$g(T, I_0) = -\ln(1 - m^*) + 2A_p \left(\frac{2f}{A_d A_t} \right)^{0.5} \exp \left[-\frac{1}{2RT} (2E_p - E_d E_t) \right] \left[\left(1 - \frac{M_0 m^*}{f I_0 X_n^*} \right)^{0.5} - 1 \right] = 0$$

(12)

Here, m^* and X_n^* are desired conversion and desired number average chain length, respectively. The kinetic constants and physical parameters were given in Table 1. The control variables in this system were taken as reactor temperature and initial initiator concentration. The best reaction temperature and initial initiator concentration to achieve a given predetermined conversion and number average chain length in a minimum time can be obtained by use of Lagrange's Multiplier [17]. According to this method, the optimal T and I_0 can be obtained by optimizing the following function:

$$W = t_f(T, I_0) + \lambda g(T, I_0) \quad (13)$$

Here, λ is the Lagrange's multiplier. For this system, the partial derivative of W with I_0 and T respectively must be zero, i.e:

$$\left(\frac{\partial W}{\partial I_0}\right)_T = \left(\frac{\partial W}{\partial T}\right)_{I_0} = 0 \quad (14)$$

From the solution of equation (11), (12), (13) and (14), the following equations for optimal T and I_0 have been obtained:

$$T = \frac{E_d + E_t - 2E_p}{2R \ln \frac{[-\ln(1-m^*)]}{A_p \left(\frac{8fI_0}{A_d A_t}\right)^{0.5} \left[1 - \left(1 - \frac{M_0 m^*}{fX_n^*}\right)^{0.5}\right]}} \quad (15)$$

$$\frac{E_d}{(3E_d + E_t) - 2E_p} \ln \left[1 - \frac{M_0 m^*}{fI_0 X_n^*}\right] + \frac{M_0 m^*}{fX_n^* \left[\frac{M_0 m^*}{fI_0 X_n^*}\right]^{0.5}} = 0 \quad (16)$$

Table1. The kinetic constants and physical parameters used in experimental studies [12].

Constant		
A_p	1.051×10^9	$l \text{ mol } s^{-1}$
A_t	1.255×10^9	$l \text{ mol } s^{-1}$
A_d	2.6×10^{16}	$cal \text{ mol } K^{-1}$
E_p	7060	$cal \text{ mol } K^{-1}$
E_t	1680	$cal \text{ mol } K^{-1}$
E_d	34200	$mol \text{ l}^{-1}$
M_0	6.092	$g \text{ mol}^{-1}$
f	0.5	

The calculated optimal temperature and initiator concentration values from these equations were implemented on the experimental reactor system.

The Generalized Minimum Variance Control

GMV controller was formed basically as a modification of the minimum variance technique of Aström and Wittenmark [15].

In MV control it is critical that the time delay integer k be correctly chosen. Wrong values of the system delay can destabilize the controller by the polynomial Q . The presence of this usually avoids problems of destabilizing effect of wrong choice of the supposed system delay. The GMV algorithm produces good set point following characteristics and is able, to a certain extent, to control non-minimum phase systems. This GMV algorithm is implicit, i.e. the evaluated parameters are employed precisely in the control law computation. However, due to the implicit character of this algorithm it can be shown that the controller design parameters cannot be varied on-line without degrading the parameter estimates [19]. An additional modification to this design permits the control weighting to be altered on-line without influencing the parameter estimates [20]. In the latter case, the control law is not completely implicit.

The GMV approach employs a system pseudo-output $\phi(t)$ determined by

$$\phi(t+k) = Py(t+k) + Qu(t) - Rr(t) \quad (17)$$

Where $r(t)$ is the set point, and P , Q and R are transfer functions in the backward shift operator z^{-1} , specified as;

$$P=1, Q=\lambda, R=1 \quad (18)$$

The function of the feed forward term is to change the system open loop zeros from B to $PB+QA$, this can be seen by substituting the system CARMA model equation (19) in equation (20);

$$y(t+k) = \frac{C}{A} e(t+k) + \frac{B}{A} u(t) \quad (19)$$

$$\phi(t+k) = \frac{PB+QA}{A} u(t) + \frac{PC}{A} e(t+k) - Rr(t) \quad (20)$$

The polynomials are written as:

$$A = 1 + a_1 z^{-1} + \dots + a_{n_a} z^{-n_a} \quad (21)$$

$$B = b_0 + b_1 z^{-1} + \dots + b_{n_b} z^{-n_b} \quad (22)$$

$$C = 1 + c_1 z^{-1} + \dots + c_{n_c} z^{-n_c} \quad (23)$$

$$P = 1 + p_1 z^{-1} + \dots + p_{n_p} z^{-n_p} \quad (24)$$

The cost function to be minimized is then the variation of the pseudo-output

$$J = E \{ \phi^2(t+k) \} \quad (25)$$

This expression for $\phi(t+k)$ is the addition of two independent terms. The first term can be defined as:

$$\phi(t+k/t) = \frac{1}{C} [(BE + QO)u(t) + Gy(t) - CR(t) + Ed] \quad (26)$$

$$G = g_0 + g_1 z^{-1} + \dots + g_{n_g} z^{-n_g} \quad (27)$$

and represents the best forecast of $\phi(t+k/t)$ established on data up to time t . The second term is:

$$Ee(t+k) = \phi(t+k) - \phi(t+k/t) \quad (28)$$

Clearly J is minimized by setting the predicted output equal to zero ie:

$$\phi(t+k/t) = 0 \quad (29)$$

This gives the control law

$$Fu(t) + Gy(t) - Hr(t) + Ed = 0 \quad (30)$$

Where,

$$F = BE + QC \quad (31)$$

$$H = CR \quad (32)$$

Hence:

$$u(t) = (Hr(t) - Gy(t) - Ed) / F \quad (33)$$

The steps in the implementation of the GMV algorithm may be summarized as:

a. Apply a PRBS to the system as a forcing function and obtain the plant output

b. Estimate F , G , H from equation (30) implementing the Bierman U-D update Algorithm[21]

c. Employ equation (33) to evaluate the control signal

d. Apply the control signal

e. Return to (a)

Deadbeat Controller (DB)

The purpose of conventional feedback control is to attain and maintain a desired condition by using the difference between the measured output and the set point in order to remove as far as possible any difference between them. When the fixed parameters of the controller have been determined, sufficient and effective control is usually attained although detuning may be required to make sure that stability and non-oscillatory behavior is maintained over the whole range of operating conditions. Deadbeat controller was used for comparison in the present work.

Deadbeat control action can be described quantitatively by:

$$D(z) = \frac{1}{HG_p(z)} \left[\frac{\hat{y}(z)/\hat{y}_{sp}(z)}{1 - \hat{y}(z)/\hat{y}_{sp}(z)} \right] \quad (34)$$

Where the unit step change in the set point is given as:

$$\hat{y}_{sp}(z) = \frac{1}{1 - z^{-1}} \quad (35)$$

The response with a unit step delayed by one sampling instant

$$\hat{y}(z) = \frac{z^{-1}}{1 - z^{-1}} \quad (36)$$

Therefore,

$$\frac{\hat{y}(z)}{\hat{y}_{sp}(z)} = z^{-1} \quad (37)$$

Transfer function for a Deadbeat controller

$$D(z) = \frac{1}{HG_p(z)} \frac{z^{-1}}{1 - z^{-1}} \quad (38)$$

and

$$HG_p(z) = \frac{b_0 z^{-1}}{1 + a_1 z^{-1} + a_2 z^{-2}} \quad (39)$$

Therefore the discrete time version of deadbeat action can be given as:

$$\frac{C(z)}{\varepsilon(z)} = \frac{1 + a_1 z^{-1} + a_2 z^{-2}}{b_0 (1 - z^{-1})} \quad (40)$$

$$C_n - C_{n-1} = \left[\frac{1}{b_0} \right] \varepsilon_n + \left[\frac{a_1}{b_0} \right] \varepsilon_{n-1} + \left[\frac{a_2}{b_0} \right] \varepsilon_{n-2} \quad (41)$$

EXPERIMENTAL

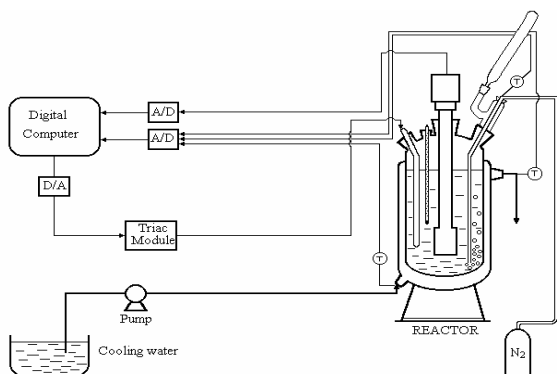


Figure 1. Experimental System

Polymerization experiments were carried out in a cylindrical glass jacketed reactor of 1100 ml. with a dished bottom equipped with a heater, thermocouples and condenser. Temperatures detected by thermocouples were transferred into a computer through A/D converters. The system was purged with nitrogen sent from the bottom of the reactor. A reflux condenser was used to regain the vaporized toluene. A thermostat through the cooling jacket pumped the coolant and its flow rate was set at a fixed value. A thyristor unit depending upon the signals from the computer manipulated the power of the heater inside the reactor. The temperatures and viscosity values

were monitored on the computer during the polymerization as shown in Fig. 1.

Styrene was vacuum distilled and benzoyl peroxide initiator was recrystallised using chloroform methanol mixture before being used.

The reactor was first charged with styrene and toluene solvent (30 % in volume). Then, the reaction mixture was heated to the desired temperature and pouring in the initiator solution started the polymerization. In all experiments, the optimum reaction temperature calculated before was tried to track by implementing GMV control method.

Next, at intervals of 15 min, 5ml. samples were taken from the reactor for the off-line analysis. The amount of monomer conversion and the number average chain length in each sample were determined by use of precipitation method and Ubbelohde viscometer respectively.

RESULTS

In this work, GMV control of polymerization reactor temperature has been realized. Experimental and theoretical applications were achieved to obtain the desired comparison. In the design of this control system, the reactor filled with styrene-toluene mixture is considered as a heat exchanger. When the reactor is heated by means of an immersed heater, cooling water is passed through the reactor-cooling jacket. So the cooling water absorbs the heat given out by the heater. If this is taken into consideration, this reactor can be considered to be continuous as regards to energy. When such a mixing chamber was used as a polymerization reactor with defined values of heat input and cooling flow rate, system can reach the steady-state condition as shown in Table 2. The design philosophy of GMV control system basically depends on this approximation. In addition, the heat released during the reaction was accepted as a disturbance for the heat exchanger. Heat input from the immersed heater is chosen as a manipulated variable.

For GMV control work, heat exchanger approximation was used. In design calculations, mathematical model equations of energy balance of the heat exchanger Eqs. (5) and (6) were solved using 4th order Runge-Kutta integration method. This simulation algorithm with initial steady-state

Table 2. Optimal operating conditions used in experimental studies and simulation

m^* (%)	X_n^*	M_0 (mole l^{-1})	I_0 (mole l^{-1})	T ($^{\circ}C$)	t_r (min)	T_{ci} ($^{\circ}C$)	m_c , (kg s^{-1})
50	500	6.0898	0.0126	103.8	152	21	5.10^{-4}
30	1000	6.0898	0.0038	105.5	124	21	5.10^{-4}

conditions shown in Table 2 was used to calculate the system model and control parameters.

A constant optimum reactor temperature for a minimum polymerization time was calculated by applying a total simulation program having energy balance, monomer and initiator material balances and Lagrange's multiplier. Theoretical, desired and optimal values for product quality and operating condition are given in Table 2. Two sets of optimal operating conditions were used for this work.

This work has been done for GMV control system. Standard CARMA model based on the relation between the reactor temperature and heat input to the reactor is used. The parameters of CARMA model were calculated by using simulation program having energy balances with initial steady-state conditions. When the reactor filled with the mixture of toluene and styrene is in the steady-state conditions, PRBS signal is given to disturb the system around the reactor temperature theoretically [22]. The identified CARMA model of the reactor was given below

$$y(t) = 0.12024 \cdot 10^{-7} u(t-1) - 0.1y(t-1) - 148997 \cdot 10^{-3} y(t-2) \quad (42)$$

In all GMV control work, this identified CARMA model was used. GMV control system based on this model has been applied to keep the reactor temperature at the optimal value. Using total simulation program, suitable control parameters, which are time delay and control weighting, were determined by observing control performance. After trying several calculations these parameters are taken as 1 min. and 0.95 respectively. In all experimental and theoretical studies, GMV control system with identified model and control parameters were applied to the polymerization reactor.

Firstly GMV control system was applied to track the reactor temperature with optimal value as $I=0.0126$ mole l^{-1} , $T=103.8$ $^{\circ}C$. Theoretical and experimental results are presented in Fig. 2. As it can be seen from this figure, agreement between the theoretical and experimental results is very satisfactory. But the reactor temperature gives some oscillations before reach the steady value. The results of GMV and Deadbeat control applied to the other optimal temperature value ($I=0.038$ mole l^{-1} , $T=105.5$ $^{\circ}C$) are given in Figure 3. This figure shows that reasonable agreement was obtained between experimental and theoretical results.

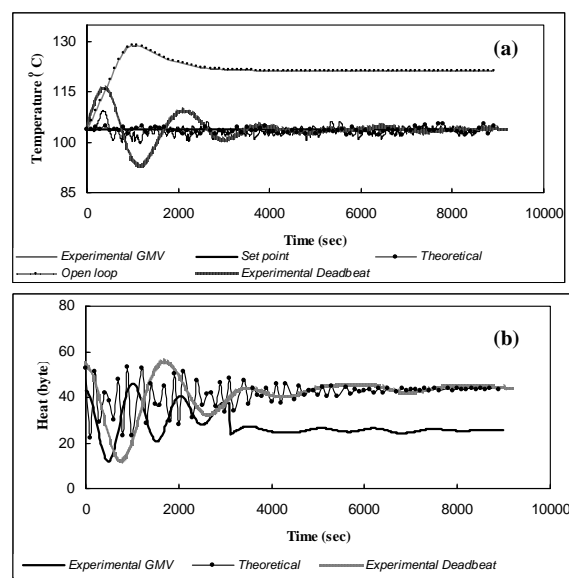


Figure 2. (a) Experimental and theoretical GMV and Deadbeat [12] control of the reactor temperature (b) The change of manipulated variable (heat-input) with time ($I_0=0.0126$ mole / l, $T=103.8$ $^{\circ}C$)

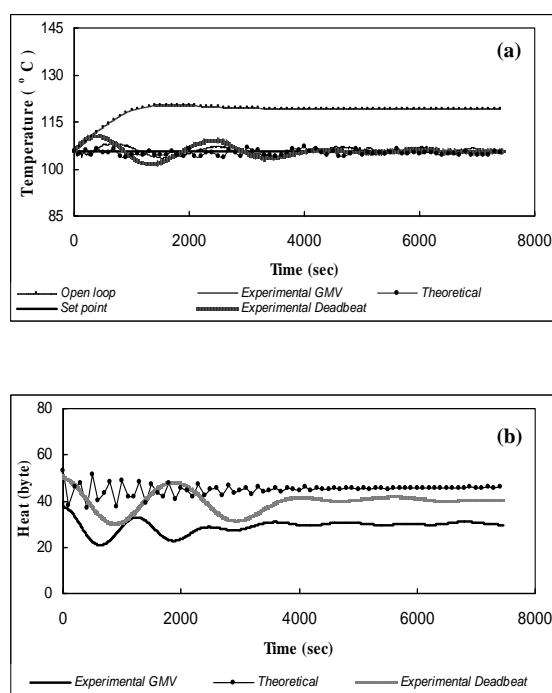
Table 3. Experimental conversion and chain lengths for GMV and DB control systems

T (°C)	103.8	105.5
I ₀ (mole l ⁻¹)	0.0126	0.0038
Desired conversion, m* (%)	50	30
Desired chain length, X _n *	500	1000
Experimental conversion, m _e (%) (GMV)	54.84	31.30
Experimental chain length, X _n (GMV)	465	516
Experimental conversion, m _e (%) (DB) [12]	57.1	32.1
Experimental chain length, X _n (DB) [12]	405.65	876

In the operating conditions of I₀=0.0126 mole l⁻¹, T=103.8 °C, t_f=152 min., m*=0.5 and I₀=0.0038 mole/l, T=105.5 °C, t_f=124 min., m*=0.3, the GMV control shows satisfactory performance. These figures (Fig. 2, 3) depict also the computer temperature profiles under control-free conditions. The temperature increase or reaction rate comes to a maximum at about 1000 sec. for the first experiment (T=103.8 °C). Up to this point, there are some undesirable oscillations and then controller performance increases. Heat variations are also given. Q is manipulated in an oscillatory but acceptable fashion.

Deadbeat control of the system was utilized to compare the efficiency of GMV methods. Deadbeat control parameters of this system were used as b₀=7.0211x10⁻⁸, a₁=1.12x10⁻⁷, a₂=-8.98x10⁻⁸ [12]. Reactor temperature closely follows the optimal trajectory. But, the response produce larger over shoot and more oscillations as it is expected. GMV yields a more moderate controlled response. In both of the control algorithms, control action produced is reducing as the temperature is increasing.

The conversion and number average chain length values measured at the end of the polymerization are given Table 3 in order to see the performance of these methods. The agreement between target values and experimental results is better under GMV control than that under Deadbeat control.

**Figure 3. (a)** Experimental and theoretical GMV and Deadbeat [12] control of the reactor temperature

(b) The change of manipulated variable (heat-input) with time (I₀=0.0038 mole / l, T=105.5 °C)

CONCLUSION

A GMV algorithm was implemented experimentally and theoretically to track the temperature on a set point in a batch jacketed polymerization reactor. Standard CARMA model based on the relation between the reactor temperature and heat input to the reactor was used. The performance of the GMV controller was compared with the results acquired using STPID. The results showed that the performance of GMV controllers was better than that of STPID. In control systems, conversion and molecular weight were taken as optimal values. Due to modeling and experimental errors, some deviations in number average chain lengths were observed. From the experimental results, it is foremost that suggested control system indicates good performance for processes with model-plant mismatch.

NOMENCLATURE

A	:heat transfer area (m ²)
A _d , A _p , A _t	:frequency factor for initiator decomposition, propagation and termination, respectively
A (z ⁻¹)	:monic polynomial in z-domain representing the poles of the discrete-time system
a _i	: parameters of A polynomial
B (z ⁻¹)	:polynomial in z-domain representing the zeros of the discrete-time
b _i	: parameters of A polynomial
C (z ⁻¹)	:monic polynomial in z-domain representing the zeros of the process noise, calculated control output for Deadbeat algorithm
C _p , C _{pc}	:specific heat of reactor content and coolant (J kmol ⁻¹ °C ⁻¹)
d	:offset
e(t)	:white noise
E _d , E _p , E _t	:activation energies for initiator decomposition, propagation and termination, respectively
f	:initiator efficiency
-ΔH	:heat of reaction (J kmol ⁻¹)
I, I ₀	:initiator concentration, initial initiator concentration (mole l ⁻¹)
k _d , k _p	:kinetic constants for initiator decomposition and propagation, respectively
k _t	:kinetic constant for termination

m _c	:flow rate of the cooling water (kg s ⁻¹)
M, M ₀	:monomer concentration, initial monomer concentration (mole l ⁻¹)
m [*]	:desired monomer conversion (%)
Q	:weighting polynomial acting on control input
Q _w	:heat power (W)
r(t)	:set-point at time t
R (z ⁻¹)	:weighting polynomial acting on set-point
T	:temperature (°C)
T _{ci}	:coolant inlet temperature (°C)
T _{co}	:coolant outlet temperature (°C)
t	:time (s)
t _f	:reaction time (min)
U	: overall heat transfer coefficient (kcal m ⁻² K ⁻¹ s ⁻¹)
u(t)	:input variable at time t
v	:constant
V	: volume of the mixture (m ³)
V _c	: volume of the cooling water jacket (m ³)
X _n , X _n [*]	: number of average chain length, desired number of average chain length respectively
y(t)	:output variable at time t
z, z ⁻¹	:forward and backward shift operators
ρ, ρ _c	: density of the mixture, density of the cooling water (kg m ⁻³)
μ ₀ , μ _r	:initial viscosity of reactor content, viscosity of reactor content (cp)
λ	:control weighting
φ	:sum of the root square of the parameters
ε	: Error
φ(t + k/t)	: Best forecast of
φ(t + k)	established on data up to time t

REFERENCES

- [1] Yüce S, Hasaltun A, Erdoğan S. and Alpbaz M., 1999. Temperature Control of a Batch Polymerization Reactor. Trans IChemE, 77 (A): 413-419.
- [2] Çetinkaya S, Zeybek Z, Hapoğlu H. and Alpbaz M., 2006. Optimal Temperature Control in a Batch Polymerization Reactor Using Fuzzy-Relational Models-Dynamics Matrix Control. Computers and Chemical Engineering, 30: 1315-1323.

- [3] Seki H, Ogawa M, Ooyama S, Akamatsu K, Ohshima M, Yang W., 2001. Industrial Application of a Nonlinear Model Predictive Control to Polymerization Reactors. *Control Engineering Practice*. 9: 819-828.
- [4] Vieira R.A.M, Embiruçu M, Sayer C, Pinto J.C, Lima E.L., 2003. Control Strategies for Complex Chemical Process. Applications in Polymerization Process. *Computer & Chemical Engineering*. 27: 1307-1327.
- [5] Richards J.R, Congalidis J.P., 2006. Measurement and Control of Polymerization Reactors. *Computer & Chemical Engineering*. 30: 1447-1463.
- [6] Linnemann A., 1995. An Algorithm to Compute State Feedback Matrices for Multi-Input Deadbeat Control. *Systems & Control Letters*. 25: 99-102.
- [7] Nešić D., 1997. A Note on Dead-Beat Controllability of Generalised Hammerstein Systems. *Systems & Control Letters*. 29: 223-231.
- [8] Chang J., 1998. Deadbeat Control with Disturbance Rejection. *Systems & Control Letters*. 33: 253-258.
- [9] Verde C., 1999. On the Use of Sensitivity Functions to Design Dead-beat Control Algorithm. *Automatica*. 35: 945-949.
- [10] Valcher M.E, Willems J.C., 1999. Dead beat Observer Synthesis. *Systems & Control Letters*. 37: 285-292.
- [11] Marlin E.T., 1995. *Process Control, Designing Process and Control Systems for Dynamic Performance*, Mc Graw Hill, New York, 3 rd Chap. 5: 121-126.
- [12] Yüce (Çetinkaya) S., 2001. Nonlinear Model Predictive Control of Reactor Temperature in Agitated Batch Polymerization Reactor Operating Optimal Condition. Ph.D Thesis, University of Ankara, Turkey.
- [13] Zeybek Z, Çetinkaya S, Hapoğlu H, Alpbaz M., 2006. Generalized Delta Rule (GDR) Algorithm with Generalized Predictive Control (GPC) for Optimum Temperature Tracking of Batch Polymerization. *Chemical Engineering Science*, 61: 6691-6700.
- [14] Shafiee G, Arefi M.M, Jahed-Motlagh M.R, Jalali A.A., 2008. Nonlinear Predictive Control of a Polymerization Reactor Based on Piecewise Linear Wiener Model. *Chemical Engineering Journal*, 143: 282-292.
- [15] Aström K.J. and Wittenmark B., 1973. On Self Tuning Regulators. *Automatica*, 9: 185-189.
- [16] Aström K.J, Borrisson U, Ljung L. and Wittenmark B., 1977. Theory and Applications of Self-Tuning Regulators. *Automatica*, 13: 457-476.
- [17] Chen S-A. and Jeng W-F., 1978. Minimum and Time Policies for Batchwise Radical Chain Polymerization. *Chemical Engineering Science*, 33: 735-743.
- [18] Güzel T, Hasan A, Karagöz A.R, Erdoğan S, Alpbaz M, Saçak M., 1993. Stirenin kesikli soğutma ceketli polimer reaktöründe radikalik polimerleşme dinamiği. *Doğa (Türk Mühendislik ve Çevre Bilimleri Dergisi)*, 17 (2): 81-90.
- [19] Clarke D.W. and Gawthrop, P.J., 1975. Self Tuning Controller. *Proc. IEE*. 122(9): 929-934.
- [20] Clarke D.W. and Gawthrop P.J., 1975. Self Tuning Controller. *Proc. IEE*. 126(6): 633-640.
- [21] Bierman, G.J., 1976. Measurement Updating Using the U-D Factorization. *Automatica*, 12: 375-382.
- [22] Karagöz A.R, Hapoğlu H. and Alpbaz M., 2000. Generalized Minimum Variance Control of Optimal Temperature Profiles in Polystyrene Polymerization Reactor. *Chemical Engineering and Processing*. 39: 253- 262.