APPLICATION OF NONLINEAR MODEL PREDICTIVE CONTROL BASED ON DIFFERENT MODELS TO BATCH POLYMERIZATION REACTOR

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ABSTRACT

In the present work, at the previously determined optimal conditions, to control batch polymerization reactor, Linear Generalized Predictive Control (LGPC) and Nonlinear Generalized Predictive Control (NLGPC) algorithm were utilized. Several system models were applied to the control algorithms. The effect of different optimal conditions has been examined on monomer conversion, average viscosity molecular weight and chain length. At the same operating conditions of LGPC and NLGPC temperature control was used for comparison. According to the experimental results, the performance of NLGPC was obtained well than LGPC control method. In addition, the results denoted that the NLGPC control performances depend on different models and the optimum conditions.

Keywords: Styrene polymerization, Generalized Predictive Control, Nonlinear Model Predictive Control

1. INTRODUCTION

Control of polymerization reactors is often difficult and sensors to provide on-line measurement of polymer properties are generally not available (Altınten, Erdogan, Hapoglu, and Alpbaz 2003; Altınten, Erdogan, Hapoglu, Alıev, and Alpbaz 2006; Cetinkaya 1996). The most significant task for a polymerization reactor control strategy is to maintain the major design because of having complex and nonlinear reaction and operational variables like product quality is also important to preserve smooth and stable operation.

Physical, chemical and mechanical properties of polymers are generally closely related with their molecular weights. But the weight of all polymer molecules within a polymer sample is not equal to each other. For this reason, the molecular weight of polymers that were determined in any way shows average number not the exact values. The full molecular weight distribution (MWD) of a specific polymer and ratio of moments of this distribution, such as the number average ($\overline{M}_n$) or the weight average ($\overline{M}_w$) molecular weight, indicate the mechanical properties of the polymer. Initial initiator concentration and temperature are the primary control ways to influence the molecular weight of a polymer produced in free radical polymerization (Barner-Kowollik and Davis 2001). The temperature change has been observed that has great influence on the kinetics of polymerization process, and physical properties and quality of produced polymer. The main objective of the temperature control of polymerization reactor is to remove a great amount of heat from the exothermic reaction to achieve the desired number average chain and a desired conversion in a minimum time (Yuce, Hasaltun, Erdogan, and Alpbaz 1999). Then, reactor temperature should be controlled effectively to satisfy the desired polymer quality.

Various control methods have been applied both theoretically and experimentally to the systems at constant and changing set points (Zeybek, Cetinkaya, Hapoglu, and Alpbaz 2006; Seki, Ogawa, Ooyama, Akamatu, Ohshima, and Yang 2001). Lewis, Nguyen, and Cohen (2007), highlighted to the effect of initiator amount on the radical polymerization. In free-radical polymerization, the reaction temperature and both the initiator and the chain-transfer agent's concentrations are usually chosen as controlled variables. These variables can also affect the rate of polymerization and the molecular weight of the polymer (Ponnuswamy, Shah, and Kiparissides 1987).

Generalized Predictive Control (GPC) algorithm is commonly used in polymerization reactors (Ozkan, Hapoglu, and Alpbaz 1998; Yuce (Çetinkaya) 2001). But, NLGPC algorithm was rarely practiced with different models and at constant temperature in a batch polymerization reactor. Zeybek, Cetinkaya, Hapoglu, and Alpbaz (2006) developed the generalized delta rule

(GDR) algorithm with generalized predictive control (GPC) for two different changing temperature path, and used experimentally in a batch polymerization reactor. The application of a GDR for system identification is a feasible alternative when model equations are not known or only historical input-output data are available.

In this study, Linear Generalized Predictive Control (LGPC) and Nonlinear Generalized Predictive Control (NLGPC) algorithm were used. Two models of NLGPC were applied to the control algorithms and compared with LGPC and both of constant and changing temperatures. According to the experimental results obtained at the constant and changing set points, the performance of NLGPC in terms of efficiency were also obtained better than LGPC control method. Desired monomer conversion, average viscosity molecular weight and chain length were affected by the controller performance. In addition, the results denoted that the NLGPC control performances depend on different models and the optimum conditions.

2. MATHEMATICAL MODEL OF THE REACTOR

In polymerization, physical, chemical and mechanical properties are based on polymer quality of the final product, which means molecular and structural characteristics of a polymer. The ability of a mathematical model is important to predict exact molecular properties of a polymer manufactured by means of polymerization reactor for optimal production cost in polymer industry.

A basic free radical polymerization mechanism has three fundamental reaction steps: initiation, propagation and termination.

Considering the standard free radical polymerization and assuming constant density, no chain transfer and no gel effects, and using quasi steady-state and long chain approaches for live radials, the equations for monomer conversion, initiator conversion and the dimensionless zeroth moment of the molecular weight distribution are used as follows (Chen and Jeng 1978):

$$\frac{dX}{dt} = k_1 (1-c)^{0.5} \frac{(1-x)}{g} \tag{1}$$

$$\frac{dc}{dt} = k_d (1-c) \tag{2}$$

$$\frac{dq_0}{dt} = (2-c)\alpha k_d (1-c) \tag{3}$$

With the initial conditions, $X(0) = c(0) = q_0 = 0$

$$k_1 = k_p \left(2 f k_d I_0 / k_t \right)^{1/2} = I_0^{1/2} A_1 \exp(-E_1 y) \tag{4}$$

$$X = 1 - \frac{M}{M_0}; \quad c = 1 - \frac{I}{I_0}; \quad g = 1;$$

$$\alpha = \frac{f I_0}{M_0}; \quad \nu = k_{tc} / k_{td} = 1 \tag{5}$$

$$E_1 = E_p + E_d/2 - E_t/2$$

$$A_1 = (2f)^{1/2} A_p A_d^{1/2} A_t^{-1/2} \tag{6}$$

The number average chain length can be given as:

$$L_n = \frac{X}{q_0} \tag{7}$$

The control variables in the isothermal batch jacketed reactor are taken as reaction temperature and initial initiator concentrations. In order to acquire optimal operating conditions, the method of Lagrange's Multiplier and Hamiltonian Maximum Principle are used and the optimum conditions are given in the Table 1.

3. DESIGN OF NONLINEAR GPC

Consider the control of a linear state space process with the output corrupted by noise. The system has the ARMAX representation:

$$\sum_{i=0}^{n} a_i y(t-i) = \sum_{i=0}^{m} b_i u(t-\tau-i) + \sum_{i=0}^{n} c_i v(t-i) \tag{8}$$

where $\tau \geq 1$ is a delay.

All zeros of $B(q^{-1}) = (b_0 + b_1 q^{-1} + ... + b_m q^{-m})$ and $C(q^{-1}) = (1 + c_1 q^{-1} + ... + c_n q^{-n})$ are strictly inside the unit circle, and $A(q^{-1}) = (1 + a_1 q^{-1} + ... + a_n q^{-n})$ is monic. The objective is to cancel the noise by manipulating the control $u(t)$. Only $y(t)$ is measured. Such problems arise in signal processing and numerous control applications. The control objective is achieved by the feed-forward law:

$$u(t) = \frac{1}{\beta_0} \left[\sum_{i=1}^{l} \alpha_i y(t+1-i) - \sum_{i=1}^{m} \beta_i u(t-i) - \sum_{i=0}^{d-1} f_i v(t+1+i) \right] \tag{9}$$

Where the coefficients f_i, α_i and β_i are found by solving a Diophantine identity:

$$C(q^{-1}) = F(q^{-1}) A(q^{-1}) + q^{-d} G(q^{-1})$$

For F and G and setting $\alpha(q^{-1}) = G(q^{-1})$ and $\beta(q^{-1}) = F(q^{-1}) B(q^{-1})$. The control u(t) (Eq. (9)) cannot be applied since noise signals are required "d " steps into the future. Stochastic control overcomes the problem by assuming that $v(t)$ is white noise, which

may be generated from the sampling of a Brownian motion. We may then exploit the fact that $E\{v(t+1)|F(t)\}=0$ for $i \geq 1$. The optimal control, in the sense of minimizing the output variance, is then implemented using measured signals Ydstie (1990). The control law that sets the predicted output to zero satisfies:

$$u(t)=\frac{1}{\beta_0}\left[\sum_{i=1}^{l}\alpha_i y(t+1-i)-\sum_{i=1}^{m}\beta_i u(t-i)-\sum_{i=0}^{d-n+1}g_i v_f(t+1+i)\right] \quad (10)$$

Where $\alpha(q^{-1})=G(q^{-1})$ and $\beta(q^{-1})=F(q^{-1})B(q^{-1})$,

$$H(q^{-1})=(1+h_1q^{-1}+...+h_mq^{-m})=F(q^{-1})C(q^{-1})$$

To establish the GPC algorithm, it is supposed that a model of the linearized plant is expressed in terms of the NARMAX . The most general NARMAX model structure would take the form below, where some function of the previous output $y(k-T)$, $y(k-2T)$ and inputs $u(k-T), u(k-2T)$, would give the current output.

$$y(k)=F[(y(k-T), y(k-2T), y(k-n_yT), u(k-T), u(k-2T),..., u(k-n_uT))] \quad (11)$$

This is a natural model for any sampled dynamic system with inputs at discrete times. It is extremely difficult to determine the function F which fits the input/output data well over a range of operating condition.

Consider the vector error (ε) composed of predicted future system errors $W(t+j)-\hat{y}(t+j)$. W is the references signal. The suggested future control sequence $\{u(t+j)\}$ is chosen by GPC at time t to minimize a cost-function such as

$$J(N_1,N_2,NU,\lambda)=\sum_{J=N_1}^{N_2}\varepsilon^2(t+j)+\lambda\sum_{j=1}^{NU}\Delta U^2(t+j-1) \quad (12)$$

N_1 is the minimum costing horizon,
N_2 is the maximum costing horizon,
N_U is the control horizon, and
λ is the (optional) control weighting
According to GPC strategy, equation (8) can be written as

$$\hat{y}=\overline{G}\Delta\overline{u}+f \quad (13)$$

$$\overline{G}=\begin{bmatrix} g_0 & 0 & \cdots & 0 \\ g_1 & g_0 & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ g_{N_U-1} & g_{N_U-2} & \cdots & g_0 \\ \vdots & \vdots & & \vdots \\ g_{N_2-1} & g_{N_2-2} & \cdots & g_{N_2-N_U} \end{bmatrix}_{N_2 x N_U}$$

$\overline{G}$ matrix is $N_2 x N_U$ dimensional lower triangular matrix.

4. EXPERIMENTAL SYSTEM

Experimental system is shown in Fig. 1. Water was used as coolant in the jacket and the temperature inside the reactor was measured at each sampling period using thermocouples. A computer with A/D and D/A converters was employed for data acquisition and the control of experimental reactor. In this study, the monomer of styrene was used and benzoyl peroxide was used as initiator. Toluene was chosen as a solvent. The reactor contents were 70% styrene and 30% toluene. The experimental studies were carried out in a cylindrical glass jacketed reactor which has the internal volume of 1600 ml and jacket volume of 980 ml. The temperature of the reactor and the temperatures of the jacket inlet and outlet were measured by thermocouples. The converter modules were connected to adjust the flow rate of the pump and the heat input which is given into the reactor. In this work, the converter module has two outputs (pump and heater) and three inputs (reactor temperature, cooling water inlet temperature and outlet temperature). All A/D connections to the reactor were realized on the converter module using VisiDAQ packet program. Reactor was heated by an immersed heater inside the reactor. In all the experiments, the manipulated variable was accepted as the heat input which was transferred by diver heater to the reactor. It was accepted that enough mixing was provided by the mixer in the reactor.

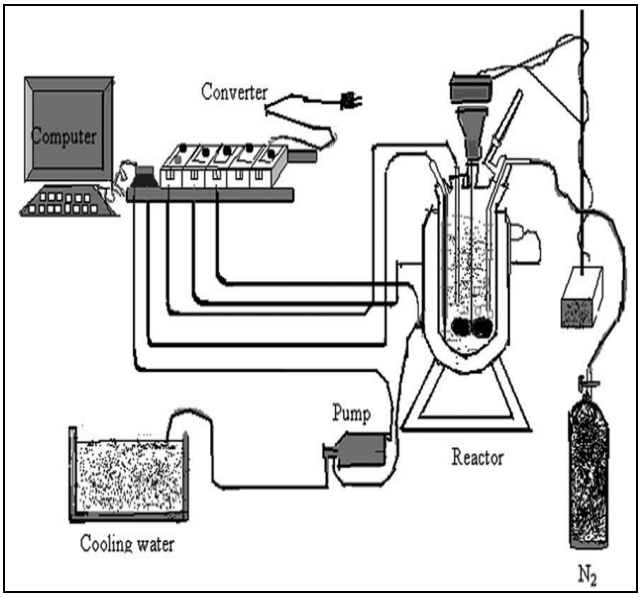


Figure 1: Experimental System

5. RESULTS AND DISCUSSION

The proposed control strategy was deeply tested on a batch polymerization reactor. The optimum reactor temperature set point for minimum polymerization time was obtained at different benylzoyl peroxide initial initiator concentrations. Two of these optimum reactor set points were selected for control purposes. GPC was applied to control the optimal temperature of a styrene polymerization reactor.

FORTRAN program has been used in the simulation of the system. For NLGPC control, models of the system have been taken in the following from (Yuce 2001).

Model1:

$$\left(y(t)-y(t-1)\right)=\frac{b_0 z^{-1}}{1+a_1 z^{-1}+a_2 z^{-2}}\left(U(t)-U(t-1)\right) \quad (14)$$

a_1= - 0.5227 a_2= 0.0001 b_1= - 0.4773

Model2:

$$\left(y(t)-y(t-1)\right)=\frac{b_0 z^{-1}}{1+a_1 z^{-1}}\left(U(t)-U(t-1)\right) \quad (15)$$

a_1= - 0.9998 b_1= 0.0001

Where $U(t)=u^4(t-1)$

These coefficients (b_0, a_1 and a_2) were found by using a pseudo random binary sequence as the input function. A second order polynomial is sufficient to represent the denominator plant dynamics. The system is defined and the model parameters are calculated using the least squares regression method given as follows. Regression coefficient (R^2) is found as 0.96.

Some experimental studies have been carried out to acquire the quality of polymer product. The effect of different optimal conditions has been examined on monomer conversion, average viscosity molecular weight and chain length. The polymerization reactor temperature was controlled by manipulating the powder to the heater and monitored to see the control performance.

To see the performance of NLGPC control and to compare with LGPC and different models, some experiments have been carried out on the experimental system. To obtain the reaction curve, a step change in the heat input was applied after the system reached to the steady state conditions. The temperature responses to this effect obtained from experimental work are given in Fig 2.

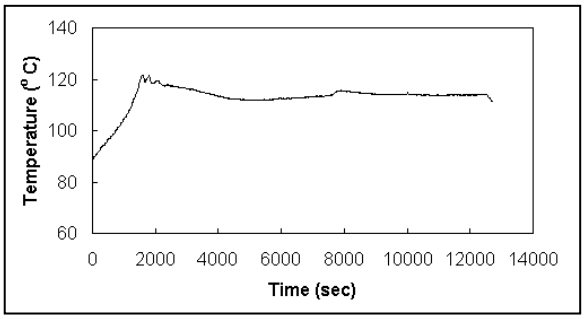


Figure 2: Temperature response of the reactor inlet (I=0.0185 mol l^{-1}, T=89.04°C)

As described in the previous section, the optimum reactor temperature for a minimum polymerization time was obtained at different benylzoil peroxide initial initiator concentrations. For control purposes, two of these optimum reactor constant temperature and paths were selected and shown in Table 1. Experimental control results are shown in Figs. 2-7 for LGPC and NLGPC.

The experimental results show some oscillations due to the fact that this is a batch exothermic reaction with constantly changing conditions and heat generation is not constant during the reaction (Zeybek, Cetinkaya, Hapoglu, and Alpbaz 2006).

As it is seen from Fig 3, 6 and 7, for variable temperature control the fluctuations occurrence are more significant and the frequency is higher than constant temperature control.

At the same operating conditions of LGPC and NLGPC temperature control was used for comparison. An overshoot is observed at the beginning of the reaction for this two control method and then the NLGPC and LGPC controller bring the reactor temperature back to set point but after 6000 seconds LGPC controller continuous as increasing oscillation until the end of the experiment (Fig. 3a).

Figs. 4-5 show the results of experimental temperature response of NLGPC for different models at constant set point. Both of model 1 and model 2 for

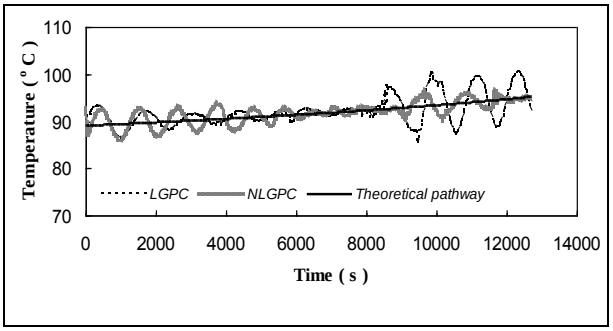
NLGPC is very suitable, fast and robust in these applications. Also, desired values chain length and conversion were closely achieved (Table 2).

In Figs. 6-7, the experiments used for two models of NLGPC were carried out using two different temperature pathways which have 89.04 and 92.7 ˚C of initial temperature respectively shown in Table 1. The chain length of 500, conversion 50% were target in these pathways. As seen in Figs.6-7, at the temperatures best result was reached for model 2 and both chain length and conversion, target was nearly reached (Table 2). The experimental results demonstrated that NLGPC at the constant temperature has been good control performance, because some oscillations in the temperature around the set point are seen due to continuously changing conditions, gel effect and strong nonlinearities (Yuce, Hasaltun, Erdogan, and Alpbaz 1999).

The progression of the manipulated variable (Q) executed by the computer during the experiments is also monitored in Figures 4(b)-7(b).

Table1: Optimal operating conditions used in experimental studies

Run	T_R (^oC)	I_0 (mole l^{-1})	M_0 (mole l^{-1})	t_f (s)	X_d (%)	L_{nd} ($gmol^{-1}$)	T_c ^oC
1	103.8	0.0126	6.092	9036	50	500	21
2	105.5	0.0038	6.092	7440	30	1000	21
3	89.04	0.0185	6.092	12720	50	500	21
4	92.7	0.0150	6.092	10200	50	500	21



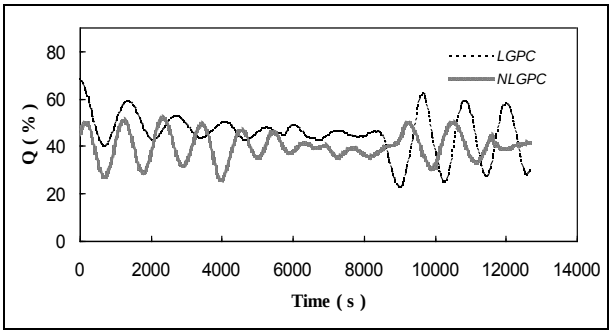
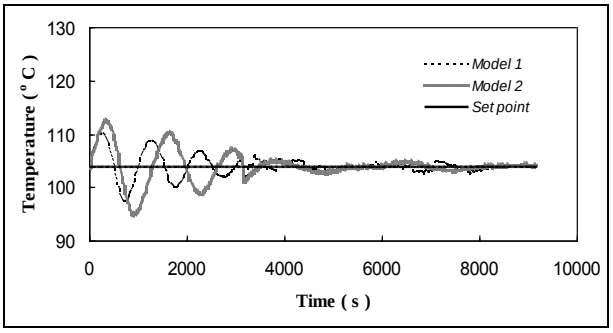


Fig. 3: (a) Temperature response, (b) manipulated variable with time under LGPC and NLGPC respectively (I_0 = 0.0185 mole l^{-1}, Model 1)



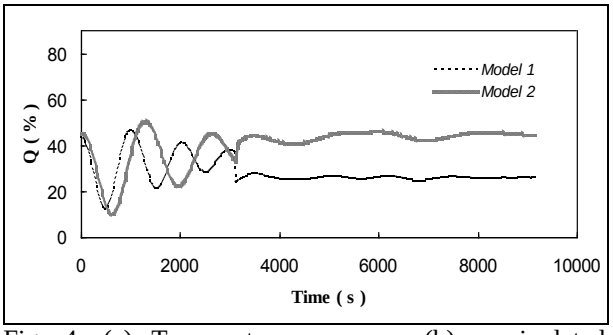
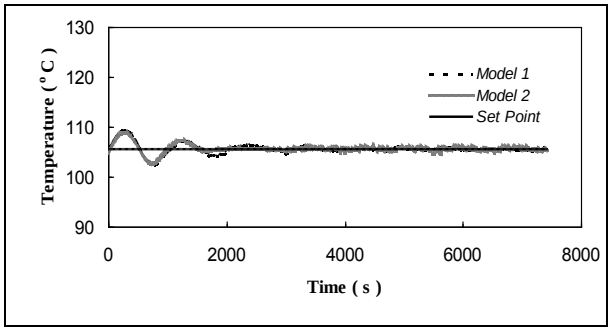


Fig. 4: (a) Temperature response, (b) manipulated variable with time under NLGPC respectively for model 1 and model 2 (I_0 = 0.0126mole l^{-1})



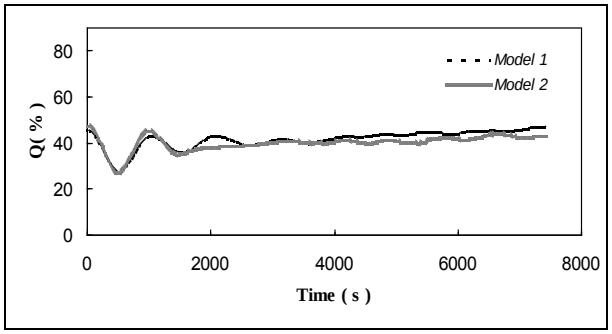
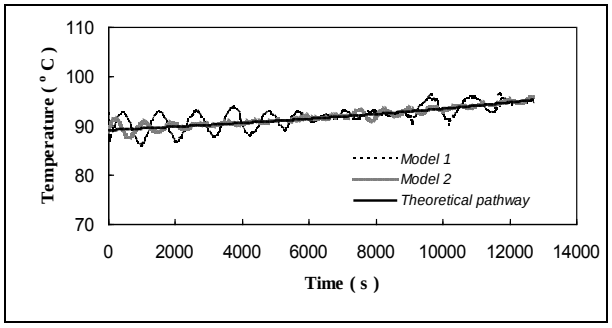


Fig. 5: (a) Temperature response, (b) manipulated variable with time under NLGPC respectively for model 1 and model 2 (I_0 = 0.0038mole l^{-1})



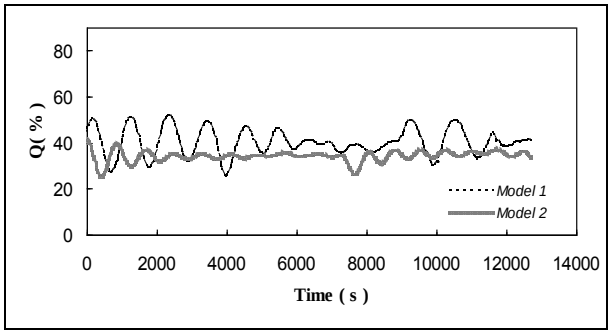
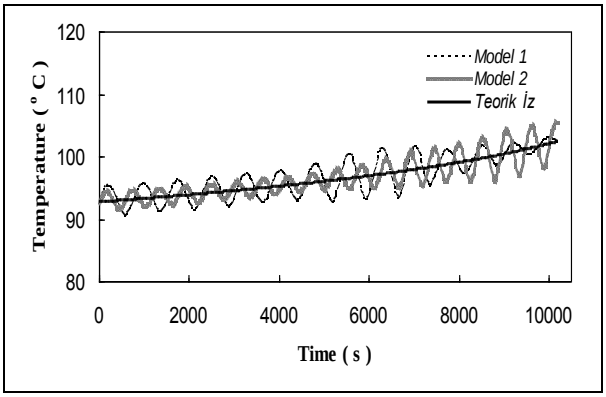


Fig. 6: (a) Temperature response, (b) manipulated variable with time under NLGPC respectively for model 1 and model 2 (I_0 = 0.0185mole l^{-1})



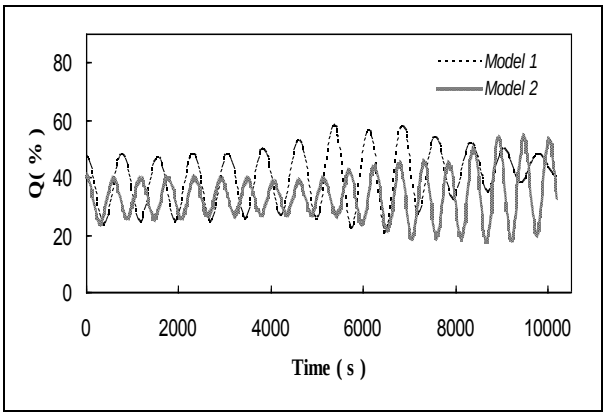


Fig.7: (a) Temperature response, (b) manipulated variable with time under NLGPC respectively for model 1 and model 2 (I_0 = 0.0150 mole l^{-1})

Table 2: Comparison of experimental results with target values

T (^{o} C)	**103.8**	**105.5**	**89.04**	**92.7**
I_0 (mole l^{-1})	**0.0126**	**0.0038**	**0.0185**	**0.0150**
Desired conversion, m^*(%)	50	30	50	50
Experimental conversion, m(%) (NLGPC,Model 1)	44	33	71.5	65
Experimental conversion, m(%) (LGPC, Model 1)	-	-	57.8	-
Experimental conversion, m(%) (NLGPC,Model 2)	45.9	29.88	69.95	54.84
Desired chain length, X_n^*	**500**	**1000**	**500**	**500**
Experimental chain length, X_n (NLGPC,Model 1)	425	940	321	269
Experimental chain length, X_n (LGPC, Model 1)	-	-	204	-
Experimental chain length, X_n (NLGPC,Model 2)	412	955	352	452
Desired average viscosity molecular weight	**52000**	**104000**	**52000**	**52000**
Experimental average viscosity molecular weight (NLGPC,Model 1)	44253.2	97869	32983.82	27681.35
Experimental average viscosity molecular weight (LGPC, Model 1)	-	-	21249	-
Experimental average viscosity molecular weight (NLGPC,Model 2)	42851.5	99445.3	36324.5	47155.6

6. CONCLUSION

Generalized Predictive Control has been presented in this paper. Linear Generalized Predictive Control (LGPC) and Nonlinear Generalized Predictive Control (NLGPC) algorithm were used to keep the temperature of a jacketed batch polymerization reactor at constant and variable optimal conditions. Good performance is achieved using NLGPC. The experimental results obtained have also confirmed that this control performs well particularly at constant optimal conditions. It is also observed that by the use of NLGPC, the desired values of molecular weight are achieved at the end of the batch. In addition, the performance of NLGPC is better than the LGPC.

APPENDIX A. NOMENCLATURE

A_d, A_p, A_t :Frequency factor for initiator decomposition, propagation and termination respectively, s^{-1}, L mol s^{-1}

a_I : Parameters of A polynomial

b_o : Parameters of A polynomial

c : initiator conversion

E : a polynomial

E_d, E_p, E_t :Activation energies for initiator decomposition, propagation and termination, respectively, kJ $kmole^{-1}K^{-1}$

f : Initiator efficiency

g : gel effect

G : a polynomial

$\overline{G}$: a matrix

I, I_0 :Initiator concentration, initial initiator concentration, respectively, mole L^{-1}

k_d : Initiator decomposition rate constant

k_p : Propagation rate constant

k_t : Termination rate constant

L_n, L_{nd} : Number average chain length, desired number average chain length

M, M_0 : Monomer concentration, initial monomer concentration, mole L^{-1}

N_1 : The minimum costing horizon

N_2 : The maximum costing horizon.

N_U : The control horizon

q_0 : Dimensionless zeroth moment

T_c, T_{ci}, T_{co} :Average, inlet and outlet coolant temperatures (^{0}C)

t, t_f : Time, polymerization time, s

u(t) : Input variable at time t

v : k_{tc}/k_t, constant

X : Monomer conversion,

y(t) : Output variable at time t

$y(t-1)$: Output variable at time t-1.

Greek symbols

α : Coefficient of momentum

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