Non-linear Model predictive control of particle size distribution in batch emulsion polymerization

Parul Arora*, Alok Gupta[1]

*-parul.arora07@gmail.com

Chemical Engineering Department

Malaviya National Institute of Technology,

1-Malaviya National Institute of Technology,

Abstract

A non-linear model predictive control has been developed for batch emulsion polymerization. Process variables can be easily measured in most reaction system but only few variables can be measured in batch system during process e.g. temperature, pressure and flow rate of jacket fluid are available. In the absence of steady state in emulsion polymerization that operates in batches is factor that shows the need of a robust control system. Temperature of polymerization has an important role on end-use property of polymer. The emulsion polymerization of Methyl methacrylate is chosen for this study, and an appropriate mathematical model is developed, which is detailed enough to explain the particle size distribution of the product. Heater power is used as manipulative variable and the result of MPC is compared with traditional controller.

Keywords: Model predictive controller, PID, Weight average molecular weight, emulsion polymerization.

Introduction

Polymerization processes are highly exothermic in nature, rapid, non-linear and sensitive towards impurities hence these processes are concerned with advance process control. Nowadays many industries are considering the transformation from big to smaller reactors for process intensification. Batch and semi-batch reactors are being replaced by continuous pilot-sized reactors, which are easy to control, this evidence that control of batch reactors is more

challenging. It is required to develop an optimum and robust control strategies to control the end-use properties in a batch reactor.

Molecular weight distribution and particle size distribution strongly depends upon temperature of the reactor, concentration of monomer in polymer particle and total number of particles but in batch reactor only temperature is available as control variable to control molecular weight of polymer. By increasing the reactor temperature the free surfactant concentration reduces more quickly, therefore its concentration reaches below the critical micelle concentration (CMC) sooner. As micellar nucleation occurs as long as free surfactant concentration is above the CMC, increasing the reactor temperature reduces the nucleation period. On the other hand, the increase of reactor temperature results in a higher rate of production of oligomeric radicals, which leads to formation of more particles in a shorter time. Consequently, increase of the reactor temperature leads to production of higher number of particles in a shorter time and therefore PSD becomes narrower with a smaller average particle size; it is required to maintain the temperature through out the process for uniform narrow distribution. We already study the conventional control and found overshoot, oscillations in disturbance rejection. A Conventional controller is able to control but our aim is to provide robust and tight control to the batch reactor system. Traditional controllers like PID are still hard to be applied to the industries processes because they still have critical problems such as lack of robustness and stability.

Authors(Ozkan, Hapoglu et al. 1998) used generalized predictive control system for ARIMAX model using Levenberg, Marquardt, kalman and bierman algorithms. A pseudo random binary sequence signal was given to the manipulated variable. Experimentally they provide heat (manipulated variable with the help of virac/triac module and maintained floe rate of cooling water at fixed value. PID controller was utilized to compare the efficiency of GPC method. They concluded, both controllers are satisfactory but GPC performed well in temperature trajectory than the PID controller.

Author(Othman, Othman et al. 2011) proposed integral strategy to control molecular weight in emulsion polymerization for polystyrene system. They implemented input/output linearization coupled with proportional controller. They found molecular weight of polymer more affected by temperature of the reactor than the concentration of monomer in polymer particle.

Authors(Hosen, Hussain et al. 2011) investigated free radical polymerization using a artificial neural network mechanistic modeling strategy. The homolysis of initiator is rate determining step of initiation. Neural network can be utilized to model the poorly known or unknown parameters such as kinetic parameters. They use least square method to optimize the model performance. In neural network they used Delta-Rule algorithm (back propagation) to train the network. The performance of NNPC is compared with PID. They notice PID took longer time(885s) to adjust closer to set-point with an overshoot but NNPC take only 145s.

Yuce and his co-workers (Yuce, Hasaltan et al. 1999) worked on control of solution batch polymerization using dynamic matrix control (DMC) and compared with internal model control (IMC). They found both controller yielded good performance in maintaining the temperature of the reactor in isothermal condition, similarly (Garcia, Prett et al. 1989; Peterson, Hernandez et al. 1992) few studies has been done with various chemical reactors.

Ohmura et al (Ohmura, Horie et al) proposed compartment reactor consisting of three well mixing compartments. This reactor has advantage that each compartment of the reactor can keep a different non-equilibrium steady-state with respect to each other. An emulsion polymerization was categorized into four function module and these modules were assigned to different compartment. First two modules Contacting and Activation were assigned to first compartment and third module chemical reaction and last compartment with particle growth function module. The reactor used by authors was set vertically, and the lower middle and upper compartment named first, second and third respectively. They found more turbine impeller used in compartment, and higher monomer conversion is obtained. They stated that strong the agitation enhances the reactivity to obtain higher monomer conversion further more the compartment reactor successfully produced stable and monodisperse particles.

Designing of Controller

Temperature control of a batch reactor depends mainly on heating and cooling system of the reactor either by flow rates of cooling water or by heater power. In this study we use heater power as manipulative variable. The model developed and solved for simulation by Runge-Kutta integration method. Model predictive control is a generic term for a widely used class of controllers(Cueli and Bordons 2008; Harnischmacher and Marquardt 2007).

Model predictive control is one of the most widely used advance control method. MPC presented a set of future manipulated variable moves is calculated to minimize the objective

function over a prediction horizon based on the sum of squares of the differences between model predictive output and desire output(Nagy and Agachi 1996).

The Tool used in MATLAB to compute the manipulated variable moves subject to constraints is cmpc, which solve optimization problem by using QP. The function requires model used for estimation the plant and model which state the controller in step format, limits of both input and output variable and weights of output and manipulated variables.

$[y_p, u]$ = cmpc (plant, model, ywt, uwt, M, P, tend, r, ulim, ylim)

There is some assumptions taken for MPC

1. The output sequence y(k) for ylim is also observed
2. The input u(k) is a continuing driving function of the process
3. The noise is a random sequence with zero mean and is uncorrelated with u(k).

In MPC future value of output variable is predicted using a dynamic model of the process and current measurements. The control calculations are based on both future predictions and current measurements. Usually MPC used when PID is unable to control the system or when there are some constraints or limitations on process variable and manipulative variable. In this study we compare the performance of PID and MPC for controlling molecular weight of a batch emulsion polymerization.

The flow chart of MPC used in matlab programming if shown in fig 1.

1. In MPC the process model calculates the predicted future outputs for the prediction horizon (N) at each sampling time t. Theses depends upon past inputs and outputs, including the current output y(t) (initial condition).
2. The sequence of future control signals is computed to optimize a performance criterion. Usually the control effort is included in the performance criterion.
3. Only the current output signal u(t) is send to the process. Step 1 is repeated for all the sequence to bring the process up to date.
4. The future moves of the manipulated variables are determined by minimizing the predicted error or the objective function

Mathematical Model

This model is zero-one model referring instantaneous bimolecular radical termination followed by some assumptions-

1. All reactions are irreversible.
2. The reactions are independent of chain length.
3. The quasi-steady state assumption is stated for free radical chain.
4. There is no chain transfer.

The energy balance for the reactor and the jacket used is tabulated in table no.1

The particles are assumed to contain either one or zero radical; this system is called zero one model. Population balance equation for particle size distribution is given by equation.

$$\frac{\partial \eta_0(r)}{\partial t} = \rho(r)\left[n_1^p(r) + n_1^m(r) - n_0(r)\right] + k_0(r) \times n_1^m(r) \quad (1)$$

$$\frac{\partial n_1^p(r)}{\partial t} = k_p \times n_1^p(r) + \rho_{ini}(r) \times n_0(r) - \rho(r) \times n_1^p(r) - k_{tr}[M]_0 n_1^p(r) + \sum_{j=z}^{jcri} k_{em,i}\,[micelles][p_i^*]\,\delta(r - r_{nuc}) \quad (2)$$

$n_0(r)$ is the population containing zero radicals and $n_1(r)$ is the population containing one radical , polymer radical $n_1^p(r)$ which would not readily diffuse out of the particle due to its size. Monomer radicals formed from chain transfer reaction is $n_1^m(r)$, which presuming can readily exit particle. j_{crit} is the chain length which become insoluble in water and participate in anew particle.

Particle growth rate is given by

$$G(r) = \frac{k_p[M]w_m}{4\pi r^2 \rho_p N_A}$$

Here k_p is propagation rate, w_m molecular weight of monomer, density of polymer represent by ρ_p.

Quasi –steady state assumption is applied and we get

$$n_1^m = \frac{k_{e,t}\,(r)[E]\,n_0\,(r) + k_{tr}\,[M]_p\,n_1^p}{\rho_r + k_{p,e}\,[M]_p + k_0\,(r)}$$

$n(r) = n_0(r) + n_1^p(r) + n_1^m(r)$

$n_1(r) = n_1^p(r) + n_1^m(r)$

Zero –one system is where entry of a radical into a particle which already contains a growing radical causes termination at a rate much faster than overall polymerization. In such a system the average number of radicals per particle cannot exceed 0.5, termination occurs only between an entering radical and a radical whom has been growing at the same time. Heater power is used as manipulative variable to maintain the control variable to its target value. Same model is used in Simulation using PID controller and the results are discussed later in this article.

The energy balances for the reactor and jacket are formulated in table no 1. Where Q is the heater power which is used as manipulative parameter, U and A are the overall heat transfer coefficient and the heat exchange area respectively. As the monomer conversion increases, the heat transfer efficiency becomes poorer and the heat generation rate increases which increases the temperature of the reactor. Hence a robust control is required.

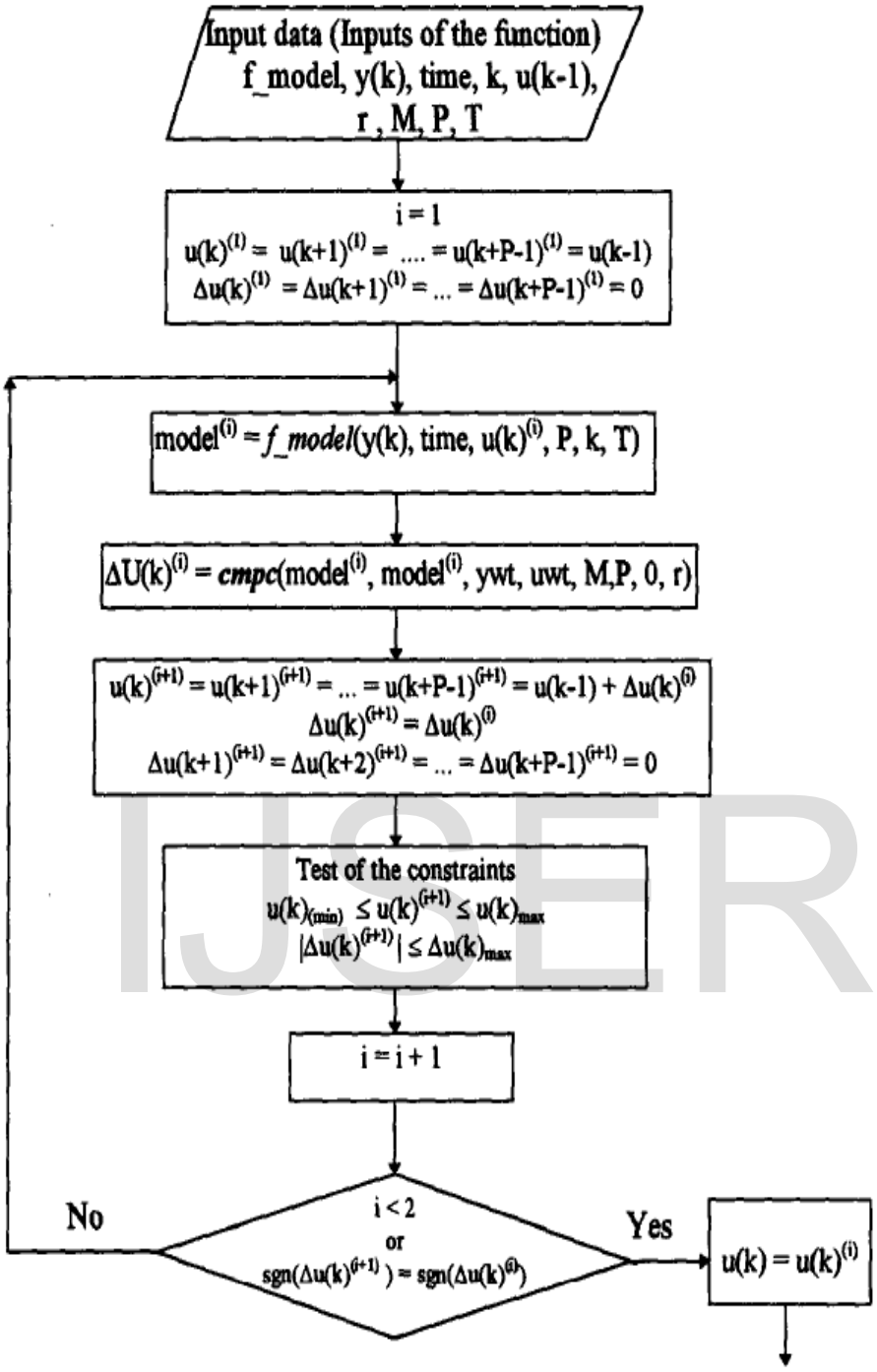

Fig 1: Flowchart for MPC

Simulation method

The MPC GUI is designed to analyze and simulate the MPC controller and plant model combinations. A simulink block diagram is designed as shown in figure 2. This simulink file contains S-Function file which is containing the model equations discussed above along with material balances for monomer, initiator and also energy balances. The initial value of

process parameters one should check before the implementation of non-linear model. The initial value of process variables of our model is tabulated in table 2.

Table 1: Energy balance for Reactor and jacket used in model

Equation for Reactor	$Q+(-\Delta H)R_m\,V-UA(T-T_J)/VC_p\rho-(T/VdV/dt)$
Jacket dynamics	$M_cC_{pc}(T_{ji}-T_{jo})+UA(T-T_j)/V_c\rho_cC_{pc}$

Table 2: Initial value of process variables.

Manipulative Variable	State variables
Q=90W	I=1.42 mol/l M=3.58 mol/l T_{jo} = 300K

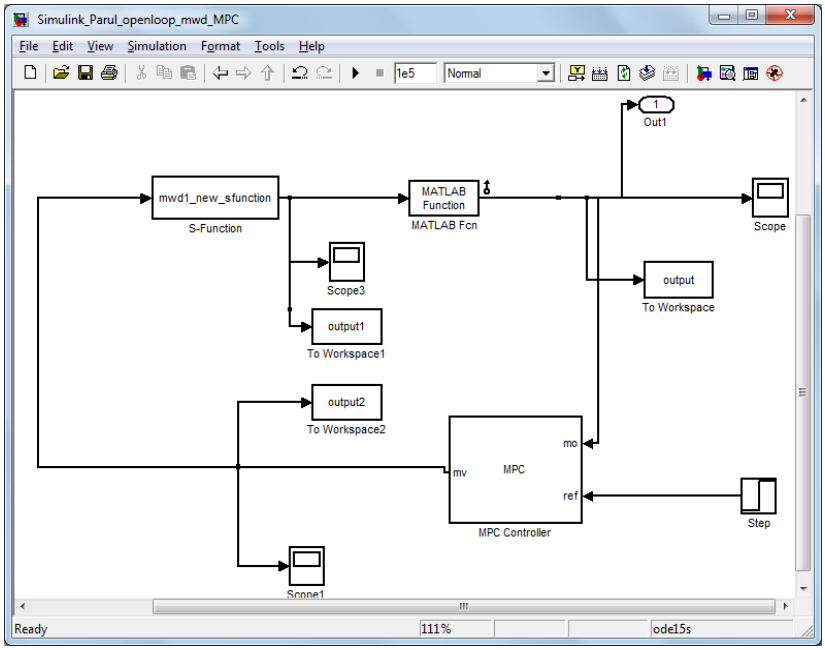

Figure 2: Simulink block diagram for MPC

Model predictive control usually follows the ideas:

1. Explicit use of model to predict the process output along future time horizon.
2. Calculation of control output to optimize the performance index.
3. A receding horizon strategy, which involves the first control signal of the sequence calculated at each step.

Results

The open-loop control strategy is very sensitive to disturbances and model mismatch. To overcome this problem, a closed-loop control strategy is proposed. Particle nucleation depends on the free surfactant concentration level above the CMC, and therefore the free surfactant concentration profile is closely related to the physical phenomena affecting the PSD. Consequently, the ionic free surfactant concentration can be used to control PSD inferentially. But it should be noted that use of flow rate of surfactant or any other process variable as manipulative variable is only possible in either continuous or semi-batch reactor. In batch reactor use of temperature to control the end-use property is the possible easiest way and the manipulative variables used for the same is either flow rate of jacket fluid or heater power, in this work we use heater power as manipulative variable to control the Particle size distribution through temperature in a batch reactor. The value of manipulative variable if defined as 90 W by may vary 40-140 W to maintain the optimum set-point. The value of state variables is given table 2.

The model predictive controller is used for control of the PSD of polymer PMMA. Heat duty of the heater is used as manipulative variable and temperature as controller variable in this study. The coolant flow rate is kept constant. The design parameters used in the simulation given in table 3, 4. The performance of MPC is compared with PID controller. Our objective is to improve the control performance. Tunning of the controllers were done by trail and error method and tabulated in table 3 for MPC and table 4 for PID.

Table 3 : Tuning parameters for MPC

Parameters	Value
Weight Tunning	0.7
Prediction Horizon (Np)	100
Control Horizon (Nc)	10

Table 4: Tuning parameters for PID

Parameters	Value
Controller Gain	10
TauI	0.01

It is worthy to note that the process model describing the PSD is non-linear, distributed and includes an important number of parameters that are not well known or sensitive to adulterations. This makes the model-based control of PSD more difficult task. Moreover, online measurement of PSD without delay is not available.

Our objective is to control PSD, instead of direct control we control through controlling the temperature of the reactor, Indeed temperature affect the propagation, solubility and entry parameters which affects the nucleation rate, hence temperature ultimately control the PSD.

The stable operational state is never achieved in emulsion polymerization reactors that operate in batch system. Hence tuning of the reactor is done only by trail and error method for this study.

Figure 3 shows the controller profile for MPC and PID. The response shows that PID controller took large time to get closer to the set-point with overshoot. MPC took 2100s where as PID adjusted within 2800s to the set-point with 4 K overshoot which depicts higher overshoot than MPC. In MPC heat duty oscillates in start-up but In PID heater power oscillated at time 50000s with initial start-up also where as MPC gives rest response smoother than PID which will prolong the life of heater. Absolute Error calculated for the reactors are given in table 3 and 4 on the basis of performance index tunning parameters was

finalized. The main control objective of our study is to bring the reactor temperature to its target value as rapidly as possible with minimal temperature overshoot. It has been observed in the figure 3 that temperature overshoot can be minimized by this proposed method.

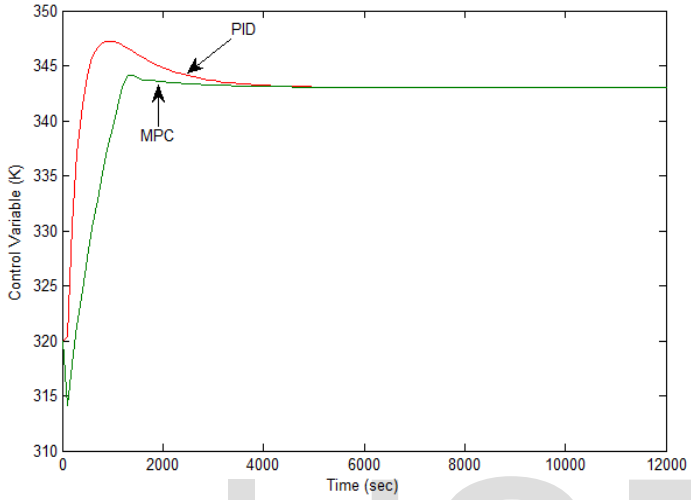

Fig 3: Comparison of control variable using MPC and PID

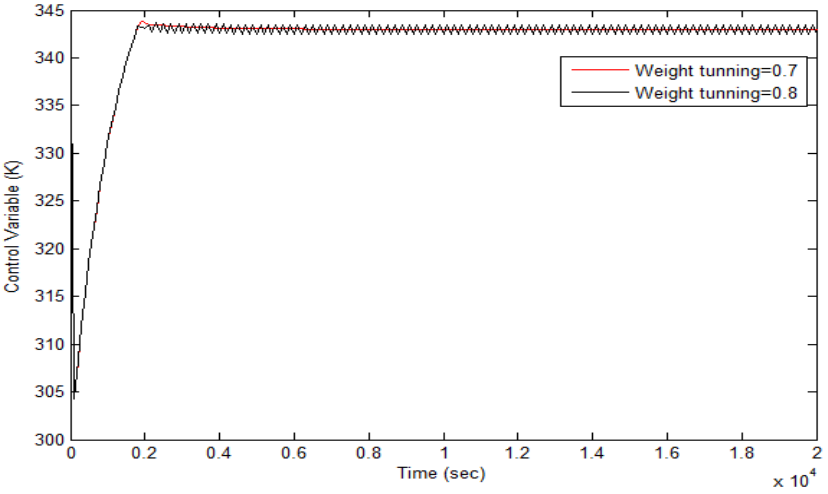

Fig 4 : Effect of weight tuning

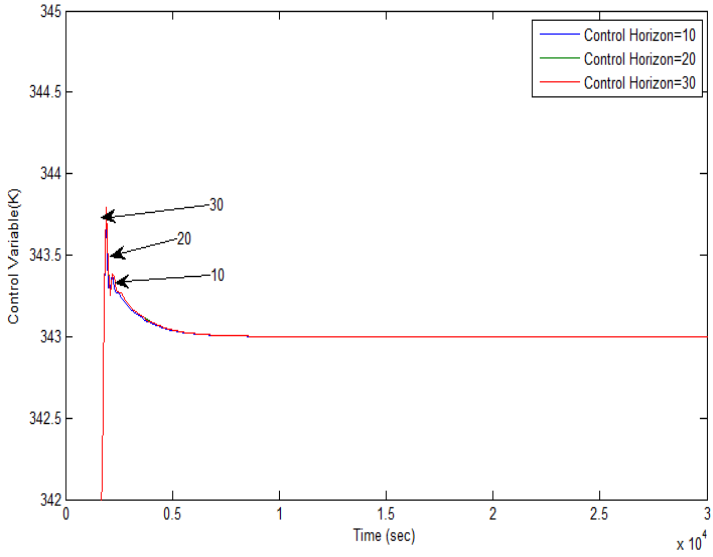

Fig 5: Effect of control horizon

As the temperature increases, initial conversion increases significantly. This is expected because the rates of reactions are exponentially temperature dependent functions according to Arrhenius law. At higher temperature, concentration of oligomeric radicals in the aqueous phase and nucleation rate increase and consequently the total number of particles increases. This in turn leads to a smaller final average particle size but run away of the temperature destroys the uniformity of the particles hence a robust control is required to maintain the particle size of the latexes.

Performance index were compared of both controllers in table 5 and 6. The control system for a polymerization reactor must be sufficiently robust to handle unmeasured disturbances, which impact polymer reactor operation. These disturbances typically result either from trace amount of polymerization inhibitors or retarders left over after monomer purification prior to the polymerization reaction or oxygen which may be present in a typical polymerization recipe and which may be affecting the reaction. Process control of batch reactors must address the main disadvantage of batch reactors versus continuous system, namely variability within a batch. This variability is particularly important in batch free radical polymerization, where the time of formation of a single chain is only a very small fraction of the batch time and therefore in-homogeneity results from the fact that polymer chains can be formed under very different conditions during the course of the batch.

Table 5 :MPC Performance Index for PSD

Parameter	ITAE	IAE	ISE	ITSE
Weight Tunning-0.7	1.7150e+012	3.4315e+007	1.1775e+010	5.8825e+014
Weight tunning-0.8	1.8158e+012	2.8626e+007	6.7263e+010	2.6668e+014
Nc-10	1.7190e+007	3.4355e+007	1.1776e+010	5.8826e+014
Nc-20	1.7150e+012	3.4315e+007	1.1775e+010	5.8825e+014
Nc-50	1.7550e+012	3.4395e+007	1.1795e+010	5.8828e+014
Np-100	1.7150e+012	3.4315e+007	1.1775e+010	5.8825e+014
Np-50	2.1150e+012	2.1315e+007	1.7715e+010	6.6825e+014

Table 6 : Performance Index PID for PSD

Parameter	ITAE	IAE	ISE	ITSE
Kc-10	1.1458e+007	1.1949e+004	1.1505e+005	3.1816e+007
Kc-9	1.1657e+007	1.2715e+004	1.2730e+005	3.8286e+007
Kc-8	1.1871e+007	1.3540e+004	1.4205e+005	4.6841e+007
Kc-6	1.4137e+007	1.6120e+004	1.8296e+005	7.4853e+007
Kc-3	3.1867e+007	2.5277e+004	3.0668e+005	1.9843e+008
TauI-0.1	5.0679e+007	3.3373e+004	4.2574e+005	3.6435e+008
TauI-0.01	1.4137e+007	1.6120e+004	1.8296e+005	7.4853e+007
TauI-0.001	1.0293e+008	2.0597e+004	1.7498e+005	6.7183e+007
TauI-0.0001	2.4418e+008	1.6576e+004	1.7379e+005	4.4335e+007

Tuning of parameters in batch reactor is always difficult as compared to continuos reactor but it is strongly required to improve the performance of the controller. Trial and error method was using for tunning of both the controllers. In MPC as the value of weight tunning decreases number of oscillations increases, Change in control horizon and prediction horizon makes little difference (fig 4,5,6) but the best value was taken after the study of performance index, the same procedure is repeated for the PID controller, decrease in the value of controller gain causes higher the value of overshoot on control variable as shown in fig, at Kc=1 highest overshoot was obtained, as the value of tauI increases oscillations in the control variable increases as in fig 7 and 8.

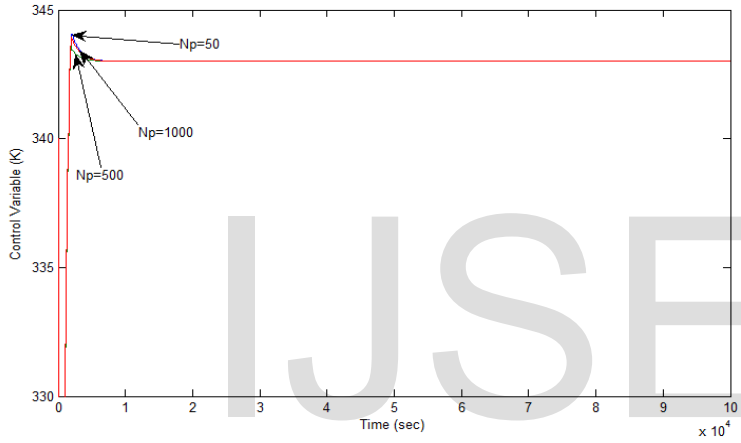

Fig 6 : Effect of prediction horizon

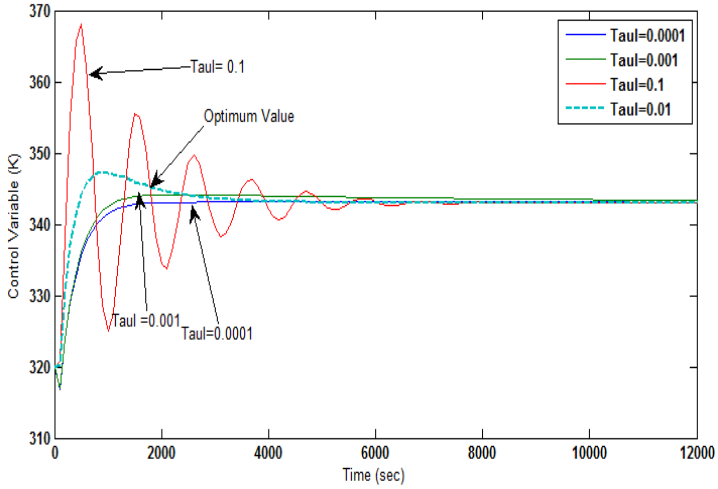

Fig 7: Effect of taui

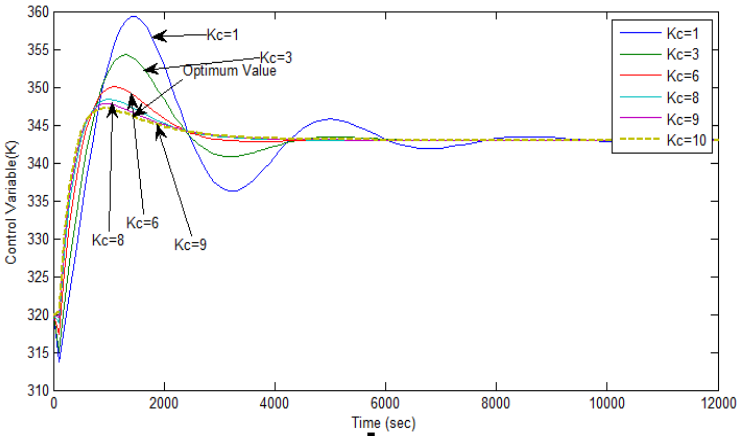

Fig 8: Analysis of controller gain

In industrial practice, some uncertainty and disturbance are normally encountered in the reactor; to explore this we introduce disturbance and set-point change in the reactor. An excitation in the cold flow rate was introduced at the time 50000s. Increasing the flow rate would decrease the temperature of the reactor and decrease in flow rate would increase the temperature because of the heat transfer rate of the reactor shown in fig 9 for MPC and PID simultaneously. Similarly set-point was also introduced into the system at time 50000s shown in fig 10 for MPC and PID. MPC took less time as compared to PID for getting closer to the set-point in load-change, and gave less oscillation in set-point change with small overshoot. This evidence that advance controllers are promising than traditional controllers in terms of there robustness for non-linear control. Comparison of particle density function is given in fig11.

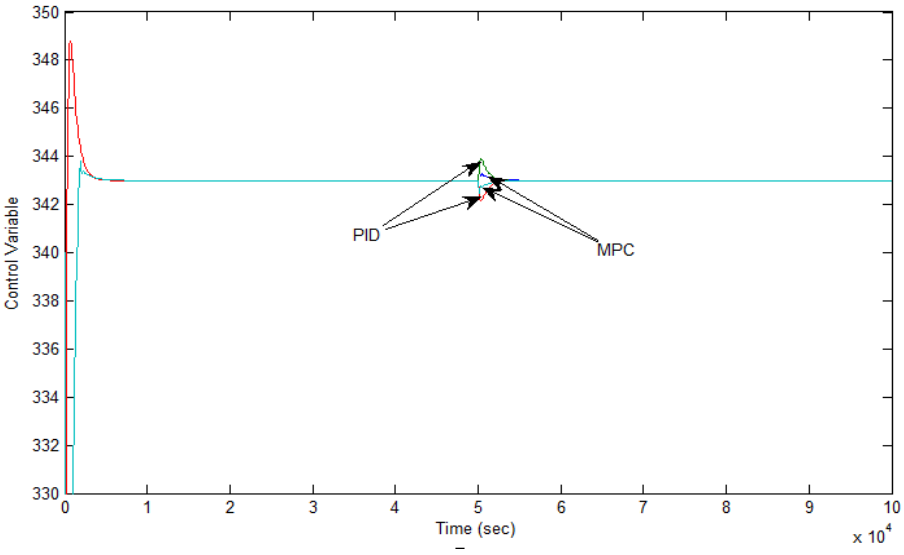

Fig 9: Load change in PID and MPC

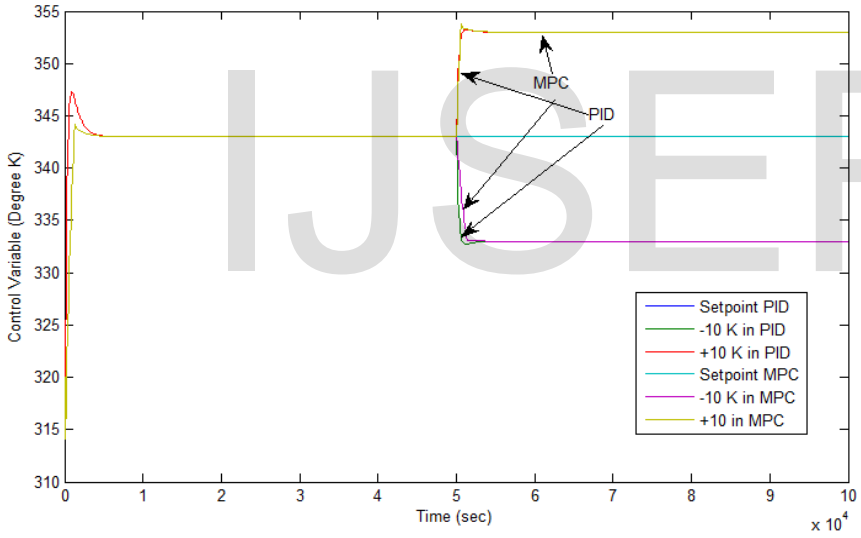

Fig 10**:** Set-point change in PID and MPC

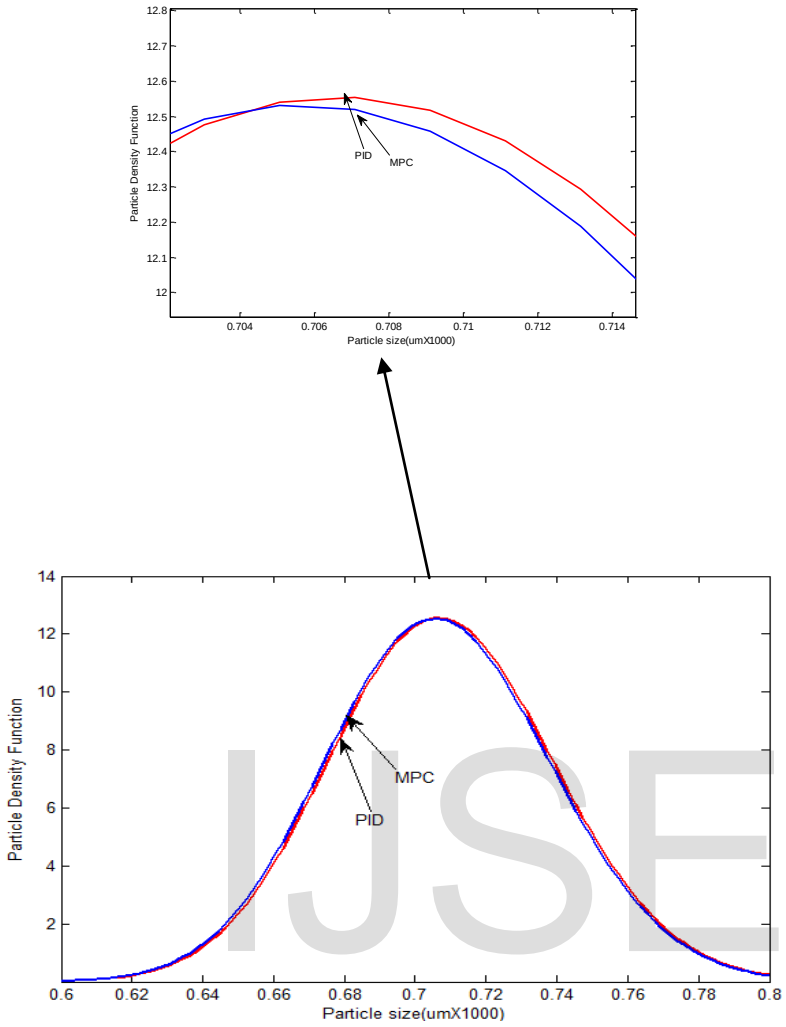

Fig 11: Particle density function as a result of controllers (Blue-MPC, Red- PID)

It can be concluded that synthesizing an optimal open loop assuming perfect modeling would leads to important errors between real and desired PSD's, due to impurities, changes in raw materials or due to degradation of the process components. Meanwhile, the closed-loop SISO MPC strategy is a much batter than PID since it reduces the impact of error.

The main advantage of MPC over PID controllers are its ability to handle constraints, non-minimum phase processes, changes in system parameters and its straightforward applicability to large, multivariable processes.

Conclusion

The challenging features in batch emulsion polymerization are highly non-linear nature, insufficient measurements, unmeasured disturbance and process model-mismatch; Inspite of all this we have formulated non-linear model predictive control for the control of batch emulsion polymerization within target points i.e. temperature of the reactor at 373 K and compared with PID controller. MPC was able to track the optimum reactor temperature profile efficiently and without a noticeable overshoot as in the case of PID, better disturbance rejection and smoother control move were also shown by MPC. This study shows advanced controllers are more robust and faster than traditional controller in the sense of fast response, better regulation and less sensitive controller output.

Symbols used

1. A_H [m^2] heat transfer area
2. Cj [J/Kg-K] heat capacity of water
3. Fhot [l/min] flow rate of the hot
4. Fcold [l/min] cold streams fed to the circulating water
5. Hs =enthalpy of incoming hot water
6. hc = enthalpy of liquid cooled
7. k0 [m3/mol-min] Radical exit rate.
8. Ktr [me/mol-min] Termination rate
9. M [kg]= molecular weight of hot water
10. N_A 6.0221x 10^{23} Avogadro number
11. Q [W] Heat duty
12. Tj [Degree Kelvin] temperature of cooling water in jacket,
13. Uj [w/m2-K] Internal energy of hot water in jacket
14. U [W/m2-k]overall heat transfer coefficient

15. ρj [kg/m3]density of water

16. Tj0 [Degree Kelvin] inlet cooling water temperature

References

[1] Cueli, J. R. and C. Bordons, *Control Engineering Practice* **,**16:,1023-1034 [2008]

[2] Garcia, C. E., D. M. Prett, et al. , *Atomatica* ,25, 335-348[1989]

[3] Harnischmacher, G. and W. Marquardt , *Control Engineering Practice* ,15,1238-1256[2007]

[4] Hosen, M. A., M. A. Hussain, et al. , *Control Engineering Practice* ,19, 454-467 [2011]

[5] Nagy, Z. and S. Agachi , *Computer chem. Engg*, 21(6),571-591 [1996]

[6] Othman, N. S., S. Othman, et al., *Journal of process control* , 21, 861-873 [2011]

[7] Ozkan, G., H. Hapoglu, et al. ,*Chemical Engineering and Processing* ,37,125-139 [1998]

[8] Peterson, T., Y. Hernandez, et al. , *chem Eng Sci* ,47, 737-753 [1992]

[9] Yuce, S., A. Hasaltan, et al. , *IChem* ,77, 413-419 [1999].

[10] Ohmura, N.,T. Horie,et al.,*Chem. Eng. Technol.*,35(7),1273-1280 , DOI : 10.1002 / ceat.201200034 [2012]

List of Tables

Table 1: Energy balance for Reactor and jacket used in model

Table 2: Initial values of process variables.

Table 3: Tuning parameters for MPC

List of Figures