

Research paper

Modelling and control of crystallization process

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Abstract

Batch crystallizers are predominantly used in chemical industries like pharmaceuticals, food industries and specialty chemicals. The nonlinear nature of the batch process leads to difficulties when the objective is to obtain a uniform **Crystal Size Distribution (CSD)**. In this study, a linear PI controller is designed using classical controller tuning methods for controlling the crystallizer outlet temperature by manipulating the inlet jacket temperature; however, the response is not satisfactory. A simple PID controller cannot guarantee a satisfactory response that is why an optimal controller is designed to keep the concentration and temperature in a range that suits our needs. Any typical process operation has constraints on states, inputs and outputs. So, a nonlinear process needs to be operated satisfying the constraints. Hence, a nonlinear controller like **Generic Model Controller (GMC)** which is similar in structure to the PI controller is implemented. It minimizes the derivative of the squared error, thus improving the output response of the process. Minimization of crystal size variation is considered as an objective function in this study. Model predictive control is also designed that uses advanced optimization algorithm to minimize the error while linearizing the process. Constraints are fed into the MPC toolbox in MATLAB and Prediction, Control horizons and Performance weights are tuned using Sridhar and Cooper Method. Performances of all the three controllers (PID, GMC and MPC) are compared and it is found that MPC is the most superior one in terms of settling time and percentage overshoot.

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Keywords: Batch crystallization; Crystal size distribution; Crystal size variation; Optimal control; Constraints; Model predictive control

1. Introduction

Crystallization is one of the oldest and widely used unit operations in chemical industry. It is commonly used in pharmaceuticals, fine chemicals and food industries. It is usually the first step in any separation process and produces high-purity products. The batch crystallization mode is widely adopted in industries for the production highly pure products in small quantities. The methodologies of batch crystallization modelling are well established and the control strategies have undergone many refinements over the past decades. Due to advancement of sensor technology, measurement of solution

concentration has been possible and the advanced control concepts related to model free controller design and robust control of crystal shape and size have been developed [1].

1.1. Operations of batch crystallizer

A pictorial representation of batch cooling crystallizer is shown in Fig. 1, in which the crystals are formed when the solubility of a solute in a solution decreases as a result of cooling the solution. Decrease in the solubility can also be achieved by evaporating the solvent from the solution or by adding another solvent which precipitates the solute. The crystallization process consists of two major events, nucleation and crystal growth which continue to occur simultaneously, and their rate is driven by the existing supersaturation in the solution. Supersaturation can be altered by changing the operating conditions [2]. Depending upon the conditions, either nucleation or growth may be predominant over the other, and as a result, crystals with different sizes and shapes are obtained. As the time progresses, supersaturation level decreases and a stage

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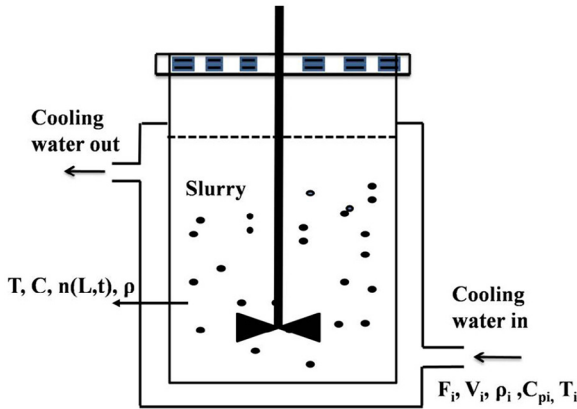


Fig. 1. Jacketed batch cooling crystallizer.

of equilibrium is reached where the crystallization process reaches a steady state. After the batch time, the slurry is discharged from the crystallizer and crystals are separated by filtration and dried. The size of the product formed is very important since it helps in increasing the efficacy of downstream processes like filtration, washing etc. Hence, the prime objective of crystallizer is to obtain the crystals with narrower size distribution.

The operating region in the crystallizer can be identified between a series of clear points (solubility) and cloud points (metastable limit) at various temperatures. For every solute–solvent combination, this can be obtained by experimentation and the region bounded by these curves is termed as the metastable zone. In this zone, spontaneous nucleation does not occur but it can be controlled. Hence, in cooling crystallization the solute concentration profile is maintained well within the metastable zone and close to the solubility curve in order to promote crystal growth and avoid spontaneous nucleation.

The classical approach to control is based on first principles models in which a model is constructed from material, energy and population balances ignoring agglomeration and breakage phenomenon. The growth and nucleation kinetics need to be accurately determined and the model should give the robust and accurate description of process. The direct design approach to control is based on following a desired trajectory within the operating region. The desired trajectory of the crystallizer falls in the metastable zone which is bounded by solubility and metastable curves. Here, a concentration trajectory as a function of temperature is followed with simple feedback control system which is very simple to adopt and does not require the accurate prediction of nucleation and growth kinetics [3–5].

The objective of this study is to analyse and compare the performance of the conventional and the advanced controllers which track the desired cooling profile. The temperature control strategy is the most widely adopted technique because of its simplicity since it requires only temperature of the crystallizer to be controlled and is used in the present study. Potassium nitrate–water system is taken as a model system and the inlet jacket temperature is taken as the manipulated variable. The paper is organized as follows. In section 2, population balance

modelling of batch cooling crystallizer, and method of moments is discussed. In section 3, the design principles of conventional PID controller, Generic Model Controller (GMC) and the Model Predictive Controller (MPC) are presented and the performance of these controllers is discussed in the subsequent section 4.

2. Population balance modelling of batch cooling crystallizer

Modelling of the batch crystallizer is done by first-principles method by solving Population Balance Equation (PBE), mass and energy balance equations simultaneously [6–8]. PB equation describes the behaviour of particulate system like crystallizer. It consists of partial differential equation which describes the evolution of CSD as a function of time and the crystal size. One dimensional PBE for a batch crystallization system is given as

$$\frac{\partial n(L, t)}{\partial t} + \frac{\partial Gn(L, t)}{\partial L} = B_0 \quad (1)$$

where $n(L, t)$ is the number density function ($1/m^4$) which describes the number of crystals per crystal length and volume of slurry, t is the time (s), L is the characteristic size of crystals (m), G is the crystal growth rate (m/s) and B_0 is the nucleation rate of crystals of infinitesimal size ($1/m^3s$). The initial and boundary conditions for solving the PBE are given as

$$n(L, 0) = 0, \quad n(0, t) = \frac{B_0}{G} \quad (2)$$

If growth rate is assumed to be independent of crystal length and initial birth rate is considered to be negligible, the simplified form of PBE can be written as

$$\frac{\partial n(L, t)}{\partial t} + G \frac{\partial n(L, t)}{\partial L} = 0 \quad (3)$$

The solute mass balance equation provides the change of concentration inside the crystallizer as the time progresses. The mass balance is written in the form of ordinary differential equation

$$\frac{dC}{dt} = -3\rho_c K_v \int_0^\infty nGL^2 dL \quad (4)$$

where C is the solute concentration, ρ_c is the density of crystals and K_v is the volumetric shape factor. The energy balances for the suspension and the cooling jacket are given below

$$\frac{dT}{dt} = \frac{-UA}{MC_p} (T - T_i) - \frac{3\rho_c K_v \Delta H}{C_p} \int_0^\infty nGL^2 dL \quad (5)$$

$$\frac{dT_i}{dt} = \frac{F_i}{V_i} (T_{isp} - T_i) + \frac{UA}{V_i C_{pi}} (T - T_i) \quad (6)$$

where T is the temperature of the crystal suspension at any time t , T_i is the time dependent temperature of fluid in the jacket (water), F_i , V_i , ρ_i and C_{pi} are respectively the flow rate, volume, density and specific heat of the inlet jacket fluid respectively,

Table 1
Parameters used for simulation of crystallization process.

Parameters	Definition	Value	Units
b	Nucleation rate exponent	1.78	
K _b	Nucleation rate constant	4.64*10 ¹¹	1/m ³ .s
g	Growth rate exponent	1.32	
K _g	Growth rate constant	1.1612*10 ⁻⁴	m/s
C ₀	Initial concentration of solution	18	kg KNO ₃ /kg water
ρ _c	Density of crystals	2110	kg/m ³
ρ _i	Density of water	1000	kg/m ³
K _v	Volume shape factor	1	
M	Mass of slurry	30	kg
C _p	Specific heat capacity of solution	95.06	J/kg.K
C _{pi}	Specific heat of water	4184	J/kg.K
V _i	Volume shape factor	0.905	m ³
F _i	Flow rate of water	0.001	kg/s
U.A.	Areas times overall heat transfer coefficient	0.125	J/K.s
μ ₀ (0)	Zeroth moments (total number of crystals)	10 ⁻⁶	
μ ₁ (0)	First moment (total length of crystals)	10 ⁻⁶	
μ ₂ (0)	Second moment (total area of crystals)	10 ⁻⁶	
μ ₃ (0)	Third moment (total volume of crystals)	10 ⁻⁶	
n (L, t)	number density function which describes the number of crystals per crystal length and volume of slurry		(1/m ⁴)
S	Supersaturation which defines the driving potential for the nucleation and growth to occur		

T_{isp} is the set point temperature provided, U and A are the overall heat transfer coefficient and area available for heat transfer respectively, ρ is the density of the slurry and ΔH is the heat of crystallization. The growth rate G and birth rate B rate at any time t in the crystallizer are obtained from empirical equations

$$G = K_g.S^g \tag{7}$$

$$B = K_b.S^b \tag{8}$$

where K_g and g are growth rate parameters and K_b and b are birth rate parameters and S is the supersaturation which is defined as the driving potential for nucleation and growth to occur. In terms of concentration, S = (C – C_s)/C_s, where C is the solution concentration and C_s is the concentration at saturated conditions.

2.1. Method of moments

PBE as described in equation (1) is in the form of a partial differential equation. The numerical schemes for solving the PBE are very complex and time consuming. In order to reduce the model complexity, it is advisable to convert this partial differential equation into a set of ordinary differential equations. The most widely used method to reduce the complexity is to use the method of moments. In principle, the method of moments defines the ith moment in terms of the population density function by

$$\mu_i = \int_0^\infty n(L, t)L^i(t)dL \tag{9}$$

where μ_i is the ith moment and n(L, t) is the population density function. Usually, first four moments are needed for solving

the model equations, which have the following physical significance; μ₀ represent total crystal number per unit volume, μ₁ represent total crystal length per unit volume, μ₂ represent total crystal surface area per unit volume, μ₃ represent total crystal volume per unit volume. The rate equations for different moments can be written as

$$\frac{d\mu_0}{dt} = B(t) \tag{10}$$

$$\frac{d\mu_i}{dt} = i * G(t) * \mu_{i-1}^n \tag{11}$$

All the above mentioned ordinary differential equations, mass and energy balance equations are solved in **SIMULINK environment of MATLAB software using ODE45 solver** as well as forward Euler method. The parameters are taken from Miller [9], for carrying out the simulation and is shown in Table 1. Metastable zone width is assumed to be constant having a value of ΔT = 2.2 K for temperature range used. The saturation concentration (C_s) for KNO₃–water system is given by the equation

$$C_s = 0.1286 + 0.00588.T^2 + 0.0001721.T^3 \tag{12}$$

3. Design of controllers

3.1. PID controller design

Crystallization process can be controlled by two ways; either by controlling the crystallizer temperature or by controlling the crystallizer concentration. Inlet jacket temperature is used as the manipulated variable. As the jacket temperature is changed, concentration and temperature of the crystallizer is changed. Since, concentration control requires some expensive sensors, temperature control is widely adopted in literatures. A basic

PID controller is designed for temperature control strategy. Output temperature of the crystallizer is fed back and difference between the set point and measured output is calculated. This error signal is then fed to the controller and an actuating signal is then generated from the controller so that the output reaches the set point. Zeigler–Nichols tuning settings are used for tuning the PID parameters.

3.2. Generic model control (GMC)

A PI controller is also designed for this case. But the response was not satisfactory. It took a lot of time for the system to reach steady state value. Controller efficiency was not improving even on changing the parameter values. So there was a need of designing a different controller which is as simple in structure as a PI controller but which will give a good output response. Hence, a Generic Model Controller for temperature control of exothermic batch crystallizer is designed in which rate of change of temperature is used as a manipulated variable.

3.2.1. Generic model controller algorithm formulation

GMC has several advantages that make it better from other control algorithms. The process model appears directly in the control algorithm and it does not need to be linearized before use, allowing for the inherent nonlinearity of the operation to be taken into account. It can be used for discrete as well as continuous process plant model. By design, GMC provides feed-back control of the rate of change of the process output. This suggests that the rate of temperature change (output) can be used directly as a manipulated variable. The relationship between feed forward and feedback control is explicitly stated in the GMC algorithm. Finally and importantly, the GMC framework permits us to develop a control algorithm that can be used for both heat-up and temperature maintenance and therefore eliminates the need for a switching criterion between different algorithms; which makes it very robust in nature.

The Generic Model Control (GMC) algorithm derived for the control of batch reactor (Lee and Sullivan, 1988) is used for control of crystallizer in this study [10]. GMC requires a dynamic model of the process written in standard state variable form. The controller is formulated by solving the dynamic process model for the derivative of the controlled variable, y , and letting it equal what is, in effect, a proportional integral term operating on the difference between the current value of y and its desired value, y_d . The desired trajectory can be assumed as,

$$\dot{y}_d = K_1(y_d - y) + K_2 \int (y_d - y) dt \quad (13)$$

where K_1 and K_2 are tunable parameters. Consider a non-linear system

$$\dot{x} = f(x) + g(x)u(t) \quad (14)$$

$$y = h(x)$$

where $f(x)$, $g(x)$ and $h(x)$ are vector fields. The scalar field describing the output function is simply the state itself. The performance index for GMC is given by [11],

$$I = \int (\dot{y}_d - \dot{y})^2 dt \quad (15)$$

where \dot{y} and \dot{y}_d denote the derivative of the process output and the desired trajectory. To minimize the performance index, let dI/dt equals zero, then

$$(\dot{y}_d - \dot{y}) = 0 \quad (16)$$

Now, \dot{y} can be written as,

$$\frac{dy}{dt} = \frac{dy}{dx} * \frac{dx}{dt}$$

$$\text{Now, } \frac{dy}{dx} = \frac{dh(x)}{dx} \text{ and,}$$

$$\frac{dy}{dt} = \frac{dh}{dx} * f(x) + \frac{dh}{dx} * g(x) * u(t) \quad (17)$$

Combining the above equations and putting in Equation (16), we will get

$$K_1(y_d - y) + K_2 \int (y_d - y) dt - \left\{ \frac{dh}{dx} * f(x) + \frac{dh}{dx} * g(x) * u(t) \right\} = 0 \quad (18)$$

From the above equation, $u(t)$ can be calculated in terms of K_1 and K_2 .

3.2.2. Tuning of the parameters

Several methods have been proposed to find the values of K_1 and K_2 . The common methods used are

1. Finding the ultimate gain and ultimate period of the output response and multiplying by a scalar.
2. By calculating the damping coefficient (ξ) and time constant (τ) of the desired trajectory. K_1 and K_2 can be written as:

$$K_1 = 1/\tau^2 \text{ and } K_2 = 2\xi/\tau$$

3. By trial and error method.

3.3. Model predictive control (MPC)

In chemical industries, there are certain inequality constraints on input and output variables that need to be considered for optimal operation. So, in order to follow those constraints, some advanced controllers need to be designed that follow advanced optimization algorithm to minimize the error. Model predictive controller predict the future output if an accurate dynamic model of process is available and then depending on predicted and measured outputs, appropriate change in measured input can be calculated.

Basic structure of MPC is given in Fig. 2. Current values of the output variables are calculated using a process model described in section 2 and then difference between predicted and actual outputs are used as a feedback signal to a prediction block. The predicted outputs are used in controlled calculation and set point calculation after considering constraints on the input and output variables. MPC configuration is analogous to both internal model control configuration and smith predictor

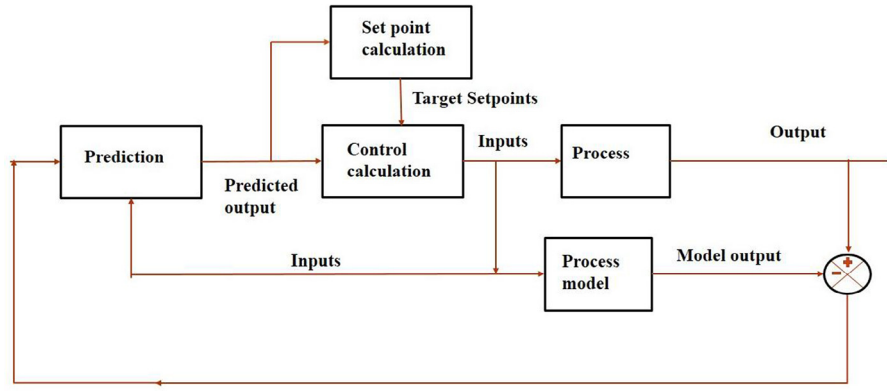


Fig. 2. Block diagram of MPC.

configuration because model and process are parallel acted and difference act as feedback control signal. But coordination of the control and set point calculations makes MPC superior than others. Moreover, MPC is largely used in MIMO control problems than IMC or Smith predictor.

The main objective of the MPC is to determine the set of control moves so that predicted model output reaches the desired set point. At any sampling time, MPC decides a set of M input values containing a present input $u(k)$ and future $(M-1)$ inputs. Set of inputs are calculated in such a way that a set of P predicted output should reach the optimum set point. Objective function is optimized to find out control moves. Number of control moves M is called the control horizon while number of prediction P is referred to as the prediction horizon.

The least square objective function for a prediction horizon P and a control horizon M is written as follows,

$$J = Q \cdot \sum_{i=1}^P (r_{k+i} - \hat{y}_{k+i})^2 + R \cdot \sum_{i=0}^{M-1} \Delta u^2_{k+i} \quad (19)$$

where \hat{y} denotes the predicted model outputs, ' r ' is the set point, Δu is the change in manipulated input from one sample time to the next, ' Q ' and ' R ' is a weight for the change in the output and manipulated input respectively. The optimization problem deals with a minimization of the objective function by manipulating control moves.

3.3.1. Tuning strategy review

1. Approximate the process dynamics of all controller output–process variable pairs with FOPDT models (Shridhar and Cooper, 1997) [12].

$$\frac{y(s)}{u(s)} = \frac{K \cdot e^{-\theta s}}{\tau s + 1}$$

2. Select the sample time as close as possible to $T = \text{Min}(0.1\tau, 0.5\theta)$
3. Compute the prediction horizon (P) and control horizon (C) using

$$P = \frac{5\tau}{T} + \frac{\theta}{T} + 1 \quad c = \frac{\tau}{T} + \frac{\theta}{T} + 1$$

MPC toolbox model advisor is used to calculate the performance weights. MPC toolbox of the SIMULINK environment is used to carry out the simulation. Input and output constraints are provided and weights and gain are tuned to find out the optimal performance.

4. Results

A classical PID controller to control the temperature of the crystallizer has been proposed in which temperature of the jacket fluid is taken as the manipulated variable to control the crystallizer temperature. The parameter values for the PID controllers were obtained minimizing the integral square error (ISE). The response of PID controller for the set point tracking is shown in Fig. 3. Ultimate period and gain are calculated by increasing the value of K_c until a sustained oscillation curve is obtained. Zeigler–Nichols tuning settings are given by $K_c = 0.6$, $\tau_i = P_u/2$, $\tau_d = P_u/8$. For this case, it is found out to be $K_c = 1.8$, $\tau_i = 0.6$, $\tau_d = 1.35$. The controller is tracking the set point changes for different types of step changes provided. But, the overshoot appears to be higher and it takes a minimum of 53 min for the response to settle at steady value. The response of GMC for set point tracking is shown in Fig. 4. It provides a better response when compared with PID controller. The overshoot has reduced and the response settles after 49 min. Both controllers were compared (Fig. 5, Table 2) in terms of percentage overshoot, settling times and it was found that GMC response was quite better than that of the PID. By designing a MPC controller with prediction horizon as 51 and control horizon as 11 with sampling times of 0.5 s for 2000 s, controlled state variables of batch crystallization process are obtained and are given in Fig. 6. There is a drastic decrease in

Table 2
Comparison between GMC, PID and MPC.

Controller	Percent overshoot	Settling time (mins)	ISE
PID	18%	53	594.55
GMC	10%	49	284.39
MPC	1%	16.67	50.73

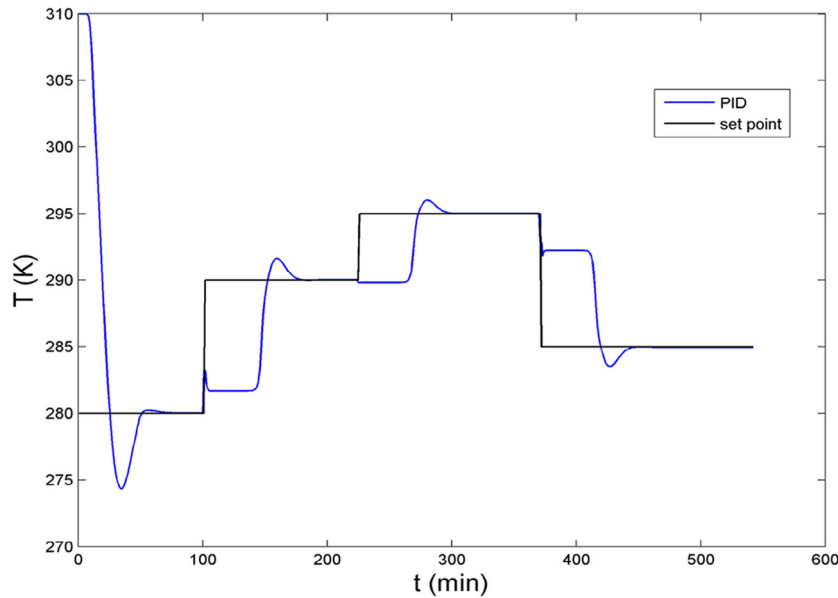


Fig. 3. PID response for set point tracking.

the percentage overshoot and the response settles sooner when compared with other controllers. The MPC response is quite better than other controllers in tracking the set point.

5. Conclusions

The primary objective of this study is to evaluate the opportunities for model based control to improve product quality and process productivity of industrial crystallizers. All the controllers have been successfully implemented and the output

response is satisfactory. Temperature set point tracking is achieved for every case by keeping the thermodynamics of the system in mind. Simulation is done for quite a number of operating points. The MPC toolbox of MATLAB simulation block is used for implementing the MPC controller. Several constraints on input and output variables are provided. Performance of MPC is then compared with that of PID and Generic Model controller (GMC) and it is found that MPC gives a better response based on percent overshoot, settling time and integral squared error.

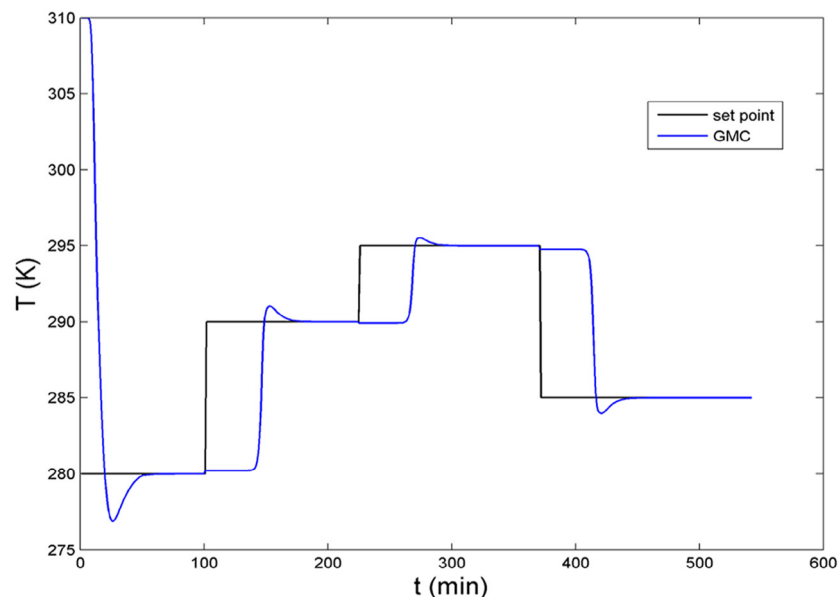


Fig. 4. GMC response for set point tracking.

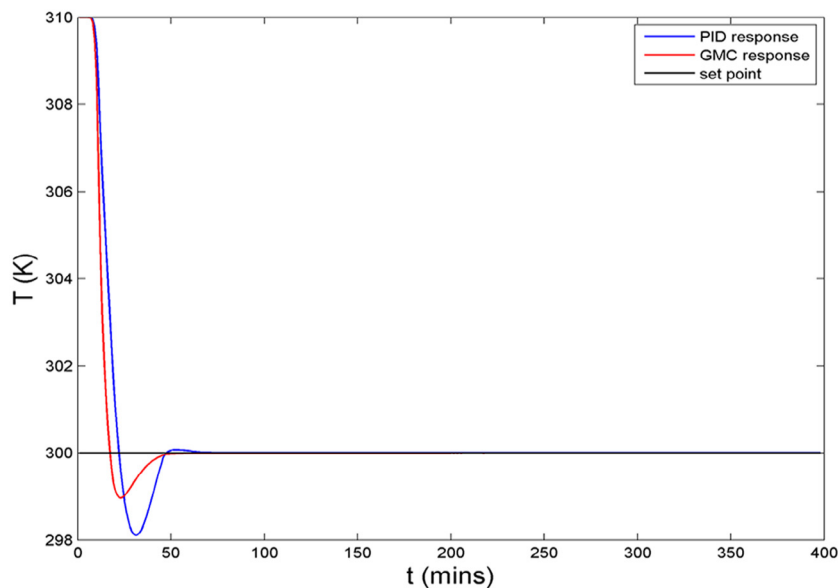


Fig. 5. Comparison of PID and GMC responses for temperature set point tracking.

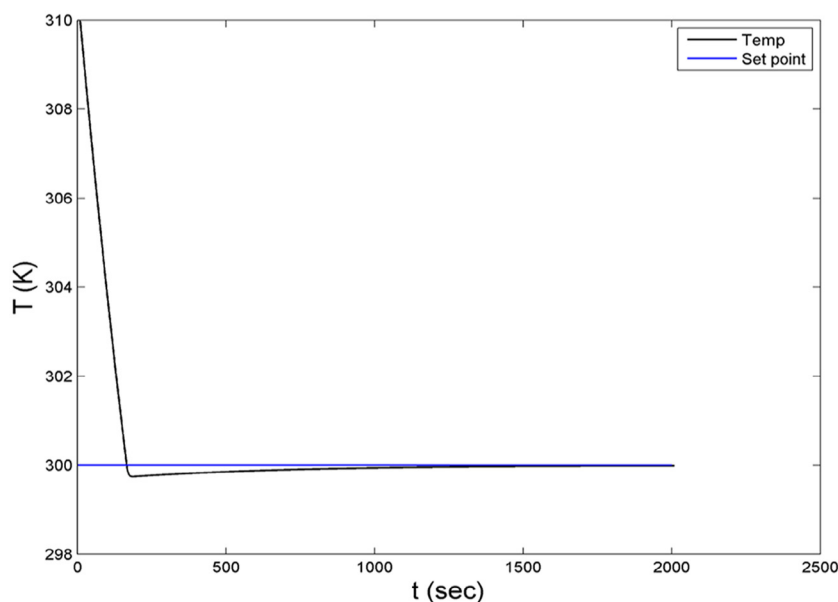


Fig. 6. MPC response for set point tracking.

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