

Article

Hybrid Neural Network Controller Design For a Batch Reactor to Produce Methyl Methacrylate

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Abstract. Methyl methacrylate (MMA) production in an exothermic batch reactor provides a challenging problem for studying its dynamics behavior and temperature control. This work presents a neural network forward model (NN) to predict a concentration of methyl methacrylate, a jacket temperature and temperature profile in the reactor. An optimal NN model has been employed to predict state variables incorporating into a model predictive control (MPC) algorithm to determine optimal control actions. To control the temperature, neural network based control approaches: a neural network direct inverse control (NNDIC) and a neural network based model predictive control (NNMPC) have been formulated. In addition, a dynamic optimization approach has been applied to find out an optimal operating temperature to achieve maximizing the MMA product at specified final time. The robustness of the proposed control strategy is investigated with respect to changes in operating parameters: rate of reaction, heat of reaction and overall heat transfer coefficient. Simulation results have indicated that the NNMPC is robust and gives the best control results among the PID and NNDIC in all cases.

Keywords: Methyl methacrylate, batch reactor, neural network based model predictive control, neural network direct inverse control, dynamic optimization.

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1. Introduction

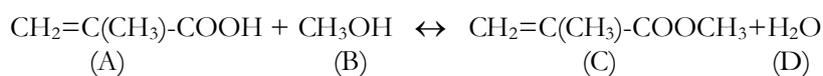
Methyl methacrylate (MMA), which is used to produce polymethyl methacrylate (acrylic plastics) and polymer dispersions, is an important chemical polymer intermediate. The world production capacity has been double increased in past 15 years and the MMA demand is still expected growth in the future [1]. The MMA can be manufactured by many routes. An esterification of methacrylic acid with methanol in a batch reactor is a successful route on account of achieving the maximum yield. In a batch reactor with exothermic reactions, the heat-released of reactions in heating period may become very large very quickly and the heat-generated can exceed the cooling capacity of the reactor and then runaway reactions take place [2-3]. To overcome the problem, model based control strategies have been proposed to solve the problem [4-7]. However, control performances of such control strategies rely upon the accuracy of mathematical models developed.

To overcome the problem, neural networks studied and provided successfully to capture the dynamics of nonlinear and complex systems have been proposed and formulated [8-13]. Neural networks have several advantages of distributed information processing and the inherent potential for parallel computation. The potential for the processing and approximation relates to operating data without the prior knowledge of the process. They can learn adequately accurate models and give good non-linear control when model equations are not known or only partial state information is available. Neural networks can be employed to be mathematical model, estimator and controller. Kittisupakorn et al. [14] demonstrated dynamic neural network modeling for hydrochloric acid recovery acid process to predict the concentration profile of a hydrochloric acid recovery process consisting of double fixed-bed ion exchange columns. Rusinowski and Stanek [15] presented a method and example results of calculations of neural modeling of steam boilers. Charoenniyom et al. [16] applied neural network to be a modeling for the methyl methacrylate production process in a batch reactor and Thammasato et al. [17] proposed neural network modeling for a batch crystallizer. For the process control, Nueaklong et al. [18] investigated neural network modeling for hard chrome electroplating process to predict the plating solution temperature in hard chrome electroplating bath and applied the neural network inverse model as a controller for controlling plating solution temperature to the desired temperature range. Daosud et al. [19] presented the neural network for inverse model to be a controller for a steel pickling process. Kittisupakorn et al. [20] presented a multi-layer feedforward neural network based model predictive control for a steel pickling process. The neural network for forward model is applied as mathematical model to predict the state variables in the model predictive control algorithm. For use the neural network as an estimator, Arpornwichanop and Shomchoam [21] applied neural network as an estimator to estimate the unmeasured state variables for fed-batch bioreactors.

The goal of this paper is to provide an effective control technique to control the system. To achieve this, this paper has developed the neural network forward model to predict the dynamics behavior and the neural network inverse model to control the process integrated with the dynamic optimization. Both neural network forward and inverse models have been developed based on the Levenberg-Marquardt training algorithm. An optimal neural network structures for forward and inverse models are chosen based on mean square error (MSE). The obtained optimal neural network structure for forward model has been employed to predict state variables over a predictive horizon within a model predictive control (MPC) algorithm for searching optimal control actions via successive quadratic programming (SQP). Robustness tests of the proposed controls have been studied with respect to changes in operating parameters.

2. Mathematical Models for Methyl Methacrylate Production Process in a Batch Reactor

Mathematical models for esterification reaction of a methyl methacrylate in a batch reactor studied in this work have been developed. The esterification reaction involved in this study is given below.



It is assumed that the kinetic equation of the esterification reaction of methacrylic acid with methanol is reversible and the reaction takes place in liquid phase. From Witczak et al. [22], the reaction rate constants for this reaction are described by the following equations.

$$R_A = k_1 C_{cat} \frac{C_A C_B}{C_C^{0.3}} - k_2 C_{cat} C_C^{0.7} C_D \quad (1)$$

$$k_1 = 960 \exp\left(-\frac{63,600}{RT_r}\right) \quad (2)$$

$$k_2 = 600 \exp\left(-\frac{67,320}{RT_r}\right) \quad (3)$$

where C_A , C_B , C_C , C_D and C_{cat} refer to concentration of methacrylic acid, methanol, methyl methacrylate, water and catalyst in mol/m³, respectively.

The mass and the energy balances described the change of each component and temperatures are:

$$\frac{dC_A}{dt} = -R_A \quad (4)$$

$$\frac{dC_B}{dt} = -R_A \quad (5)$$

$$\frac{dC_C}{dt} = R_A \quad (6)$$

$$\frac{dC_D}{dt} = R_A \quad (7)$$

$$\frac{dT_r}{dt} = \frac{Q_r + Q_j}{C_r C_{pr}} \quad (8)$$

$$\frac{dT_j}{dt} = \frac{F_j p_j C_{pr} (T_{jsp} - T_j) - Q_j}{V_j p_j C_{pj}} \quad (9)$$

$$Q_r = -\Delta H R_A \quad (10)$$

$$Q_j = UA(T_j - T_r) \quad (11)$$

$$C_{pr} = \frac{C_{pA} C_A + C_{pB} C_B + C_{pC} C_C + C_{pD} C_D}{C_A + C_B + C_C + C_D} \quad (12)$$

where Q , ΔH and U are heat released from reaction (kJ/min), heat of reaction (kJ/kmol) and heat transfer coefficient (kJ/(min m² °C)), respectively.

3. Neural Network Modeling

Neural networks consist of an input layer, hidden layer(s) and an output layer with each layer composing of a processing unit. The input layer receives the external inputs from the outside of the network. The output layer will produce the output of the network according to the input data sets. In the hidden layers, the value from each input neuron is multiplied by a weight and is calculated by transfer function.

In this part, neural network forward model is applied to predict the dynamic behavior of the system. The training, testing and validating data sets for neural network modeling are generated from the first principle mathematical model covering several possible scenarios consisting of nominal and plant uncertainty cases. Rate of reaction, heat of reaction and overall heat transfer coefficient are considered as plant/model mismatches. In addition, the manipulated variable (the set point of the jacket temperature) is adjusted as step change and random change.

Table 1. Physical properties simulation system geometric characteristics.

Heat capacity of methacrylic acid (J/mol K)	167.817
Heat capacity of methanol (J/mol K)	81.080
Heat capacity of methyl methacrylate (J/mol K)	191.202
Heat capacity of water (J/mol K)	1000
Heat capacity of water in jacket (J/mol K)	1000
Density of methacrylic acid (kg/m ³)	1015
Density of methanol (kg/m ³)	791.8
Density of methyl methacrylate (kg/m ³)	940
Density of water (kg/m ³)	1000
Density of water in jacket (kg/m ³)	1000
Heat of reaction (J/mol)	-57500
Molecular weight of methacrylic acid (kg/kmol)	86.08
Molecular weight of methanol (kg/kmol)	32.04
Molecular weight of methyl methacrylate (kg/kmol)	100.12
Molecular weight of water (kg/kmol)	18.00
Volume of reactor (m)	0.0025
Overall heat transfer coefficient (J/sec m ² K)	274.42
Area of jacket (m ²)	0.05
Volume of jacket (m ³)	0.001
Gas constant (kJ/kmol K)	8.314
Flow rate of jacket (m ³ /sec)	3.5×10 ⁻⁵

After that, all generated data subsets are integrated to one overall set consisting of 4,500 patterns and the pattern in the overall set is randomized. The obtained data set are classified into 3 sets consisting of 60%, 30% and 10% of the obtained total data as a training, testing and validating data set, respectively. The number of samples for training, testing and validating data sets are shown in Table 2.

The generated data requires normalization for achieving a good performance neural network modeling. In the normalization step, all data were scaled in range of minimum and maximum value. The minimum and maximum data were compared to 0.05 and 0.95, respectively. The equations for data normalization and converting back can be expressed as follows:

$$x_{nor} = \frac{(x - x_{min})(0.95 - 0.05)}{(x_{max} - x_{min})} + 0.05 \quad (13)$$

$$x = \frac{(x_{nor} - 0.05)(x_{max} - x_{min})}{(0.95 - 0.05)} + x_{min} \quad (14)$$

where x and x_{nor} are the actual and normalized values, respectively.

In the training step, the Levenberg Marquardt algorithm is used to train the both neural network models. During the training, the neural network adjusts the weights and biases in each node connection but there does not adjust during the testing and validating to evaluate the neural network performance. The structure for the neural network forward model consists of 8 nodes in an input layer and 3 nodes in an output layer as shown in Fig. 1. The input layer nodes are composed of the past and present values of the reactor temperature, the concentration of methyl methacrylate, the jacket temperature and the set point of

the jacket temperature. The output layer nodes are the future values of the reactor temperature, the jacket temperature and the concentration of methyl methacrylate.

$C_c(k-1)$	$C_c(k)$	$C_c(k+1)$	Output
$T_r(k-1)$	$T_r(k)$	$T_r(k+1)$	
$T_j(k-1)$	$T_j(k)$	$T_j(k+1)$	
$T_{jsp}(k-1)$	$T_{jsp}(k)$		
			Input

Fig. 1. Input/output pattern for the forward model.

In the neural network design step, the appropriated neural network structure is defined by choosing the number of nodes in the hidden layer. Both neural network forward and inverse models have been developed based on the Levenberg-Marquardt training algorithm. The sigmoid function is used as the activation function of the nodes in the hidden layer and the linear transfer function is used as the activation function in its output layer. A common objective of the neural network training is to minimize an error between predicted neural network values and actual targeted values. An equation for MSE calculation is shown below:

$$MSE = \frac{1}{n} \sum_{i=1}^n (T_{aci} - T_{pi})^2 \quad (15)$$

where T_{ac} is the actual targeted values and T_p is the predicted neural network values.

After training step, the trained neural network model is tested with testing data sets. If the MSE values of the testing data are out of a specified value, the obtained neural network model is inapplicable and more training is required by reinitialize the weights and biases. Alternatively, another neural network structure is considered. On the other hand, if the MSE values of the testing data sets are satisfied, the obtained neural network model is validated by validating data set. If the MSE values of validating set are not out of a specified value, the network structure is changed by changing the number of hidden layers and the number of nodes in the hidden layer. In this paper, the number of hidden layer is varied from 1 node to 20 nodes.

Basic steps of the neural network designing are shown in Fig. 2. Many procedures of the neural network designing are summarized in this figure.

Table 2. The MSE of the testing and validation sets of forward and inverse models.

Data sets	Forward model (MSE)	Inverse model (MSE)	No. of samples
Training	4.6068 x10 ⁻⁵	3.6992 x10 ⁻⁵	2700
Testing set 1	3.5618 x10 ⁻⁵	2.0184 x10 ⁻⁵	675
Testing set 2	3.1400 x10 ⁻⁵	1.0795 x10 ⁻⁵	675
Validating set	5.0784 x10 ⁻⁵	2.0489 x10 ⁻⁵	450

4. Neural Network Direct Inverse Control (NNDIC)

In this part, a neural network inverse model is applied to formulate a neural network direct inverse controller to control the process. The detailed procedures to find the inverse neural network model are summarized in the Fig. 2. A neural network inverse model structure consists of 8 nodes in an input layer and a node in an output layer.

An optimal neural network inverse model is utilized to predict a manipulated variable (a set point of a jacket temperature). The neural network inverse model for the set point of the jacket temperature prediction requires the past and present values of the process outputs and the past values of manipulated variable and the set point of the reactor temperature. The input and output patterns for the neural network inverse model is shown in Fig. 3.

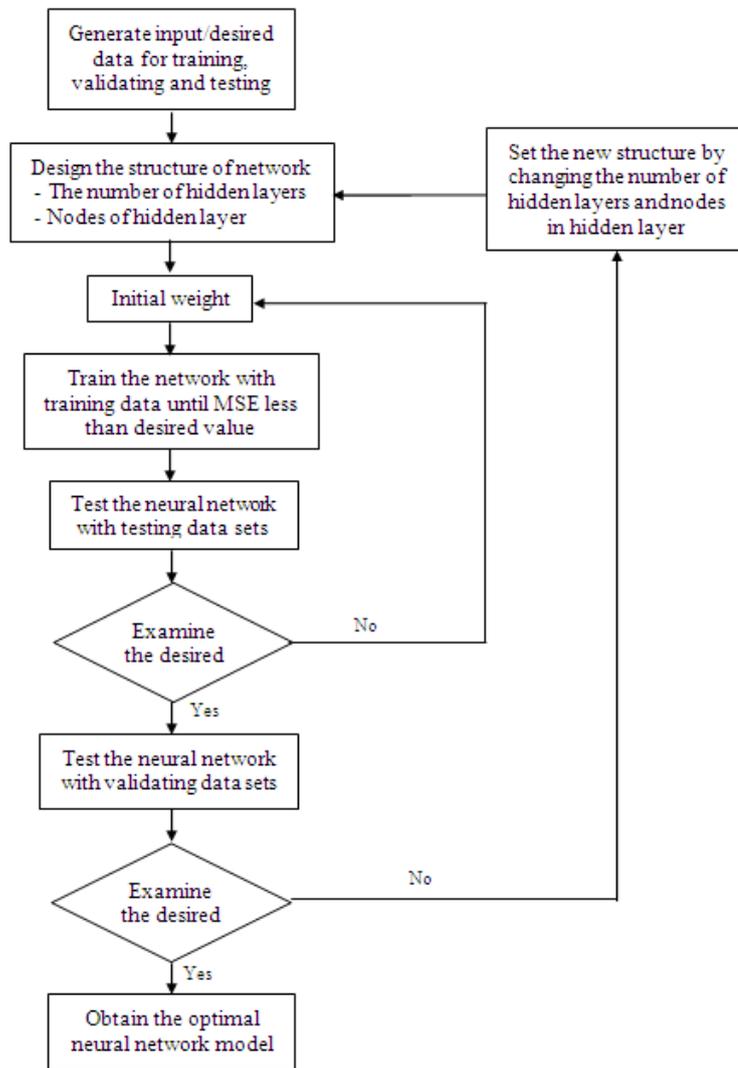


Fig. 2. Procedures of the neural network designing.

$C_c(k-1)$	$C_c(k)$		Output
$T_r(k-1)$	$T_r(k)$	$T_r(k+1)$	Input
$T_{jsp}(k-1)$	$T_{jsp}(k)$		
$T_j(k-1)$	$T_j(k)$		

Fig. 3. Input/output patterns for the inverse model.

The NNDIC strategy for controlling the temperature is shown in Fig. 4. The control performance is tested under the uncertainty of the process including changes of kinetic rate, heat of reactions and heat transfer coefficient.

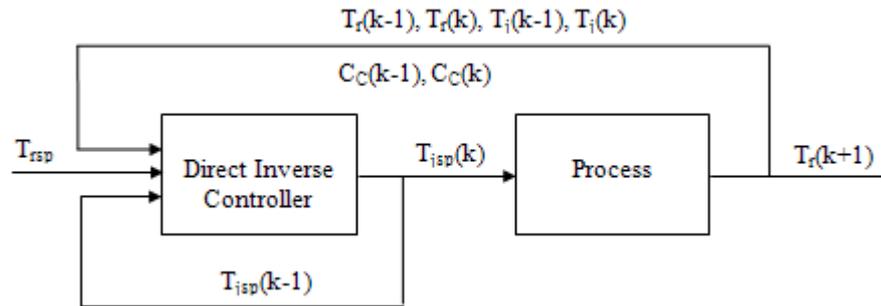


Fig. 4. The structure of NNDIC strategy.

5. Dynamic Optimization

The goal of a dynamic optimization problem is to find an optimal control profile of one or more control variables or control parameters of a system. Optimality is defined as a minimization or maximization of an objective function without violating given the process constraints [23-24]. In this work, a program is written to solve the optimization problem using a sequential quadratic programming (SQP) algorithm in Matlab Optimization Toolbox. The written program is tested to determine an optimal temperature of the exothermic batch reactor studied by Aziz et al. [25].

In this work, an objective is to determine the optimal temperature policy maximizing the amount of a desired product concentration for a given fixed batch time subject to bounds on the reactor temperature. The problem can be written mathematically as:

$$\max_{T_r} C_c(t_f)$$

Subject to

$$\dot{x} = f(x(t), T_r, p, t) \quad \text{process model} \quad (16)$$

$$x(t_o) = x(o) \quad \text{initial condition} \quad (17)$$

$$298K \leq T_r \leq 348K \quad \text{boundary condition} \quad (18)$$

$$t_f = t_f^* \quad (19)$$

where x is the vector of state variables, \dot{x} is the derivative of x with respect to time (t) and p is the process parameters. The batch time (t_f) is specified at 10 hours. The lower bound on the temperature is the initial temperature that operates at the ambient condition and the upper bound is dictated by the maximum temperature of the experimental data used by Witczak et al. [22].

6. Neural Network Based Model Predictive Control (NNMPC)

MPC appears to be one of general approaches which can handle most common process characteristics and industrial requirements in a satisfactory way [26-27]. The key success factor in the use of MPC in these process problems is the existence of accurate process models. [28-29] Then, the obtained neural network forward model in the neural network modeling part is applied as a predictor to predict the future values of outputs over a prediction horizon (p) within a model predictive control algorithm. An optimal manipulated variable (T_{jsp}) is determined by solving the optimization problem to minimize a specified objective function subject to the neural network model and lower and upper bounds of the manipulated variable (a set point of a jacket temperature). Figure 5 illustrates the structure of a neural network model predictive control (NNMPC) strategy. A program is used to solve the optimization problem using a successive quadratic programming (SQP) algorithm. The optimization problem is shown below:

$$\min_{T_{jsp}} \sum_{i=1}^p \left[W_1 \{T_r(k+i) - T_{rsp}(k+i)\}^2 + W_2 \{\Delta T_{jsp}(k+i)\}^2 \right] \quad (20)$$

Subject to

Neural network forward model

$$260 \text{ K} \leq T_{j\text{sp}}(k+1) \leq 380 \text{ K}, i = 1,2,3,\dots,P \quad (21)$$

$$T_r(k+p) = T_{\text{rsp}}(k+p) \quad (22)$$

where p is a parameter specifying the prediction horizon, M is the control horizon, W_i is the weighting parameter used to give different weights to different squared tracking error and T_{rsp} is the set point of the reactor temperature obtained off-line optimization.

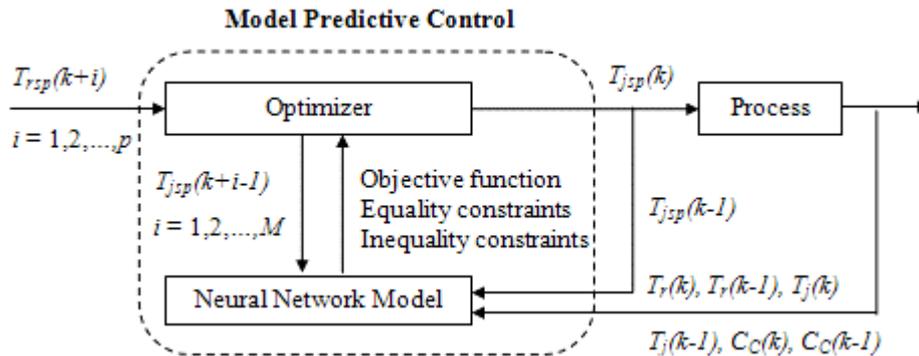


Fig. 5. The NNMP strategy.

7. Simulation Results

7.1 Neural Network Forward and Inverse Models

After training the neural network models, the neural network forward and inverse models are validated by the sets of validating data for the performance monitoring. Optimal structures for the forward and inverse models are [8-6-8-3] and [8-4-8-1] respectively and their MSEs for the testing and validating sets are presented in the Table 2. This table shows that the neural network forward model can predict the values of the concentration of MMA, the jacket temperature and the reactor temperature with great accuracy and the neural network inverse model gives good prediction of the set point of jacket temperature.

7.2 Dynamic Optimization

The dynamic optimization maximizing production concentration with respect to variations of time intervals: 1, 2, 4 and 8 intervals have been carried out. The simulation results with different time intervals are shown in Fig. 6. Table 3 reports the temperature and the concentration of MMA (desire product) of each time intervals. It has been found that at the final time, the maximum product achieves at the case of 8 intervals. The obtained optimal temperature profile is then applied as set points of the reactor temperature for control design.

Table 3. Optimization results

Interval	Off-line optimal temperature (K)	$C_c(t_i)$
1	342.45	13.932
2	348.00, 329.77	13.949
4	348.00, 347.98, 336.32, 325.21	13.952
8	348.00, 348.00, 347.47, 345.88, 341.60, 333.48, 328.15, 324.84	13.962

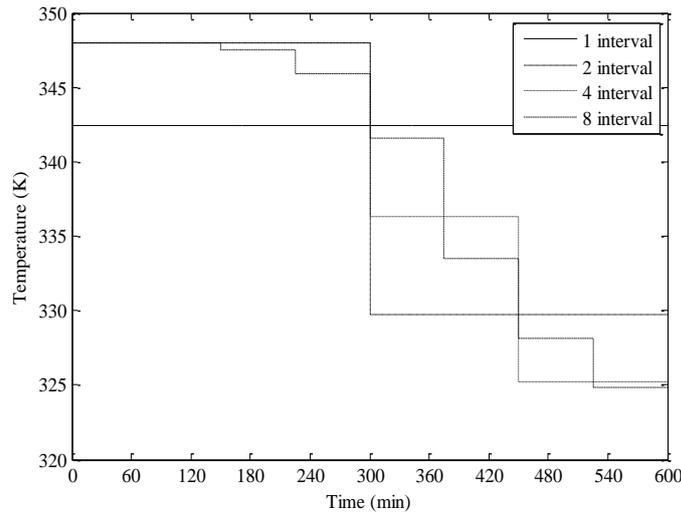


Fig. 6. Temperature profile for the optimization problem.

7.3 Neural Network Model Based Control

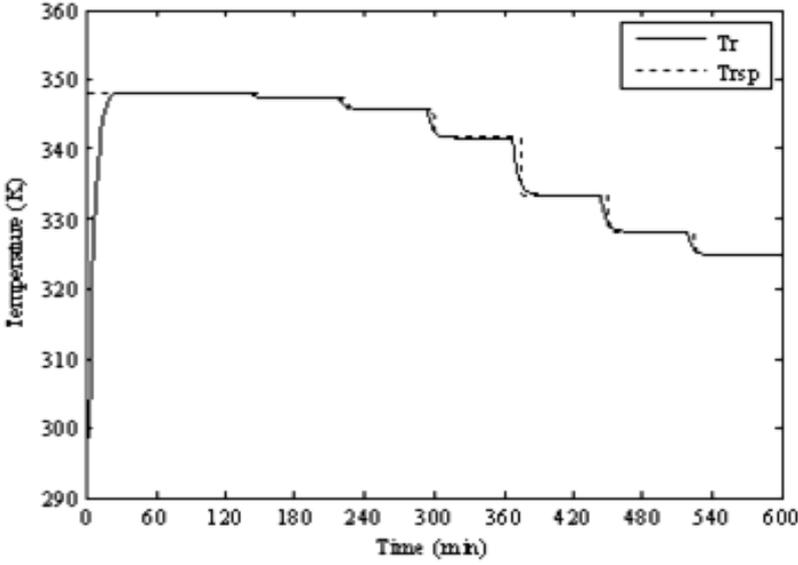
For this case, the controllers are adjusted with the same IAE for the same control performance. The objective of the control is to control the reactor temperature to the desired set point by adjusting the set point of a jacket temperature. The controlled simulations are divided into two cases which are nominal case and plant/model mismatch case. The conditions of the mismatch case are defined by 30% increasing and 30% decreasing of the model parameter values from the nominal condition (Table 1., Eq. (2) and Eq (3)) such as k_1 , k_2 , ΔH and U . The closed-loop performance of the NNMPC, the NNDIC and the PID control are indicated by the integral of absolute value of an error (IAE).

The simulation results of the nominal case with the process parameters presented in Table 1 indicate that the NNMPC can bring the temperature closely to the set point without overshoot, oscillations and offset as show in Fig. 7. In contrast, the NNDIC and the PID control cause the overshoot and oscillation of the control variable (reactor temperature) as show in Fig. 8 and 9, respectively. Moreover, the dynamic behavior of NNMPC manipulated variable is smoother than the NNDIC and the PID control.

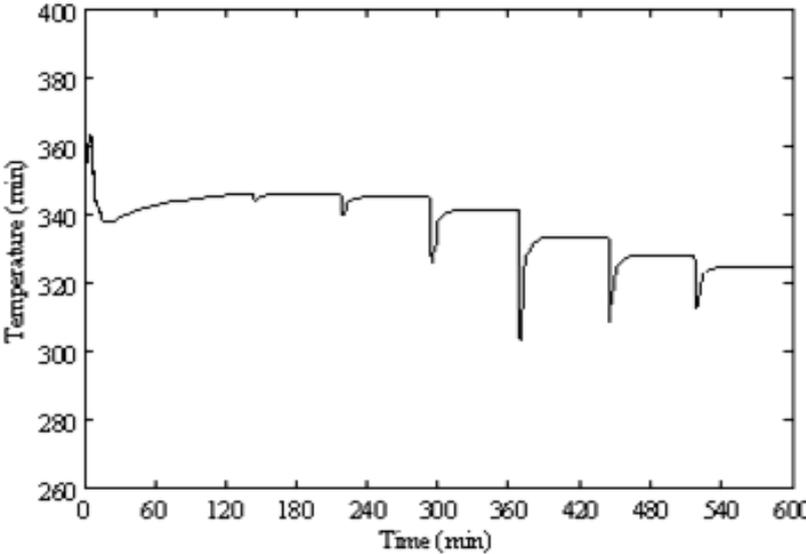
For the plant/model mismatch cases, a rate of reaction for forward reaction, a rate reaction for reverse reaction, a heat of reaction and an overall heat transfer coefficient are considered as the plant/model mismatch parameters because these parameters are prone to errors in the real process. Then the model mismatch cases are divided into 6 cases as follows

- 30% increasing of k_1 case
- 30% decreasing of k_2 case
- 30% increasing of k_1 and 30% decreasing of k_2 case
- 30% increasing of ΔH case
- 30% decreasing of U case
- 30% increasing of k_1 , 30% decreasing of k_2 , 30% increasing of ΔH and 30% decreasing of U case

The simulation results in the case of the decrease in U of 30% for the NNMPC, PID and NNDIC are shown in Fig. 10, 11 and 12, respectively. These figures illustrate that the NNMPC strategy can bring the reactor temperature to the set point by gradually adjusting the jacket temperature set point which then gives smooth control response. The NNDIC and PID, on the other hand, can control the reactor temperature to the set point but with drastic adjustment of the jacket temperature set point causing the oscillation and overshoot in the process response. For the control comparison, the NNMPC give the best control performance in all parameter mismatch cases and the NNDIC is more robust than PID control. The performance index in term of the absolute error (IAE) of three different controllers in nominal case and parameter mismatch cases are summarized in Table 4.

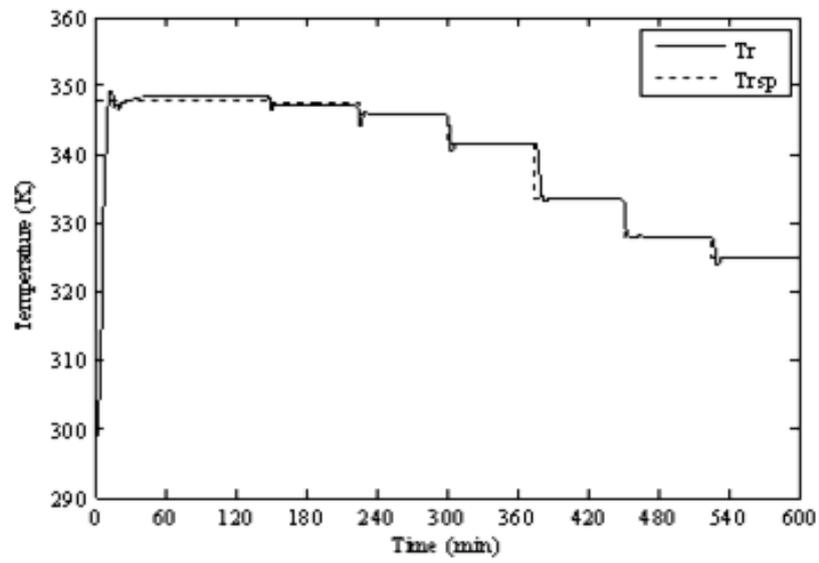


(a)

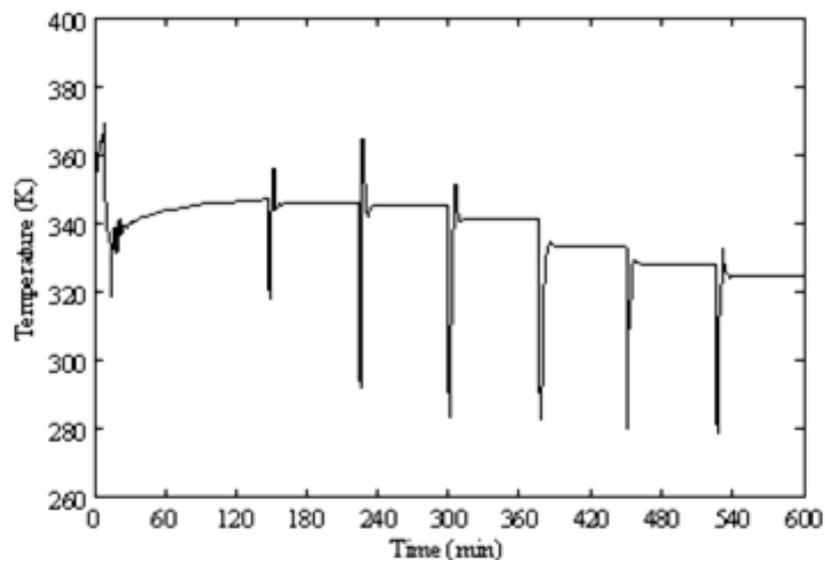


(b)

Fig. 7. The temperature control using NNMPC under the nominal case: (a) the control variable (T_r) and (b) the manipulated variable (T_{jsp}).

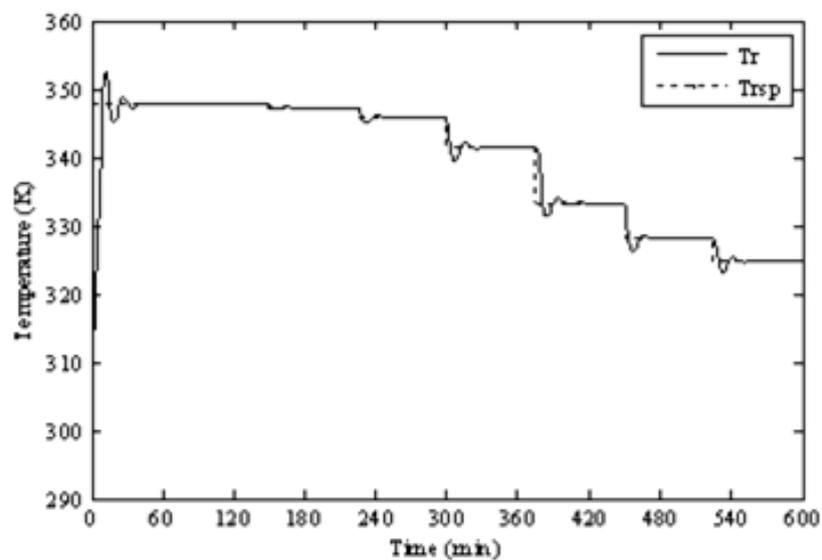


(a)

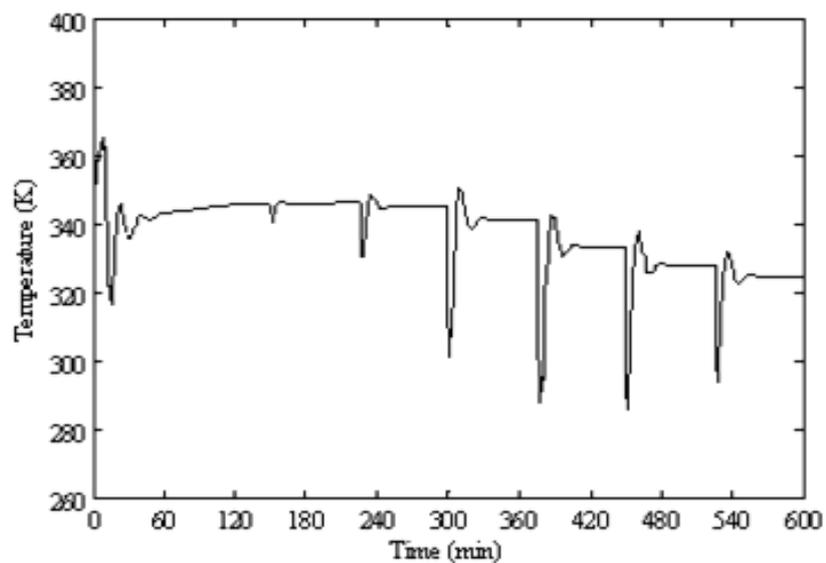


(b)

Fig. 8. The temperature control using NNDIC under the nominal case: (a) the control variable (T_r) and (b) the manipulated variable (T_{jsp}).



(a)

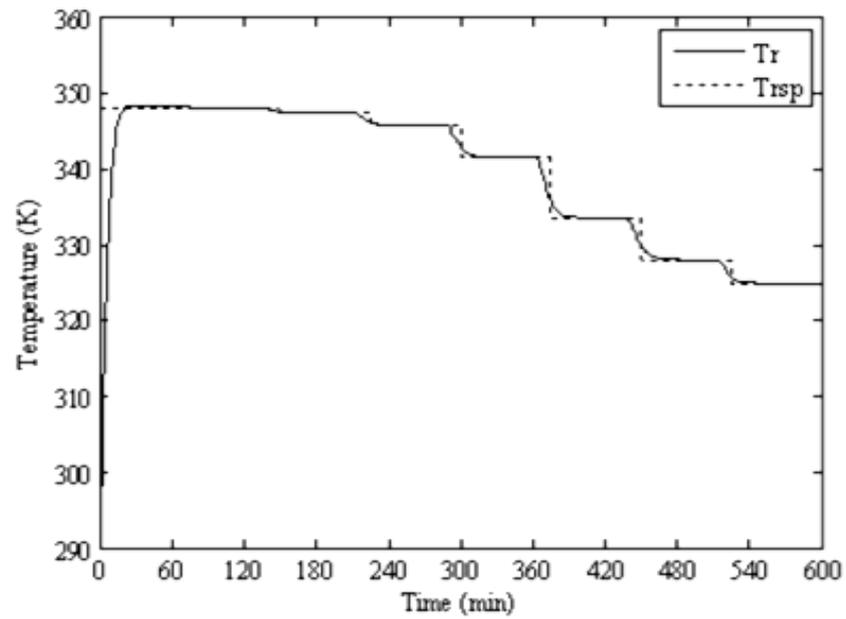


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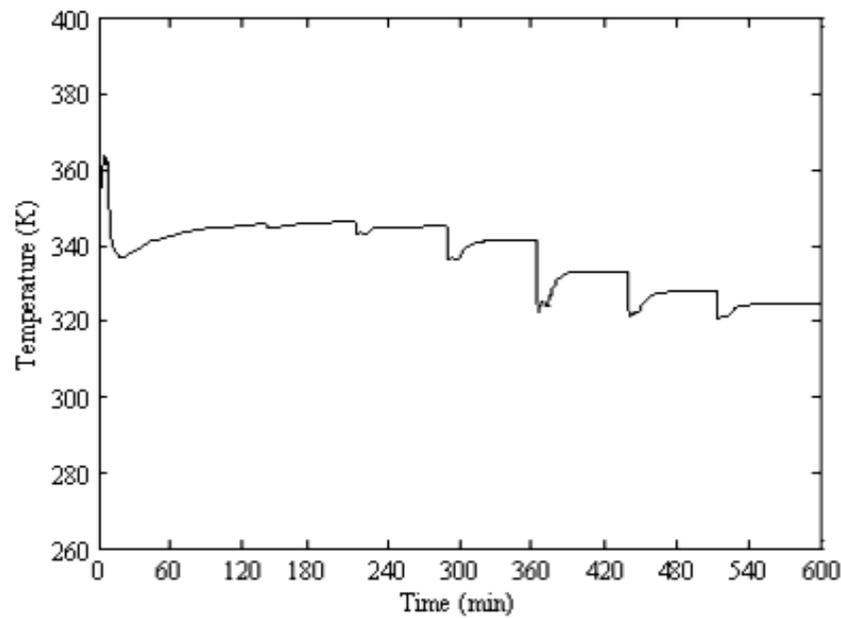
Fig. 9. The temperature control using PID control under the nominal case: (a) the control variable (T_r) and (b) the manipulated variable (T_{isp}).

Table 4. Performance indices of NN MPC strategy, PID control strategy and NNDIC strategy for nominal and model mismatch cases.

Cases	NNMPC	PID	NNDIC
Nominal	422.0569	422.9892	422.4777
+30% k_1	421.0025	433.2399	421.2645
-30% k_2	421.1529	424.3498	422.0130
+30% k_1 , -30% k_2	421.4568	434.5736	421.6259
+30% ΔH	415.2102	433.8296	419.5642
-30%U	435.0598	550.6485	456.2135
+30% k_1 , -30% k_2 , +30% ΔH , -30%U	447.0252	584.9196	462.7851

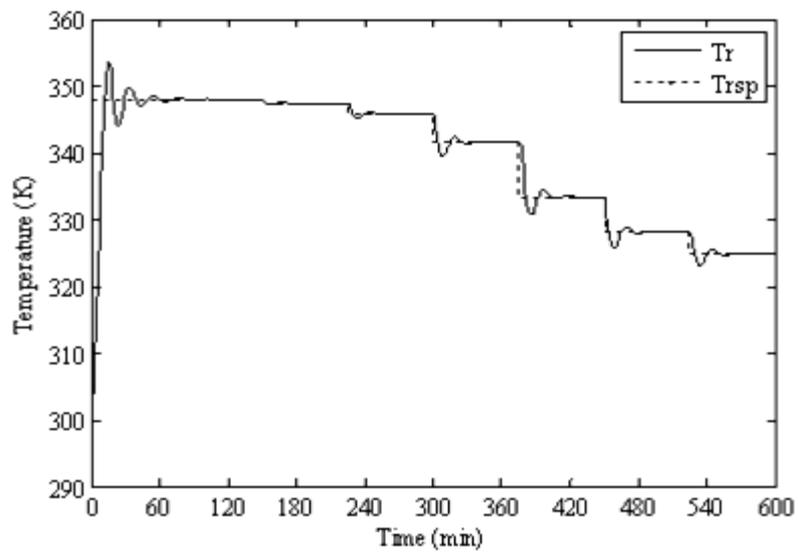


(a)

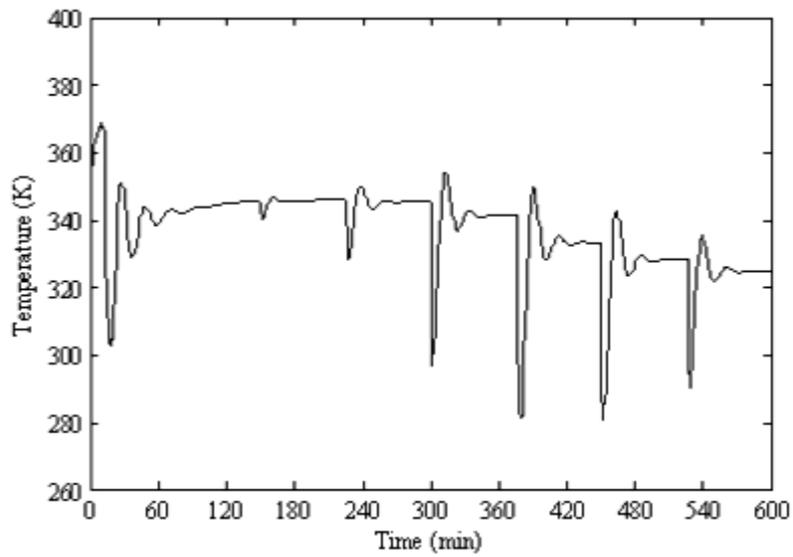


(b)

Fig. 10. The temperature control using NNMPC under the parameter mismatch case (-30% U): (a) the control variable (T_r) and (b) the manipulated variable (T_{jsp}).



(a)



(b)

Fig. 11. The temperature control using PID control under the parameter mismatch case (-30% U): (a) the control variable (T_r) and (b) the manipulated variable (T_{rsp}).

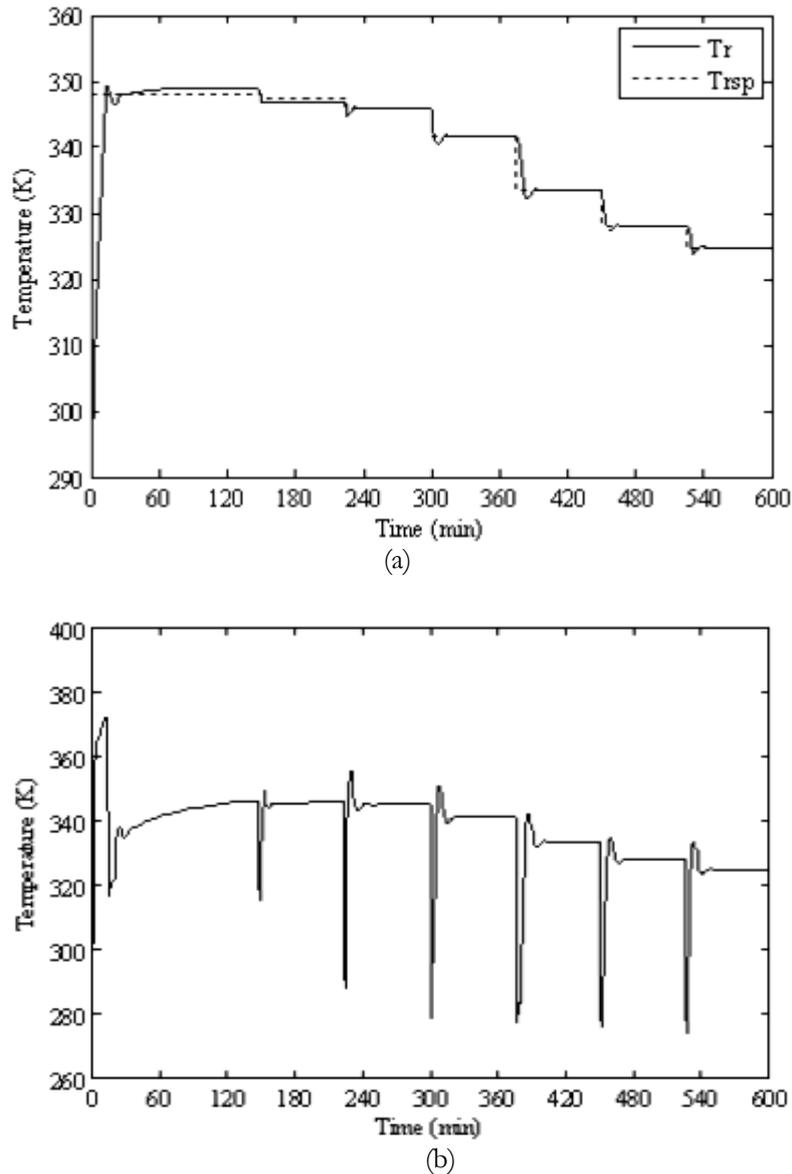


Fig. 12. The temperature control using NNDIC under the parameter mismatch case (-30% U): (a) the control variable (T_r) and (b) the manipulated variable (T_{jsp})

8. Conclusions

In this paper, a dynamic optimization, neural network modeling and neural network based controls are proposed to provide effective control performance for the production of methyl methacrylate (MMA) production in an exothermic batch reactor. To control the temperature of the reactor, neural network based control approaches consisting of a neural network direct inverse control (NNDIC) and a neural network based model predictive control (NNMPC) have been formulated. For the neural network modeling, an optimal structure composes of 8 nodes in an input layer, 6 nodes in first hidden layer, 8 nodes in second hidden layer and 3 nodes in an output layer. An obtained neural network forward model is used to predict a dynamics behavior in the NNMPC algorithm. For a neural network inverse model, an optimal structure consists of 8 nodes in an input layer, 4 nodes in first hidden layer, 8 nodes in second hidden layer and a node in an output layer. In a dynamic optimization, a maximum product achieves at the case of 8 intervals. The obtained temperature profile is applied as set points of the process. Robustness tests of three different controls have been studied with respect to changes in operating parameters. Comparisons of control performances among the PID, the NNDIC and the NNMPC indicated that the NNMPC gives the best control performance in the nominal case and plant/model mismatch cases.

9. Acknowledgment

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10. Nomenclature

A, B, C, D	Correlation constants for each compound
C_i	Concentration of component i (mol/m ³)
C_{pi}	Liquid heat capacity at 293.15 K (J/(mol K))
E_a	Activation energy (J/mol)
k_0	Frequency factor
k_1, k_2	Reaction rate constants (m ^{5.1} /(mol ^{1.7} min))
M	Control horizon
MW_i	Molecular weight of component i (kg/kmol)
P	Prediction horizon
R_i	Reaction rate of component i (mol/(m ³ min))
T_r	Reactor temperature (K)
T_j	Jacket temperature (K)
T_{jsp}	Set point of jacket temperature (K)
V_j	Volume of jacket (m ³)
w_j	Weight of neural network
W_j	Weight for tuning of NNMPC
ΔH	Heat of reaction (J/mol)
ρ_j	Density of component i (kmol/m ³)

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