

**DYNAMIC MODELING AND CONTROLLABILITY ANALYSIS OF DME
PRODUCTION IN AN ISOTHERMAL FIXED BED REACTOR**

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Received 11 April 2014; received in revised form 20 March 2015

Abstract: In this study, to enhance Dimethyl Ether (DME) production from methanol dehydration, an isothermal fixed-bed reactor has been modeled heterogeneously against a commercial adiabatic reactor at dynamic condition. A sensitivity analysis has been carried out to evaluate the influence of input variables on the process conditions using considered dynamic model. The results of sensitivity analysis showed that the feed composition is the main input variable in this system. Also, the process controllability has been investigated using a conventional feedback PID controller. The outlet DME composition from the reactor and circulated boiling water pressure in the steam drum are selected as controlled and manipulated variables, respectively. The responses of the control system to disturbance rejection and setpoint tracking showed that the considered control structure can maintain the process at the desired condition.

Keywords: *Dimethyl Ether; Heterogeneous Model; Methanol Dehydration; PID Controller.*

Available online at <http://www.banglajol.info/index.php/CERB>

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INTRODUCTION

Dimethyl ether as the simplest ether is a colorless gas at the ambient condition that is easily liquefied under light pressure. It does not produce any particulate matter and toxic gases such as NO_x and SO_x at burning [1]. It is useful for a variety of applications such as liquefied petroleum gas substitute, transportation fuel, fuel cell, solvent and propellant in various aerosol products [2].

DME can be produced from a variety of feed-stock such as natural gas, crude oil, coal, waste products and biomass [3]. Recently, biomass is recognized as an extensive feedstock to produce synthesis gas and consequently DME production [4]. Although the commercial technology for DME production is methanol dehydration in conventional reactors, many researchers have focused on the direct conversion of synthesis gas to DME in a single step process [5]. Methanol dehydration can be occurred in a slurry reactor because of the better heat removal capability by coil or jacket and higher catalyst surface area due to small catalyst particle size. However, slurry reactors suffer from significant mass transfer resistance due to use of inert liquid phase [6]. At the present, DME is commercially produced through methanol dehydration using acidic porous catalysts [7]. Due to simplicity and lower capital cost, adiabatic fixed bed reactors are the first choice for catalytic reactions which have low or intermediate reaction heat [8]. However, in case of highly endothermic or exothermic reactions, the catalyst sintering and deactivation may happen [9]. Sintering is a solid state transformation of catalyst, which occurs at high temperatures and is promoted by water vapor molecules. In spite of catalyst loss in fluidized reactors due to collision between catalyst particles and the reactor wall, the fluidized bed reactor has

been suggested as a perfect reactor for DME synthesis [10,11].

There are several articles in the literature that discuss modeling of catalytic packed bed reactors. Shahrokhi et al. modeled a fixed-bed reactor for methanol production from synthesis gas at dynamic state and proposed an optimizer to maximize methanol production rate [12]. Jahanmiri and Eslamloueyan modeled a low pressure methanol reactor and showed that the difference between one and two-dimensional model is negligible [13]. Also, they calculated the optimal shell side temperature considering methanol production as objective function. Lee et al. modeled DME production from synthesis gas in a fixed-bed reactor at steady state condition [14]. Farsi et al. modeled an industrial adiabatic methanol dehydration reactor to DME production at dynamic condition and investigated the controllability of the considered process [15]. Omata et al. studied DME production from synthesis gas in a temperature gradient reactor to overcome the both equilibrium limitations and catalyst activity, experimentally [16]. Then, they optimized the reactor operating condition for higher CO conversion by combining genetic algorithm and neural network. Farsi et al. proposed an optimized isothermal reactor for large scale DME production from crude methanol [17]. They optimized the proposed reactor at steady state condition. The results showed that isothermal reactor is more efficient compared to the traditional adiabatic reactor. Farsi et al. modeled and optimized the isothermal membrane fixed bed reactor for DME production from methanol dehydration [18]. The simulation results of the membrane reactor indicated that the methanol conversion is improved about 6.2% compared to the conventional reactor.

The main objective of the process control is to control the required product rate and concentration at the desired level. Fixed bed reactors are a nonlinear distributed parameter system with dead time that are more difficult to control [19]. Jorgensen presented a good review for dynamic modeling and control of fixed bed reactors [20]. Aguilar et al. developed a robust PID control to control the temperature of a fluid catalytic cracking reactor [21]. This control strategy was robust against model uncertainties and noisy measurements.

Mainly, the objective of this paper is dynamic modeling, simulation and control of an isothermal DME synthesis reactor that has been coupled with a steam drum. In this study, a dynamic one-dimensional heterogeneous model is developed to simulate the isothermal reactor based on the mass and energy conservation laws. Also, to verify the accuracy of the proposed model and considered assumptions, results of the steady state model of adiabatic reactor are compared with the design data (Petrochemical Zagros Complex in Iran). Then, a conventional feedback control system is designed to maintain the outlet DME concentration at the desired value.

MODELING

The main operations in the DME process are the feed preparation, DME synthesis convertor, product separation and methanol recycling. Figure 1 shows the process flow sheet of DME synthesis in an isothermal fixed bed reactor. The isothermal fixed bed reactor is a vertical shell and tube heat exchanger, which tubes are packed with catalyst pellets and surrounded by the boiling water. The heat of reaction is removed from the reactor by circulating the boiling water as coolant in the shell side of the reactor. The outlet coolant from the

reactor is passed through a steam generator, and then recycled to the shell side of the reactor.

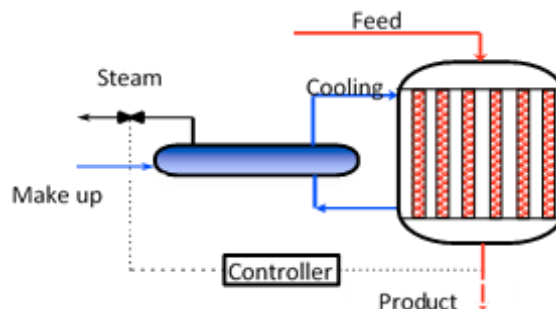
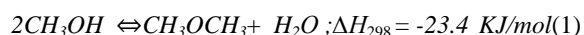


Figure 1: Process Flow Diagram of DME Production

Reaction Kinetics

The reaction of DME synthesis is mainly dehydration of methanol that is an exothermic and equilibrium reaction. Many researches have focused on DME synthesis through methanol dehydration and synthesis gas conversion [22-24]. The reaction equation of DME synthesis from dehydration of methanol is as the following.



In this work, the rate expressions have been selected from the kinetic model proposed by Berčić et al. [25].

$$r = \frac{k \cdot K_{CH_3OH}^2 \left(C_{CH_3OH} - \frac{C_{CH_3O} C_{H_2O}}{K_{eq}} \right)}{(1 + 2(K_{CH_3OH} C_{CH_3OH})^2 + K_{H_2O} C_{H_2O})^2} \quad (2)$$

where,

$$\ln(K_{eq}) = 0.86 \log T + \frac{3138}{T} + 1.33 \times 10^{-3} T - 1.23 \times 10^{-5} T^2 + 3.5 \times 10^{-10} T \quad (3)$$

K_{CH_3OH} and K_{H_2O} are the reaction rate constant and the adsorption equilibrium k_1 , constants for methanol and water vapor, respectively, which has been tabulated in Table 1. Commercially, $\gamma\text{-Al}_2\text{O}_3$ catalyst is used in methanol dehydration reaction.

Table 1: Kinetic and Equilibrium Constants

Parameter	A _i	B _i
K ₀	3.7 e10	-105000
K _{H2O}	0.84 e ⁻¹	41100
K _{CH3OH}	7.9 e ⁻⁴	7050

Process Modeling

Reactor Modeling

In this study, a one-dimensional heterogeneous model has been considered to simulate DME reactor and steam drum at steady state and dynamic condition. The basic structure of the model is composed of heat and mass conservation equations coupled with thermodynamic and kinetic relations as well as auxiliary correlations to predict physical properties. In the process modeling the following assumptions have been considered.

- Plug flow pattern and negligible concentration and temperature gradient in radial direction.
- The bed void fraction is constant along the reactor.
- Uniform temperature in the catalyst pellet.
- Bed porosity in axial and radial directions is constant.
- The gas mixture is an ideal gas.
- Heat losses via surrounding is neglected

Thus, mass and energy balances for the gas phase are expressed by:

$$\varepsilon \frac{\partial C_i}{\partial t} = -u_s \frac{\partial C_i}{\partial z} - k_{gi} a_v (C_i - C_{is}^s) \quad (5)$$

$$\rho_g c_{pg} \varepsilon \frac{\partial T}{\partial t} = -u_s \rho_g c_p \frac{\partial T}{\partial z} + h_f a_v (T_s^s - T) - \frac{(\pi D) U}{A_c} (T - T_j) \quad (6)$$

The mass and energy balances for the solid phase are expressed by:

$$(1 - \varepsilon) \frac{\partial C_i}{\partial t} = -k_{gi} a_v (C_i - C_{is}^s) + \eta_i \rho_B r_i \quad (7)$$

$$\rho_B c_{ps} (1 - \varepsilon) \frac{\partial T_s}{\partial t} = -h_f a_v (T_s^s - T) - \eta_i \rho_B (-\Delta H) r_i \quad (8)$$

The pressure drop in the catalytic packed bed is calculated by Ergun equation [26]:

$$-\frac{\Delta P}{L} = 150 \frac{(1 - \varepsilon)^2 \mu u_c}{d^2 \varepsilon^3} + 1.75 \frac{(1 - \varepsilon) \rho u_c^2}{d \varepsilon^3} \quad (9)$$

The feed condition, characteristics of the catalyst pellets and the reactor design specifications of DME reactor is reported in Table 2 [20].

Table 2: Feed Specification and Reactor Data

Feed Composition (Mole Fraction)	
CH ₃ OH	0.94
DME	0.05
H ₂ O	0.01
Total molar flow rate (kmol hr ⁻¹)	5600
Inlet temperature (K)	533
Inlet pressure (bar)	18.18
Catalyst particle	
Particle diameter (m)	0.3175 × 10 ⁻²
Specific surface area (m ² m ⁻³)	673
Adiabatic reactor	
Reactor length (m)	8
Reactor diameter (m)	4
Isothermal reactor	
Reactor length (m)	8
Tube diameter (m)	0.09
Tube number	2000

Auxiliary equations are used to predict the model parameters. In the heterogeneous models physical properties of components and overall mass and heat transfer coefficients between catalyst solid phase and gas phase should be considered. In the mass

and energy balance equations, η_i is the effectiveness factor which is defined as the actual reaction rate per particle to theoretical reaction rate based on the external pellet surface concentration. The effectiveness factor is calculated from the dusty gas model [27]. The detail of dusty gas model is given in Appendix A. The methods used for calculating the physical properties, overall heat and mass transfer coefficients between solid and gas phase and mass diffusion needed for simulation are given in Table 3.

Table 3. Methods Used for Calculating the Physical Properties, Heat and Mass Transfer Coefficients

Parameter	Reference
Mass transfer coefficient	Cussler [28]
Binary diffusion coefficient	Reid et al. [29]
Gas conductivity	Lindsay and Bromley [30]
Effective diffusion coefficient in pellet	[31]
Mixed gas heat capacity	Ideal gas [32]
Gas viscosity	Lucas [32]
Gas–solid heat transfer coefficient	[33]

Steam Drum Modeling

The steam drum is one of the most important components in a water cooled isothermal reactor. To develop a steam drum model, the following assumptions are made:

- Heat losses via surrounding is neglected
- In energy balance, pseudo-steady state condition is considered for vapor phase

The dynamic of this process can be described by the following set of mass and energy balance equations for liquid and vapor phases [28]. The schematic diagram

of the steam drum is shown in Figure 2.

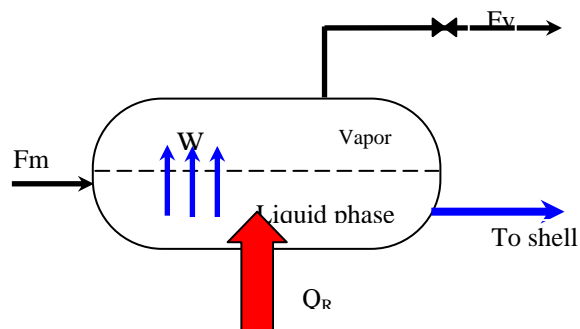


Figure 2: Schematic of the Control Structure for Isothermal Reactor

Subject to these assumptions, the gas and liquid mass balances are expressed by:

$$\frac{d(\rho_l V_l)}{dt} = \rho_m V_m - w \quad (10)$$

$$\frac{d(\rho_v V_v)}{dt} = w - \rho_v V_v \quad (11)$$

The liquid energy balances is expressed by:

$$C_p \frac{d(\rho_l V_l T)}{dt} = (\rho_m F_m) h_m - w h_s + Q_R \quad (12)$$

Numerical Solution

To solve the set of nonlinear partial differential equations (PDEs) obtained from dynamic modeling, the reactor length is divided into equal discrete intervals. Then, using finite difference method the PDEs are converted into a set of ordinary differential equations (ODEs) in the time domain. The set of reactor and steam drum equations are solved by 4th order Runge–Kutta [35]. Before carrying out dynamic simulation, the initial condition of the system should be obtained using steady state equations. The objective of the steady state simulation of the DME reactor is to determine the concentration and temperature profiles along the reactor in normal operation. After

rearranging the modeling equations for the steady state condition, a set of ordinary differential equation is obtained. This set of equations was solved by the method of 4th order Runge–Kutta.

Control Structure

The main objective of control systems in industrial processes is to maintain the operating condition at the optimum state in spite of disturbances and uncertainties. The considered control structure in the DME process is shown in Figure 2. In this study, a conventional feedback PID controller has been used to investigate the load rejection and setpoint tracking tasks. In this configuration, the outlet DME mole fraction is controlled considering the circulating boiling water pressure as the manipulated variable. The heated coolant is passed through a steam generator to produce steam and recycled to the shell side of the reactor.

RESULTS AND DISCUSSION

Steady State Simulation

In this section, the steady state simulation results of the isothermal DME reactor have been presented. The model of methanol dehydration is validated against design data of a conventional adiabatic DME synthesis reactor. Table 4 shows the absolute errors between the simulation results and available plant data. The simulation results show that the proposed model is capable to predict concentration and temperature profiles along the adiabatic reactor with a high accuracy.

The validation of the isothermal reactor is impossible due to lack of data for the isothermal reactor. In this section, the steady state results of simulation, such as temperature and concentration profiles, in the isothermal reactor are presented. Figure

3 illustrates the molar flow rate of DME, methanol and water vapor along the isothermal reactor at steady state condition.

Table 4: Comparison of the Steady State Simulation Results of the Adiabatic Reactor with Plant Data

	Simulation Result	Plant Data	A.R.E
Outlet DME (kmol/hr)	2457	2506	1.95%
Outlet MeOH (kmol/hr)	940.6	937.7	0.31%
Exit temp (°C)	652.2	644	1.27%

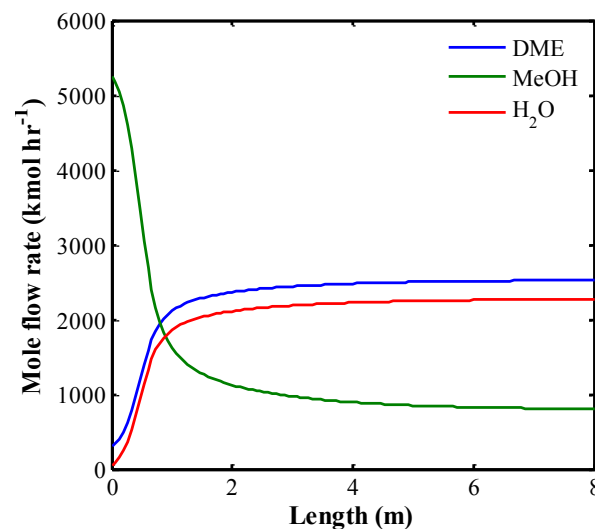


Figure 3: The Profiles of Mole Flow Rates of DME, Methanol and Water along the Reactor

DME mole fraction along the reactor in the solid and gas phases predicted by the heterogeneous model has been compared in Figure 4. In the second half of the reactor, where the reaction approaches to the equilibrium, the difference between gas and solid phase concentrations is negligible.

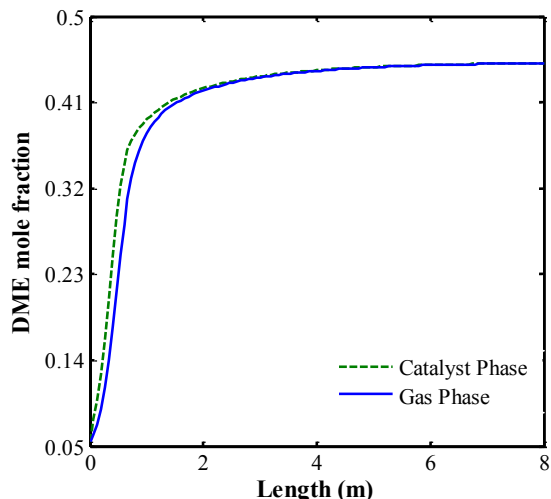


Figure 4: DME Mole Fraction in Solid and Gas phase

The predicted temperature profile along the reactor is shown in Figure 5. According to Le Châtelier's principle, temperature reduction in the second half of the isothermal reactor leads to shift the methanol dehydration reaction in the exothermic direction, which results the higher DME production. At the reactor entrance, temperature increases rapidly and results increasing kinetic constant of reaction rate. In the isothermal reactor, after a certain position along the reactor the temperature maintains constant due to heat transfer between exothermic reaction and boiling water through the tube wall.

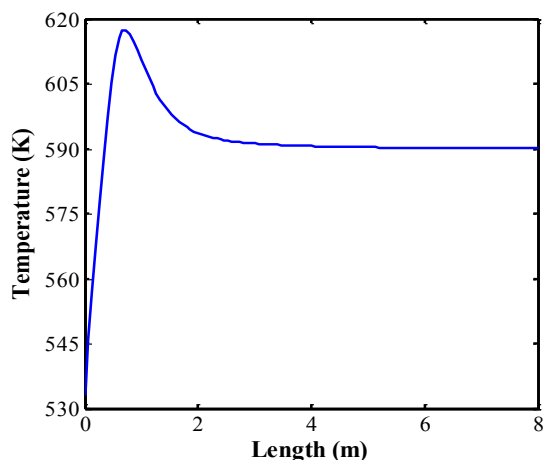


Figure 5: Predicted Temperature Profile along the Reactor

Open Loop Dynamic Simulation

Dynamic simulation is mainly used to investigate the process stability, safety and controllability. To investigate the influence of input variables on the dynamic behavior of DME reactor, feed temperature, feed pressure and composition have been considered as the main loads in the system. In this section, the effect of $\pm 10^{\circ}\text{C}$ step change in the feed temperature on the outlet DME mole flow rate is presented in Figure 7. This figure shows that the outlet DME mole flow rate from the reactor reaches to a new steady state condition after 40 seconds. Since the optimal temperature of shell side is inserted in the simulation (590K), DME production decreases with variation of feed temperature in this configuration. This process exhibits inverse response behavior due to integration of kinetic constant and equilibrium limitations. Due to these disturbances in the feed temperature, the outlet DME mole flow rate has changed less than 10 kmol hr^{-1} .

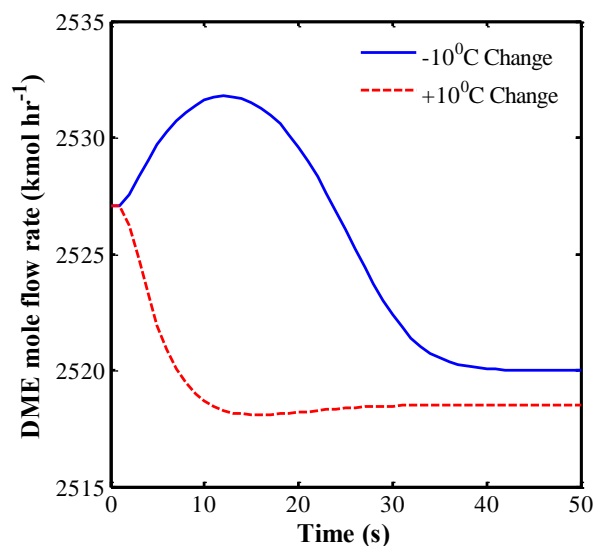


Figure 6: The Outlet DME Mole Flow Rate Profile from the Reactor to $\pm 10^{\circ}\text{C}$ Step Change in the Shell Side Temperature

In the literature, the expression of “inverse response” is used for processes that have an initial response direction opposite to

the final direction. These processes are difficult to be identified and controlled due to the presence of right half plane zeros. According to Le Châtelier's principle, changing an independent variable of a system at equilibrium such as temperature, pressure and concentration, shifts the equilibrium toward that tends to reduce the effect of the change. Thus, when the inlet pressure of the DME synthesis reactor is changed, the equilibrium remains constant due to equimolar stoichiometry in the methanol dehydration reaction. Consequently, the effect of feed pressure variation on DME production isn't presented. The feed composition effect is studied considering -0.05 step changes in the methanol mole fractions and +0.05 step changes in the water vapor mole fraction. This disturbance in the mole fraction is feasible and reasonable in industrial scale. Figure 7 shows the outlet DME mole flow rate profile with the passage of time. According to Le Châtelier's principle, decreasing the feed composition shifts thermodynamic equilibrium to neutralize this effect by decreasing methanol conversion. Due to these variations in the feed composition, the outlet DME mole flow rate decreases about 60 kmol hr^{-1} . This figure shows that the process has a time delay about 20 second. The most of industrial processes, particularly distributed systems such as tubular reactors, exhibit time delays or dead times. Generally, dead time reduces the controller system ability and performance.

The results in Figure 7 indicate that the feed composition is the major disturbance that should be rejected by the designed control system. In this process, maintaining DME production rate at the desired level is possible using variation of circulating saturated water pressure in the shell side as the manipulated variable inserting a

conventional control valve.

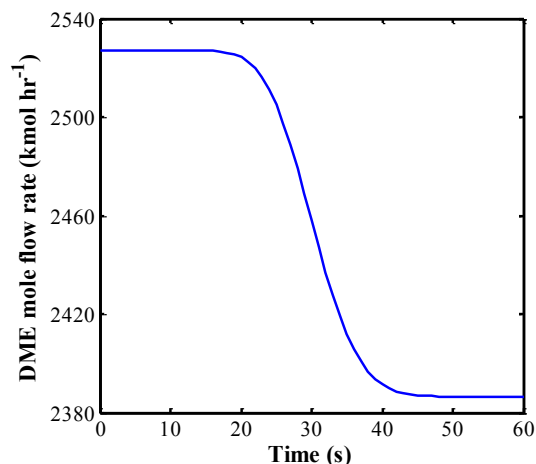


Figure 7: The outlet DME Mole Flow Rate Profile from the Reactor to ± 0.05 Step Change in Feed Mole Fractions

Closed Loop Process Simulation

In order to control the outlet DME mole flow rate at the desired level a feedback control loop is designed based on the conventional PID (Proportional–Integral–Derivative) controller. The PID feedback controller is widely used in industries, because it requires minimal process knowledge to design and controller tuning is simple [36]. A properly tuned PID controller can be robust against disturbances and uncertainty. In this study, the PID controller has been tuned using Closed Loop Ziegler–Nichols method. This algorithm is one of the most common methods for tuning PID controllers. The parameters of the controller, i.e. the proportional gain, reset time and derivative value are derived from the ultimate gain and the sustained period of oscillation [37]. According to this procedure, the controller gain, reset time and derivative constant has been calculated about 12, 0.45 min and 0.08 min, respectively. In this section, the dynamic closed-loop behavior of the process to predefined disturbances and set point variation are presented and analyzed.

Load Rejection

The considered control system should be capable to reject process disturbances using available manipulated variable without saturation of the control valve. To investigate the performance of the proposed control structure in disturbance rejection, -0.02 and $+0.02$ step change in the inlet methanol and DME mole fraction has been introduced as the main disturbance. Figure 8 shows the closed loop response of the process due to this disturbance. As can be seen from this figure, the considered load is completely rejected after about 5 minutes using the tuned conventional PID controller and process output approached to the desired setpoint. Also, although the overshoot of the controller response is almost high, the decay ratio, which states how oscillations decay in a system, is negligible. Thus, the response of the considered PID controller is acceptable in the load rejection.

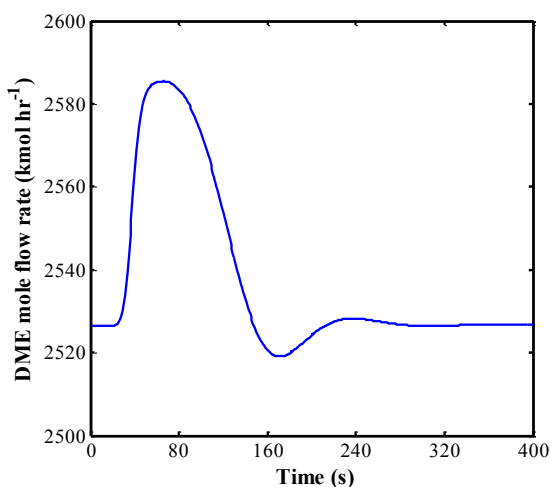


Figure 8: Closed Loop Response for ± 0.02 Step Change in the Feed Mole Fraction

Setpoint Tracking

In case of servo tracking, the considered control loop should be capable to track the process setpoint using available manipulated variable without saturation of the control valve. The ability of the proposed control

system to track setpoint has been investigated considering -0.02 step changes in the process setpoint. Figure 9 shows the efficiency of considered control loop to track the desired outlet DME mole flow rate.

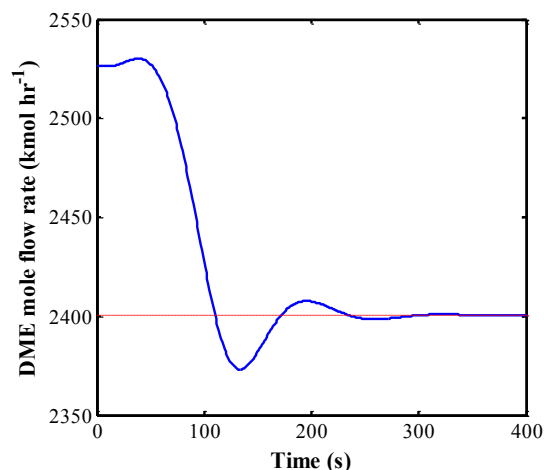


Figure 9: Closed Loop Response for -0.02 Step Change in Setpoint

Decreasing shell side temperature reduces outlet methanol mole flow rate and process approaches to the new setpoint. In this configuration, the outlet DME mole flow rate approaches to the new setpoint after 5 min. Decreasing shell side temperature is occurred using shell side pressure reduction with common control valves. Table 5 presents the dynamic characteristics of the control loop response for the set point tracking. In this system, the overshoot and particularly decay ratio are almost small which result low oscillation of the control system. This table shows that characteristics of considered PID controller are acceptable in the servo tracking.

Table 5: Dynamic Properties of the Closed Loop Response for Setpoint Tracking

Overshoot	% 26
Decay ratio	% 12
Settling time (sec)	285

CONCLUSION

In this study, methanol dehydration in an isothermal DME synthesis reactor was modeled and simulated, dynamically. A one-dimensional heterogeneous model was considered based on the mass and energy conservation laws. The results of steady state simulation of the adiabatic model were compared to the industrial plant data. This comparison showed a good agreement between the simulation results and plant data. Open loop responses of the process against important disturbances were investigated to discover the dynamic properties of the process. These results showed that the process has reverse response due to integration of reaction, heat transfer and equilibrium limitations. To maintain the methanol production rate at the desired setpoint and process safety, a tuned PID controller was used considering cooling water pressure as manipulated variable. The performance of considered feedback closed loop system was investigated for disturbance rejection and setpoint tracking. The dynamic properties of the control system indicated that this configuration has good performance in the load rejection and servo problem.

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