Steady State Operability Characteristics of an Adiabatic Fixed-Bed Reactor for Methanol Dehydration

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ABSTRACT: *Operability analysis as one of the most important bridges between process design and process control helps the process designer to investigate the control issues quite early in the design stage. Recently, Vinson and Georgakis suggested a steady state geometrical operability index as a quantitative measure for assessment of process operability. In this paper, DME fixed-bed reactor is heterogeneously modeled based on mass and energy conservation law at steady state condition. To verify the accuracy of the model, simulation results of the conventional reactor is compared with the available industrial plant data. It is observed that there is a good agreement between the simulation result and the plant data. Then, the steady state operability characteristics of DME reactor have been analyzed by using the framework of Vinson and Georgakis. This paper demonstrates the utility of the approach to industrial chemical reactors. By measuring the operability characteristics of the reactor, the input and output operability index of the process is calculated equal to 43.31% and 57.58% respectively. The results showed the low process ability for creation of desired output by available input.*

KEY WORDS: *Operability analysis, Modeling, Fixed-bed reactor, Operability index.*

INTRODUCTION

Making decision in the stage of process design has a significant effect on the operability characteristics of processes. The fundamental concept behind the interaction between design and process is consideration of operability at the design stage in order to create a process with proper dynamic and steady state characteristics. Operability analysis refers to intrinsic property of a process to obtain acceptable control and operation performance in spite of bounded inputs and model uncertainty using the available manipulated variable [1]. Ignoring the operability characteristics of a plant can lead to serious operational difficulties during operation [2]. In order to investigate operability of a process, a tool is required that measures operability quantitatively. These measures produce valuable tools in order to separate or classify alternative designs. Currently, a large number of operability measures have been proposed and developed to assist the operability properties of processes. The most of these operability indicators can be easily computed, especially when they only refer to the steady state information of the process [3]. Consideration of interaction between process design and control was documented almost six decades ago. *Ziegler & Nichols* delineated limitations of control on a poorly designed process [4]. Farr and Aris investigated the operability and stability analysis of reactors

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1021-9986/11/4/45 6/$/2.60

by comparison of heat generation and heat withdrawal curves [5]. *Russo & Bequette* demonstrated a different method to investigation of the operability of CSTRs with exothermic reactions [6]. *Lyman et al.* adopted an approach based on case studies to investigate the interaction between process design and control [7]. *Georgakis et al.* proposed a geometric approach for process operability analysis [8]. The proposed approach is applicable for both linear and nonlinear processes and requires small computation. Recently, *Vinson & Georgakis* presented a direct geometrical measure for the steady state operability analysis of the process [9, 10]. This approach is easy to understand and apply. *Uzturk & Georgakis* showed that this geometrical measure is extendable to dynamic situations [11, 12].

In this paper, the operability of dimethyl ether fixed-bead reactor is studied. Currently, DME is commercially produced by methanol dehydration in an adiabatic fixed-bed reactor using acidic porous catalysts [13]. This paper is structured as follows: First, DME production process is illustrated and DME adiabatic fixed-bed reactor is modeled at steady state condition. Then, the operability index proposed by *Vinson & Georgakis* is reviewed. Finally, the steady state operability indexes are calculated for DME reactor and results of simulation and operability analysis is presented and concluded the paper.

PROCESS DESCRIPTION

The reaction of DME synthesis is mainly dehydration of methanol on γ-Al_2O_3 catalyst. Preparation of DME from methanol can be represented by:

$$2CH_3OH \Leftrightarrow CH_3OCH_3 + H_2O$$
$$\Delta H = -23.4\ {}^{kJ}\!/_{mol} \qquad (1)$$

DME reactor is an adiabatic fixed-bed reactor which tubes is homogenously packed by catalyst slice. Schematic diagram of a conventional adiabatic methanol dehydration reactor is shown in Fig. 1. In a heat exchanger, the heat of product stream is used to preheat the feed stream.

The feed specifications and catalyst characteristics of methanol dehydration reactor are presented in Table 1.

Process modeling

In this study, DME reactor is heterogeneously modeled with consideration of heat and mass transfer between catalyst and reactant gas at steady state condition. The rate expression which is used in the mathematical model has been selected from Bercic et al. [14]. The assumptions in the one-dimensional modeling are listed at below:

- The system is well insulated.
- Since the reactor is adiabatic with low heat of reaction, radial mass and energy transfer are neglected.
- Lumped body is assumed for catalyst slice (Biot= 0.0125).
- The gas mixture is assumed an ideal gas.

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

$$u_s \frac{\partial C_i}{\partial z} = -k_{gi} a_v \left(C_i - C_{is}^s\right) \qquad (2)$$

$$k_{gi} a_v \left(C_i - C_{is}^s\right) = a\eta_i \rho_{Bs} r_i \qquad (3)$$

Table 1: The feed specifications of an industrial adiabatic fixed bed reactor.

Parameters	Value
Feed composition (mole fraction)	
CH_3OH	0.94
DME	0.05
H_2O	0.01
Total molar flow rate (kmol hr^{-1})	5600
Inlet temperature (K)	533
Inlet pressure (bar)	18.18
Rector	
Reactor diameter (m)	4
Reactor length (m)	8
Catalyst particle	
Particle diameter (m)	0.318×10^{-2}

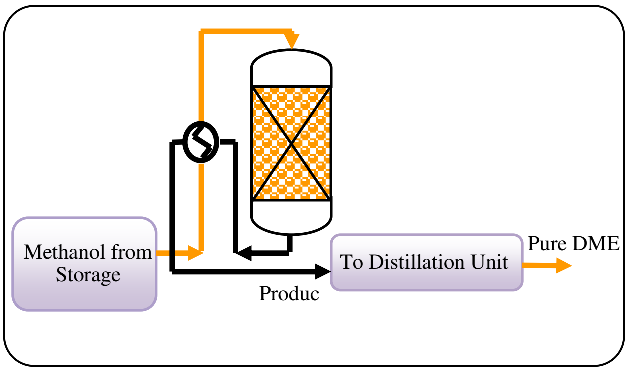

Fig. 1: Schematic diagram of a traditional adiabatic DME reactor.

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

$$u_s \rho_g c_p \frac{\partial T}{\partial z} = h_f a_v \left(T_s^s - T\right) \quad (4)$$

$$h_f a_v \left(T_s^s - T\right) = -a\eta_i \rho_B \left(-\Delta H\right) r_i \quad (5)$$

The pressure drop through the catalytic bed is calculated based on the Ergun equation. The related equation for tubular reactors is [15]:

$$-\frac{dP}{dz} = \frac{150\,\mu}{\varphi_s^2\, d_p^2} \frac{\left(1-\varepsilon\right)^2}{\varepsilon^3} \frac{Q}{A_c} + \frac{1.75\,\rho}{\varphi_s\, d_p} \frac{\left(1-\varepsilon\right)}{\varepsilon^3} \frac{Q^2}{A_c^2} \quad (6)$$

To complete the simulation, auxiliary correlations should be added to the model. In the heterogeneous model, due to transfer phenomena, proper correlations for estimation of heat and mass transfer between two phases, physical properties of chemical species should be considered. The correlations which are used for prediction of mass an heat transfer coefficient between gas and solid phases are as follows [16, 17]:

$$k_{gi} = 1.17\, Re^{-0.42}\, Sc_i^{-0.67} u_g \quad (7)$$

$$\frac{h}{C_p \rho \mu}\left(\frac{C_p \mu}{K}\right)^{2/3} = \frac{0.458}{\varepsilon_B}\left(\frac{\rho u d_p}{\mu}\right)^{-0.407} \quad (8)$$

OPERABILITY INDEX

To operability analysis of the DME reactor, the operability approach introduced by *Vinson & Georgakis* is considered. The process inputs that are changeable over a certain range are called Available Input Space (AIS). The model can be solved in the range of AIS to obtain outputs that are referred as the Achievable Output Space (AOS). Also, Desired Output Space (DOS) is specified as the desired operating window for the process outputs. The set of input values required to obtain DOS can be calculated from the model inverse. The collection of all obtained input values is denoted as desired input space (DIS). Operability index is defined in the output space as:

$$OI_y = \frac{\mu\left[AOS \cap DOS\right]}{\mu\left[DOS\right]} \quad (9)$$

where, μ is a measure function to calculate the size of the corresponding space. For example, in two dimensions, it represents the area. If it is less than unity, it would imply that our expectation of the design is higher than it can possibly deliver. This index would be useful in analyzing the operability of existing plant designs as it indicates how much of the desired process output region is achievable with the available inputs. Operability index is defined in the input space as:

$$OI_u = \frac{\mu\left[AIS \cap DIS\right]}{\mu\left[DIS\right]} \quad (10)$$

The AIS and DOS are user-specified, whereas AOS and DIS are derived through the model and model inverse respectively. Specifying the AIS for a particular system is fairly straightforward as this would mean identifying the ranges of different manipulated variables in a given plant. In the case of DOS, it needs to represent the correct operating window which is envisioned for the plant.

SELECTION OF VARIABLES

The selection of the input and output variables is important. The output variables should be chosen, so that they represent the state of the system well. The input variables should have rapid dynamic and significant effect on the output variables. In adiabatic and exothermic reactors, temperature is an important parameter that has a direct and severe effect on reaction rate, thermodynamic equilibrium and catalyst activity. Thus, the best choice for the input variables is the inlet feed temperature and pressure. In this reactor, outlet temperature and DME concentration are selected as output variables due to process safety, productivity and outlet product purity.

RESULTS AND DISCUSSION

In this section, the steady state simulation results of DME reactor are presented. The set of model equations is solved with 4th order Runge-Kutta method. First, the accuracy of the model is proved and then the process operability is considered. The model accuracy is evaluated through comparing the simulations results of the adiabatic fixed-bed reactor with the operating data from Zagros Petroleum Complex. As can be found in Table 2, a good agreement is observed between the simulation result and the plant data.

Table 2: Comparison between simulation and plant data.

	Simulation result	Plant data	Absolute relative error
DME mole flow (kmol/hr)	2457	2506	1.95%
MeOH mole flow (kmol/hr)	940.6	937.7	0.31%
Temperature (K)	652.2	644	1.27%

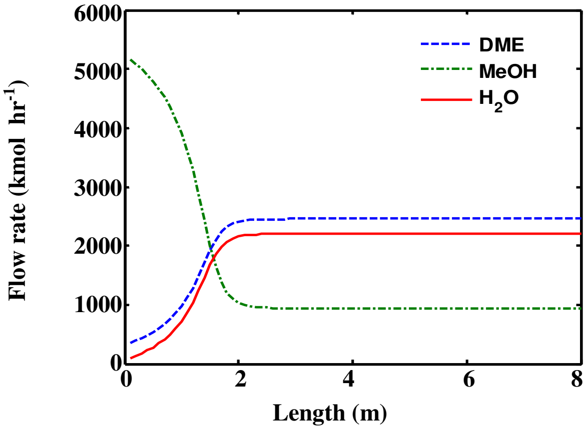

Fig. 2: Mole flow rate of DME, methanol and water vapor along the reactor.

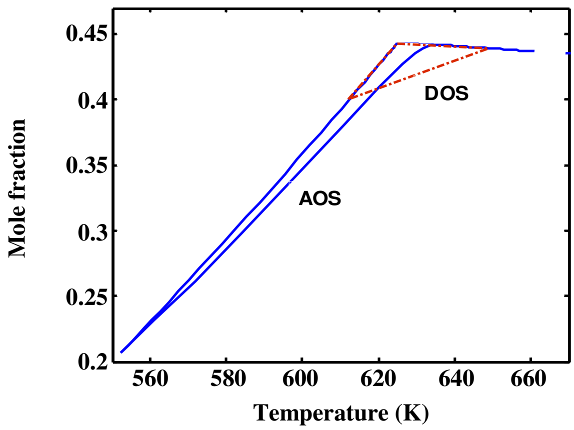

Fig. 3: Available and desired output state (AOS and DOS).

The mole flow rate of DME, methanol and water vapor along the reactor are shown in Fig. 2. This figure shows that the reaction reaches to the thermodynamic equilibrium and conversion remains constant.

This research mainly focuses on analyzing the importance of the interaction between design and operability issues. Operability is the ability of the plant to achieve acceptable operation at steady and dynamic state. Operability includes flexibility, switchability and controllability as well as many other issues. The goal of the process operability analysis is to ensure that there is an adequate equipment overdesign so that the process constraints can be satisfied while a combination of the operation costs and overdesign cost is minimized. In this section, the input-output steady state operability characteristics of DME adiabatic fixed-bed reactor are studied as a nonlinear and complicated model. The feed temperature and pressure are considered as input variables and changed in range of 16.18-20.18 bar and 500-550 K, respectively. The AOS is calculated using steady state mathematical model in range of AIS. The low temperature-high conversion region is chosen as the DOS due to catalyst deactivation, process safety, productivity and product purification cost. The selected DOS and AOS are shown in Fig. 3. This figure shows that outlet DME mole fraction changes from 0.21 to 0.44 due to variation of feed temperature and pressure. Also, outlet temperature changes from 572K to 660K. The upper bound of DME mole fraction and lower bound for outlet temperature in DOS is limited by equilibrium. After the intersection calculations, the output operability index of the process is estimated about 43.31%. In an ideal process, the operability value is close to unity. The computed output operability for DME reactor is far from an ideal process.

Thus, AIS cannot produce the desired output space in this structure. DIS, that is required to operate the process at DOS, can be calculated from inverse of mathematical model of the reactor. The calculated DIS and AIS are shown in Fig. 4. By comparison DIS and AIS, the input operability index is estimated to be 57.58%. Fig. 4 shows that the AIS has a large enough range in feed temperature. However, the feed pressure is clearly limiting of process operability.

These results prove that considered design for methanol dehydration process in process design stage is not sufficient and capable to achieve desired process output ranges using the available input ranges in presence of expected disturbances. Also, the input operability index is different from one in the output space.

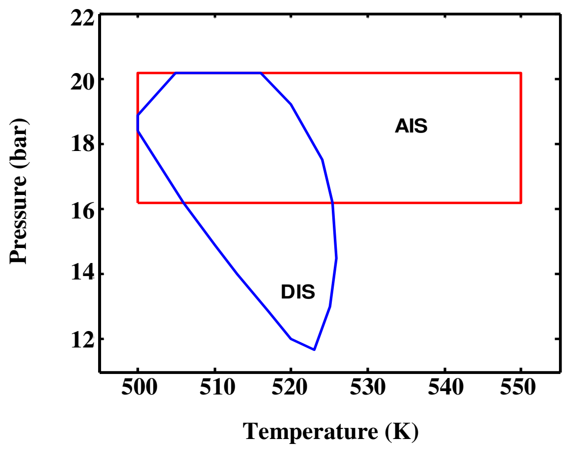

Fig. 4: Desired and available input state (DIS and AIS).

This is due to the nonlinear relationship between the input and output variables. The nonlinearity of the process has significant effects on the steady state operability of the DME reactor. The considered inputs in the real process cannot create proper and desired outlet conversion and temperature (*DOS*). By decreasing the operating pressure in real process, higher DME conversion can be attainable. The input and output operability indexes show the low operability of adiabatic fixed-bed reactor for DME production.

CONCLUSIONS

In this paper, interaction between process design and process operability of DME fixed-bed reactor was analyzed at steady state condition. This analysis can be used in the early design stages where the information of the process is very limited. First, the DME fixed-bed reactor was heterogeneously modeled based on mass energy conservation laws. The output temperature and DME concentration as AOS were calculated using specified input variables and considered model. Also, desired feed temperature and pressure as DIS were calculated to operate the process in the desired output region. Then, the DIS and DOS were compared with the AIS and AOS to determine the input and output operability index, respectively. The results showed the low input and output operability index of process and consequently low process ability to creation of desired output by available inputs. The considered design for methanol dehydration process in process design stage is poor and incapable in handling of disturbances and desired process output ranges.

Nomenclature

A_c	Cross section area of each tube, m^2
C_i	Molar concentration of component I, mol m^{-3}
C_p	Specific heat of the gas at constant pressure, J mol^{-1}
d_P	Particle diameter, m
F	Total molar flow rate, mol s^{-1}
h_f	Gas-solid heat transfer coefficient in reactor, W m^{-2} K^{-1}
P	Total pressure, Bar
Q	Volume flow rate, m^3 s^{-1}
r	Rate of reaction for DME synthesis, mol kg^{-1} s^{-1}
Re	Reynolds number
Sc_i	Schmidt number of component i
T	Temperature, K
u	Velocity, m s^{-1}
z	Axial reactor coordinate, m

Greek letters

μ	Viscosity of fluid phase, kg m^{-1} s^{-1}
ρ	Density of fluid phase, kg m^{-3}
ε	Void fraction

Superscripts

g	In bulk gas phase
s	At surface catalyst
i	Chemical species

Received : Feb. 22, 2010 ; Accepted : Feb. 28, 2011

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