

REAL TIME OPTIMIZATION OF SHELL AND TUBE METHANOL REACTOR USING EVOLUTIONARY AND GENETIC ALGORITHMS

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Abstract

Real Time Optimization (RTO) of industrial shell and tube methanol reactor is presented. Mathematical modeling of the reactor is considered as the case study. Inlet concentrations of CO, CO₂, H₂, H₂O and methanol are used as disturbances. Methanol output is maximized by adjusting the temperature of the shell as manipulating variable. Differential evolution (DE) and genetic algorithm are utilized for optimization purpose; in which the optimizer initiates every hour and determines the optimal shell temperature. Catalyst activity reduction is updated as model parameter every 24-hour periods. Comparing the optimization results demonstrates that DE algorithm requires less CPU time than genetic algorithm. Consequently, using the optimization algorithm and control loop, methanol production increases up to 15%.

Key words: real time optimization (RTO); differential evolution; methanol reactor.

1. Introduction

Online optimization of chemical processes has attracted considerable attention in the past few decades, and it has become increasingly important in achieving a competitive advantage in industries [1,2]. In advanced control system, process model is used to predict process responses to set point or manipulated variable changes. Real time optimization is a technique of using process model combined with economic information to determine the optimum operation policy. There are two types of optimization methods: direct search and model based algorithms. Direct search method is utilized when obtaining the process model is difficult. This method uses online plant experimentation in order to perturb the plant and directly measure the performance index, which is used to determine the direction of the next RTO step [3]. The real time optimization model, process model is updated by using correction based on the plant data to overcome disturbances and model errors effects. Also, optimization algorithms are applied to estimate the optimum operating point [4]. In this paper we focused on model based method.

A general structure for model based real time optimization is shown in Figure 1 [5]. The general steps taken in one cycle of the real time optimizer are: steady state detection, data reconciliation and gross error detection, parameter estimation, process model, optimization and updating of process set points. In steady state detection when the process is close enough to its steady state condition the real time optimizer is allowed to update the model of the process. Data reconciliation adjusts the measured variables and if it is possible estimates any unmeasured variables. Gross error detection takes suitable corrective actions data from gross errors found in the process data. In the process of parameter estimation, the model parameters are adjusted with process data and then these parameters are used in the process model for optimization of set points.

Several projects have been conducted on real time optimization in the recent years; Real time optimization of boiler network with three different modeling types was compared to each other [6]. Online optimization based on simplex method was used in order to minimize the cycle time and reduce the percentage of rejects for an injection process [7]. Model based real-time optimization was applied on the pulp mill benchmark problem [8].

Economic optimization studies for petroleum and chemicals production systems have proven to be very beneficial while the algorithm tools have been implemented by the industry

and several commercial software packages such as HYSYS.RTO [9], PROFIT [10] and Aspen Technology RT.OPT [11] have been developed.

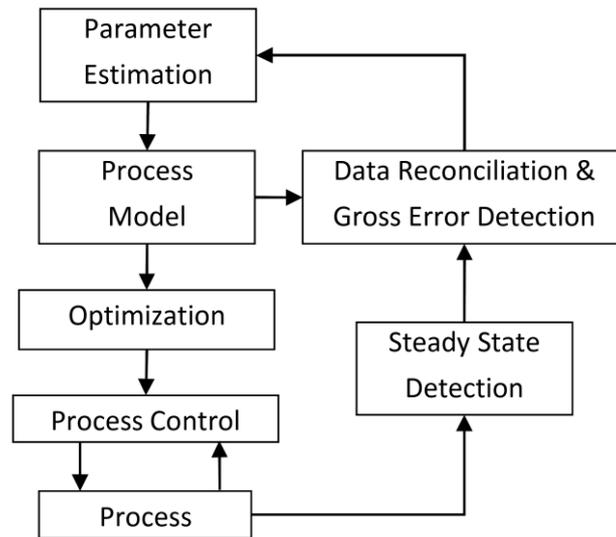


Figure 1 General structure of the model based real time optimization.

2. Process description

The case study in this work is low pressure methanol process accompanied syngas as feed shown in Figure 2.

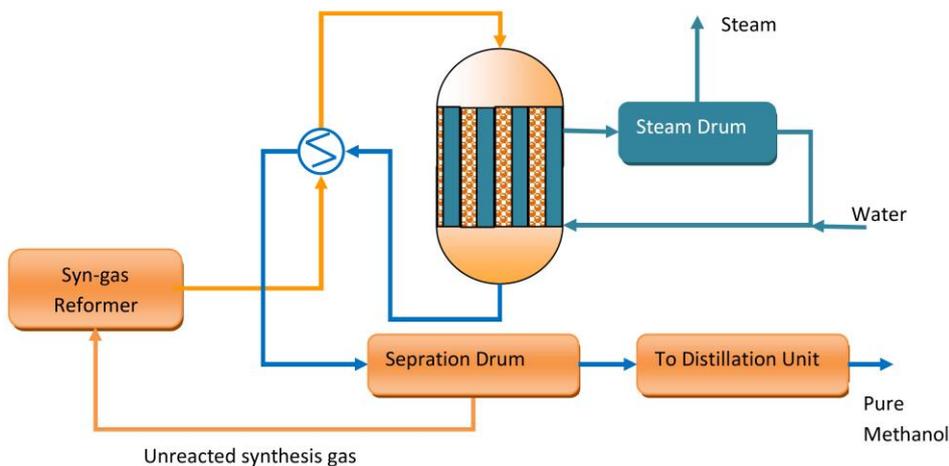


Figure 2 Flow sheet of the methanol synthesis loop

In the first step, syngas is converted to methanol in the shell and tube reactor. Then unreacted syngas is separated from crude methanol in the separation drum and after that compressed and recycled to feed stream. A part of recycle gas is purged to remove inert gases. The heat of reaction is transferred to shell side to produce saturated steam. The methanol reactor is basically a vertical shell and tube heat exchanger with fixed tube sheets, in which the tubes are packed with $\text{CuO}/\text{ZnO}/\text{Al}_2\text{O}_3$ catalysts and boiling water is circulated in the shell side. Adjusting of reactor temperature is persuaded by adjusting the steam pressure in the shell. This makes the reactor operate under isothermal condition in the shell.

Efficient heat transfer gives small temperature gradients through the reactor. Typical operating conditions are 521.15 K and 76.98 bars.

Three overall reactions are occurred in methanol reactors consist of hydrogenation of carbon monoxide, hydrogenation of carbon dioxide and the reverse water-gas shift reaction:



Kinetics of the low pressure methanol synthesis over commercial $\text{CuO}/\text{ZnO}/\text{Al}_2\text{O}_3$ catalysts has been widely investigated [12,13].

Table 1 Operating parameters of the Shiraz petrochemical plant methanol synthesis reactor

Feed composition	Mole fraction	Parameters	Value
CH ₃ OH	0.0050	Inlet pressure (bar)	76.98
CO	0.0460	Length of reactor (m)	7.022
CO ₂	0.0940	Bed void fraction	0.39
H ₂	0.6690	Tube inner diameter (m)	3.8 × 10 ⁻²
H ₂ O	0.0004	Tube outer diameter (m)	4.2 × 10 ⁻²
CH ₄	0.0926	Wall thermal conductivity (Wm ⁻¹ K ⁻¹)	48
N ₂	0.0930		

3. Model development

In this study two models are used: dynamic model for real plant and the steady state model for optimization studies. In the dynamic modeling, deactivation of catalyst is considered as it is represented by the following expression [14]:

$$\exp\left(\frac{-E_d}{R}\left(\frac{1}{T}-\frac{1}{T_R}\right)\right)a^5 \frac{da}{dt} = -K_d \quad (4)$$

where T_R , E_d and K_d are reference temperature, activation energy and deactivation constant of catalyst, with numerical value of 513 K, 91270 J/mol and 0.00439 h⁻¹ respectively [15].

The temperature and concentrations gradients between the phases are ignored and gas film on the catalyst surface is considered negligible. The chemical reactions are assumed to take place only in the fluid phase [16]. The mathematical model is carried out by use of homogeneous model. Mass and heat balances in fluid phase are as follows:

$$\varepsilon_B c_t \frac{\partial y_i}{\partial t} = -\frac{F_t}{A_c} \frac{\partial y_i}{\partial z} + r_i \rho_B a \quad i = 1, 2, \dots, N-1 \quad (5)$$

$$\varepsilon_B c_t c_{pg} \frac{\partial T}{\partial t} = -\frac{F_t}{A_c} \frac{\partial T}{\partial z} + \frac{\pi D_i}{A_c} U_{shell} (T_{shell} - T) + \rho_B a \sum_{i=1}^N r_i (-\Delta H_{f,i}) \quad (6)$$

where T and y_i are the temperature and mole fraction of component i in the fluid-phase, and a is the activity of catalyst.

The rates of reaction are as follow [12]:
Hydrogenation of carbon monoxide:

$$r_1 = \frac{k_1 K_{CO} [f_{CO} f_{H_2}^{3/2} - f_{CH_3OH} / (f_{H_2}^{1/2} K_{P1})]}{(1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2}) [f_{H_2}^{1/2} + k f_{H_2O}]} \quad (7)$$

Hydrogenation of carbon dioxide:

$$r_2 = \frac{k_2 K_{CO_2} [f_{CO_2} f_{H_2}^{3/2} - f_{CH_3OH} f_{H_2O} / (f_{H_2}^{3/2} K_{P2})]}{(1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2}) [f_{H_2}^{1/2} + k f_{H_2O}]} \quad (8)$$

Reversed water-gas shift reaction:

$$r_3 = \frac{k_3 K_{CO_2} [f_{CO_2} f_{H_2} - f_{H_2O} f_{CO} / K_{P3}]}{(1 + K_{CO} f_{CO} + K_{CO_2} f_{CO_2}) [f_{H_2}^{1/2} + k f_{H_2O}]} \quad (9)$$

where, the different parameters which observed in the reaction rate expressions have been offered by the following equations.

Reaction rate constants:

$$k_1 = (4.89 \pm 0.29) \times 10^7 \exp\left(\frac{-113000 \pm 300}{RT}\right) \quad (10)$$

$$k_2 = (1.09 \pm 0.07) \times 10^5 \exp\left(\frac{-87500 \pm 300}{RT}\right) \quad (11)$$

$$k_3 = (9.64 \pm 7.30) \times 10^{11} \exp\left(\frac{-152900 \pm 11800}{RT}\right) \quad (12)$$

Adsorption equilibrium constants:

$$K_{CO} = (2.16 \pm 0.44) \times 10^{-5} \exp\left(\frac{46800 \pm 800}{RT}\right) \quad (13)$$

$$K_{CO_2} = (7.05 \pm 1.39) \times 10^{-7} \exp\left(\frac{61700 \pm 800}{RT}\right) \quad (14)$$

$$k = \frac{K_{H_2O}}{K_{H_2}^{1/2}} = (6.37 \pm 2.88) \times 10^{-9} \exp\left(\frac{84000 \pm 1400}{RT}\right) \quad (15)$$

Equilibrium constants based on partial pressure ^[13]:

$$K_{P1} = 10^{\left(\frac{5139}{T} - 12.621\right)} \quad (16)$$

$$K_{P2} = 10^{\left(\frac{3066}{T} - 10.592\right)} \quad (17)$$

$$K_{P3} = 10^{\left(\frac{-2073}{T} + 2.029\right)} \quad (18)$$

4. Process control hierarchy

Typical process control hierarchy is broken down into scheduling, real-time optimization and process control as shown in Figure 3 ^[2].

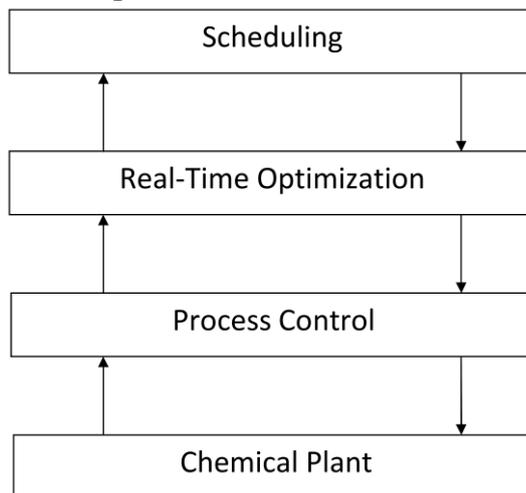


Figure 3 Process control hierarchy

The cascade design in process control hierarchy enables the decision to be made at different frequency in each layer. The upper scheduling layer deal with long term economic goals. Real-time optimization layer, in which the economics are considered explicitly, determines the best operating policy by maximizing the operating profit or minimizing the operating cost. The optimizer results are applied to the plant as controller set points in the process control layer. The lower process control layer includes conventional control and advanced process control. Proportional control is used as conventional control in this work.

5. Optimization algorithm

The method of global optimization consists of deterministic and stochastic approaches. The deterministic or gradient based methods need more computational efforts and it is possible to trap in local optima. Stochastic optimization methods which randomly search the optimum are somehow robust and able to find global optimum. Some of the most recognized stochastic search methods are Differential Evolution (DE), Genetic Algorithm (GA) and ant colony. In this study Differential Evolution and Genetic Algorithm are used and the results are compared.

5. 1 Genetic algorithm

Genetic algorithm (GA) is one of the stochastic methods based on biological evolution process to minimize or maximize the specified objective function. This algorithm chooses random pattern from a large space as initial population of individuals represented by chromosomes. It include three steps to find the best point: reproduction, crossover and mutation. In the reproduction step, the most fitness individuals are selected and placed in new population. Crossover step creates new individuals by mixing of some part of individuals randomly. The main purpose of mutation is to prevent the system from reaching and staying at local minima or maxima. When the individuals are bit string, the mutation points are often randomly selected bits, which are then complemented to create the new generation. After some number of generations the best individual would represent the optimum point.

5. 2 Differential evolution

Differential Evolution (DE) is an improved version of genetic algorithms. This method is simple, fast, robust and easy to use [17]. Based on the type of the problem, different types of strategies can be used in DE algorithm. As it is reported, as well as using different strategies, different values for number of population (NP), scaling factor (F), and crossover constant (CR) could be chosen in order to achieve better results. Selection of appropriate parameters in specific problems has a direct effect on runtime of optimization [18]. The parameters that used in this work is 15, 1 and 0.5 for NP, F and CR, respectively.

In these two algorithms the objective function is methanol output mole fraction and the shell temperature is considered as the decision variable.

6. Real time optimization:

The real time optimization algorithm proposed in this work applied on methanol synthesis reactor. Mathematical simulation of reactor is considered as the real plant. The schematic diagram of real time optimization and control loop is shown in Figure 4. The disturbances are input mole fractions of feed and input gas pressure, which are changed in the range of +20% of initial value. Optimization is carried out every hour and applied as the optimum set point to the plant. In the reactor the catalyst deactivity increases by chemical poisoning and thermal sintering. Sulphur compounds, chlorine and heavy metals act as poisons for the catalyst. Sintering is a solid state transformation which occurs at high temperatures and is promoted by water. Under normal operation only sintering occurs, because the catalyst poisons are removed from the syngas earlier in the process. Reducing the catalyst activity causes the model miss match in this study. In order to exterminate this miss match the model must be updated. Thus, the activity of the catalyst in the model is updated. Because the reduction of activity is low, updating the activity is considered every 24 hour.

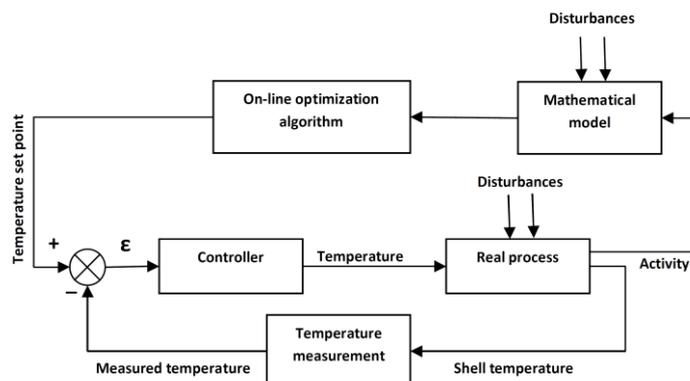


Figure 4 Schematic diagram of real time optimization and control

7. Result and discussions

In this study, an RTO system for low pressure methanol synthesis reactor is formulated. A dynamic model, considering deactivation model, is used as a real plant and DE and genetic algorithms are used as optimization tools. Due to the results, for one model-based optimization step, the DE and GA associated CPU time values are obtained as 203s and 1640s, respectively. Thus, DE algorithm is chosen in the rest of this study. The input mole fraction of feed components H_2O , methanol, CO , CO_2 , H_2 and CH_4 are considered as disturbances and the effects of each one are separately studied. The range of variation of disturbances is 20%. The results are shown in Figures 5 to Figure 9. Comparing the variation of shell temperature shows that the input mole fraction of H_2 is considerable and has significant effect on optimization than the other disturbances.

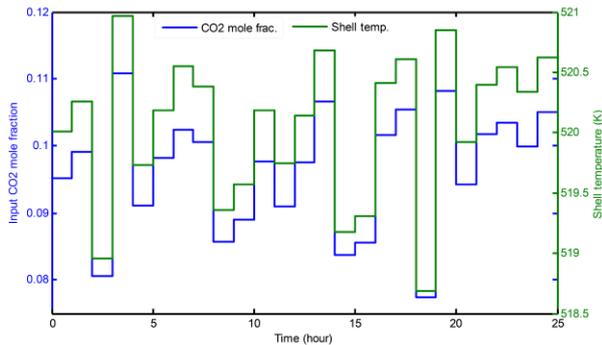


Fig. 5 Effect of CO_2 mole fraction variation on optimum shell temperature.

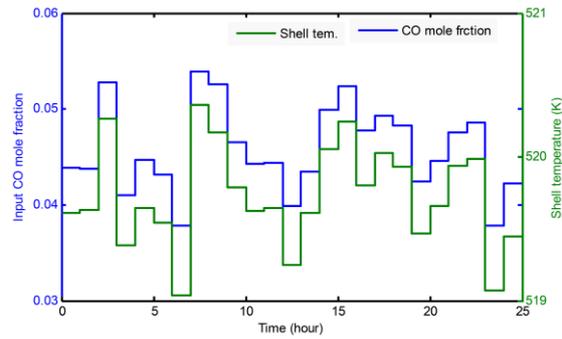


Fig. 6 Effect of CO mole fraction variation on optimum shell temperature.

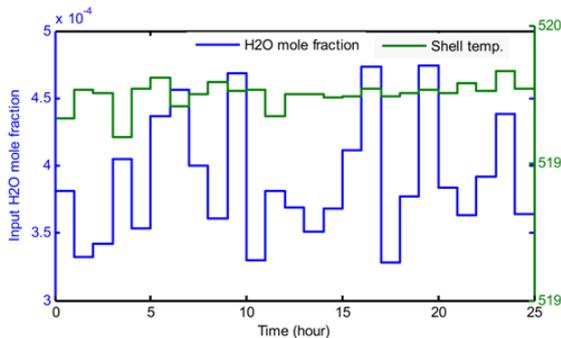


Fig. 7 Effect of H_2O mole fraction variation on optimum shell temperature

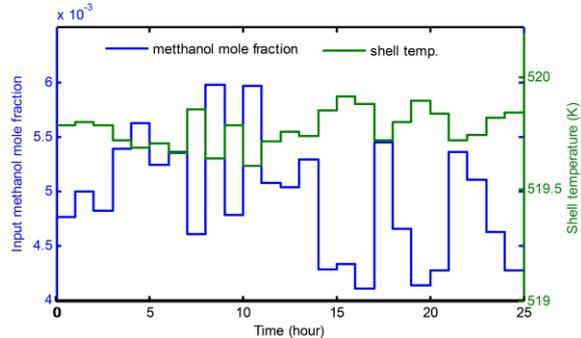


Fig. 8 Effect of methanol mole fraction (from recycle stream) variation on optimum shell temperature.

In the next step effects of input pressure change (up to +20%) investigated as a disturbance and the result of optimization is shown in the Figure 10.

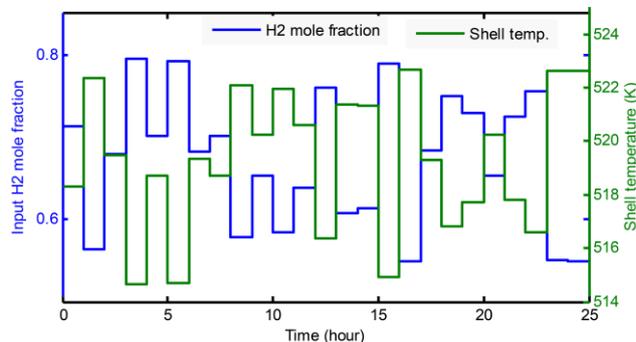


Fig. 9 Effect of Hydrogen mole fraction variation on optimum shell temperature

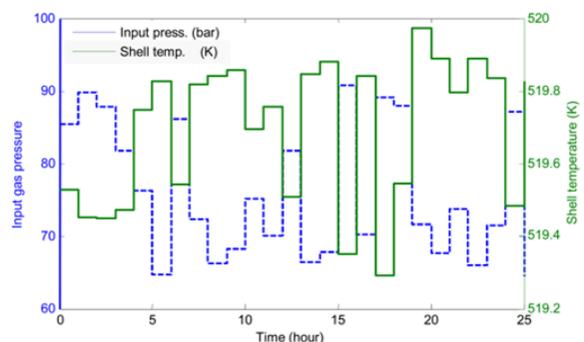


Fig. 10 Effect of input gas pressure variation on optimum shell temperature.

As it is seen, the input pressure does not have significant effect on the optimum shell temperature as decision variable.

In order to show the effect of catalyst activity reduction on optimum shell temperature, the simulation is conducted for 4 years without disturbance on input syngas. The result is shown in the Figure 11.

It can be seen that the catalyst deactivation rate cause the optimum shell temperature increase during the 4 years of operation.

In the next step one disturbance is considered, input hydrogen mole fraction (up to +20%) and shell temperature is considered as a decision variable, the result is shown in Figure 12.

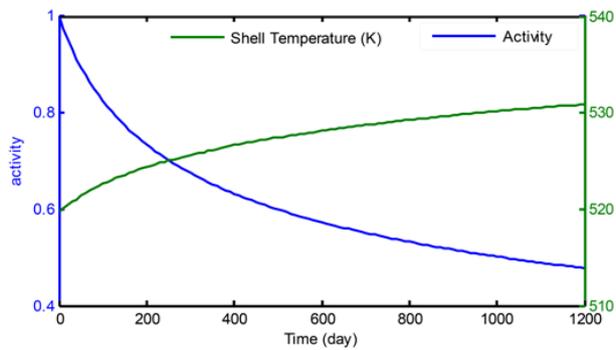


Fig. 11 Effect of catalyst activity reduction on optimum shell temperature.

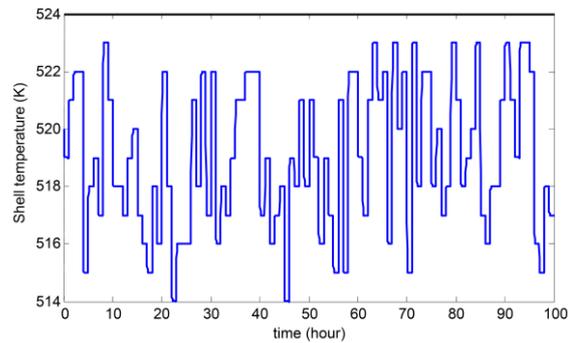


Fig. 12 Shell temperature of the reactor in RTO simulation when mole fraction of H_2 is considered as disturbance.

Using real time optimization on methanol reactor leads to up to 15% production. For a plant with a capacity of 5000 ton/day the profit will be 45 M\$/year if the price of methanol is considered as 200\$/ton.

8. Conclusions

Real time optimization of low pressure type of methanol synthesis reactor is the main goal of this study. Methanol production is the objective function while input concentrations and pressure are considered as disturbances. As the results show, changes in H_2 mole fraction have the most significant effect on optimum shell temperature. DE and genetic algorithms are employed as optimization strategies. The results show that DE algorithm needs less CPU time than the genetic algorithm. A feedback control loop is designed for controlling the shell temperature while set point is estimated by online optimization. It is concluded that using RTO for this process can be very beneficial especially in high capacity plants.

9. Nomenclature

A_c	cross section area of each tube (m^2)
a	activity of catalyst
c_{Pg}	specific heat of the gas at constant pressure ($J.mol^{-1}$)
c_t	total concentration ($mol.m^{-3}$)
D_i	tube inside diameter (m)
E_d	activation energy used in the deactivation model ($J.mol^{-1}$)
F_t	molar flow in each tube ($mol.s^{-1}$)
f_i	partial fugacity of component i (bar)
$\Delta H_{f,i}$	enthalpy of formation of component i ($J.mol^{-1}$)
K_d	deactivation model parameter constant (s^{-1})
K_i	adsorption equilibrium constant for component i (bar^{-1})
K_{Pi}	equilibrium constant for component i
K_w	thermal conductivity of reactor wall ($W.m.K^{-1}$)
k	$K_{H_2O} / K_{H_2}^{1/2}$
k_1	reaction rate constant for the 1 st rate equation ($mol.kg^{-1}.s^{-1}.bar^{-1}$)
k_2	reaction rate constant for the 2 nd rate equation ($mol.kg^{-1}.s^{-1}.bar^{-1/2}$)
k_3	reaction rate constant for the 3 rd rate equation ($mol.kg^{-1}.s^{-1}.bar^{-1}$)
R	universal gas constant ($J.mol^{-1}.K^{-1}$)

r_i	reaction rate of component i (mol.kg ⁻¹ .s ⁻¹)
r_1	rate of reaction for hydrogenation of CO (mol.kg ⁻¹ .s ⁻¹)
r_2	rate of reaction for hydrogenation of CO ₂ (mol.kg ⁻¹ .s ⁻¹)
r_3	reversed water-gas shift reaction (mol.kg ⁻¹ .s ⁻¹)
T	bulk gas phase temperature (K)
T_R	reference temperature used in the deactivation model (K)
T_{shell}	temperature of coolant stream (K)
t	time (s)
U_{shell}	overall heat transfer coefficient between coolant and process streams (W.m ⁻² .K ⁻¹)
y_i	mole fraction of component i in the fluid phase (mol.mol ⁻¹)
z	axial reactor coordinate (m)

Greek letters

ε_B	void fraction of catalytic bed (m ³ .m ⁻³)
ρ_B	density of catalytic bed (Kg.m ⁻³)

10. References

- [1] Cutler, C.R., Perry, R.T.:Real time optimization with multi-variable control is required to maximize profits, *Computers and Chemical Engineering*. 7 (1983) 663-667.
- [2] Marlin, T., Hrymark, A.N.:Real time operations optimization of continuous processes, *AIChE Symposium Series Fifth International Conference on Chemical Process Control*, 93 (1996)156-164.
- [3] Forbes, J.J.F., McGregor, F., Marlin, T.E.:Model adequacy requirement for optimization plant operation, *Computers and Chemical Engineering*, 18 (6) (1994) 497-510.
- [4] Roberts, P.D.: An algorithm for steady state system optimization and parameter estimation. , *Int. J. of System Sci.* 10 (7) (1979) 719-734.
- [5] Naysmith, M.R. :Doctorian Thesis ,Real time optimization of chemical process, A comparision of closed versus open form equation using commertial simulation package . Waterloo, 1997.
- [6] Yip, W.S., Marlin, T.E.:The effect of model fidelity on real-time optimization performance, *Computers and Chemical Engineering*, 28(2004) 267-280.
- [7] Jaziri, M., Chaabouni, M., Kamoun, A.:The use of the simplex method and its derivatives to the online optimization of theparameters of an injection moulding process, *Chemometrics and Intelligent Laboratory Systems*, 2008.
- [8] Mercangöz, M., Doyle, F. J.: Real-time optimization of the pulp mill benchmark problem, *Computers and Chemical Engineering*, 32 (2008) 789-804.
- [9] HYSYS.RTO Reference Guide. Hyprotech, a subsidiary of Aspen Technology, Inc. Anonymous nc. 2002.
- [10] PROFIT optimizer (DQP) concepts and implementation course notes. Honeywell Training Document. Anonymous, 1998.
- [11] Inc., Aspen Technology. The Aspen Leaf. Massachusetts s.n., 1996.
- [12] Scholtens, H., Stamhuis, E.J., Beenackers, A.A.C. M., Graaf, G.H.: Intra-particle diffusion limitations in low-pressure methanol synthesis, *Chem. Eng. Sci.* 45 (4) (1990) 773-783.
- [13] Graaf, G.H., Sijtsema, P.J.J.M., Stamhuis E.J. and Joosten, G.E.H.: Chemical equilibrium in methanol synthesis, *Chem. Eng. Sci.* 41(11) (1986) 2883-2890.
- [14] Hanken, L.:Optimization of methanol reactor. Master's thesis, The Norwegian University of Science and Technology, 1995.
- [15] Løvik, I.:Modeling, Estimation and Optimization of the Methanol Synthesis with Catalyst Deactivation, PhD thesis, Norwegian University of Science and Technology, 2001.
- [16] Rezaie, N., Jahanmiri, A., Moghtaderi, B., Rahimpour, M.R.: A comparison of homogeneous and heterogeneous dynamic models for industrial methanol reactors in the presence of catalyst deactivation, *Chem. Eng. Progress*, 44 (2005) 911-921
- [17] Babu, B.V., Angira, R.:Modified differential evolution (MDE) for optimization of non-linear chemical processes, *Computers & Chemical Engineering*, 30 (2006) 989-1002.
- [18] Babu, B.V., Munawar, S.A.: Differential evolution strategies for optimal design of shell-and-tube heat exchangers, *Chem Eng Sci.* 62 (2007) 3720-39.

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