Characteristics Dynamics and Control of Oxidative and Coupling Methane Catalytic Membrane Reactor

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Abstract. Oxidative and Coupling Methane (OCM) can be a promising technology for converting natural gas to Ethylene. Ethylene itself is an intermediate polymer product which is widely used for various needs of the chemical industry. The Oxidative and Coupling Methane reactor is currently in the researched to produce the most favorable conditions. The reactor configuration that is currently the most advantageous for application to oxidative and coupling methane reactors is the dual catalytic membrane reactor. This research is about controlling Methane and Ethane conversion systems in natural gas to produce Ethylene with the greatest conversion, determining the best tuning parameters for the three control structures, and comparing the three to get the best control arrangement. For this reason, control simulations are carried out on 9 controlled variables using three control structures, namely Proportional-Integral, Proportional Integral-Derivative, and Predictive Control Model. The disturbances given in this study were +20% and -20% in the composition of the feed flow operating conditions (temperature, pressure, and flow rate). A comparison of the IAE values of each controller is used to determine the best controller configuration.

1. Introduction

Ethylene is a very important intermediate chemical commodity. Ethylene production in the world exceeds the production of other organic compounds. Ethylene is used to produce polyethylene, polyvinyl chloride, ethylene oxide, ethylene chloride, ethylbenzene, alpha-olefins, linear alcohol, vinyl acetate, fuel mixtures, as well as aromatic compounds, alkanes and alkenes. Currently, the largest production of ethylene is produced by means of thermal cracking. Ethylene production by this process requires raw material in the form of naphtha [1]. In the thermal cracking process, heat is required to break the C-C and C-H chains. The main problem is the very high energy demand and very high environmental pollution [2]. It can be said that, 3 tonnes of CO2, a dangerous greenhouse gas, can be produced for every tonne of ethylene production. For this reason, efforts to conduct research related to processes that are simpler, cheaper, and environmentally friendly have been carried out for the last 30 years [3]. With the abundant availability of natural gas in the world (7299 TCF), Oxidative Coupling of Methane (OCM) has been researched as a potential solution. The OCM process involves the sequential partial oxidation Process that convert methane to ethane and then ethylene, the reaction in the OCM reactor is as follows [4]:

(Main Reaction)

\[ 2 \text{CH}_4 + \frac{1}{2} \text{O}_2 \leftrightarrow \text{C}_2\text{H}_6 + \text{H}_2\text{O} \quad \Delta H_{298} = -177 \text{kJ/mol (1)} \]

(Side Reaction)

\[ \text{C}_2\text{H}_6 + \frac{1}{2} \text{O}_2 \leftrightarrow \text{C}_2\text{H}_4 + \text{H}_2\text{O} \quad \Delta H_{298} = -105 \text{kJ/mol (2)} \]
CH₄ + 2 O₂ → CO₂ + 2 H₂O \quad \Delta H^{298}_r = -105 \text{ kJ/mol (3)}

CH₄ + \frac{1}{2} O₂ → CO + 2 H₂O \quad \text{(4)}

The entire process is divided into two units: FT reactor for increasing olefins produced in OCM as well as carbon monoxide and hydrogen consumption toward more C₅+ and in second reactor a reactor for OCM. First reactor is using fisher tropsch (FT) to change CO & CO₂ from overall process to become hydrocarbon, and second reactor for OCM reactor with catalyst to convert hydrocarbon (Methane, Ethane) to become Ethylene and minimizing side product from process[5]. The membrane FT reactor consists of shell and tube. Reaction takes place in shell side after natural gas process exiting gas passes through the tube side[6]. The tube wall in this system is capable of hydrogen selective permeation and hydrogen partial pressure gradient between the tube and shell tube allows diffusion of hydrogen through the Pd-based membrane layer to reaction section[7].

The formulation of the problems that arise in this study is the absence of dynamic simulation research on the OCM reactor system using Aspen Hysys software, the absence of research on control and optimization of the OCM Reactor system using Proportional Integral (PI) controllers, Proportional Integral Derivative (PID), Model Predictive Control (MPC). [8]. In terms of the process it is very expensive, besides the constraints to minimize unwanted by-products, namely CO and CO₂[9]. The purpose of this research is to know the dynamic characteristics of the OCM reactor system using Aspen Hysys software, to know the control strategy of Proportional Integral (PI), Proportional Integral Derivative (PID), the best Predictive Control (MPC) Model in the OCM Reactor system[10].

It is very important to determine the optimal operating conditions of the reactor to get the greatest ethylene conversion[11]. This research was conducted to control the methane reaction system with oxygen to ethylene by comparing the performance of the PI, PID & MPC controllers[12]. There is also a benefit from this research is that the latest reactor configuration and models are expected to be taken into consideration for further study so that it can be applied to the manufacture of OCM reactors at a factory scale. Steady state and dynamic process evaluation, Reducing or preventing operational risks, From the existing Reactor configurations obtained the best operating conditions under which to obtain the best possible Ethylene conversion. [13].

2. Experimental Section

2.1 Case Study

The case studied of this configuration is based on the research of A.Ghareghasi et al in figure 1. There is substantial information in the literature that deals with reaction kinetics of OCM and FT. Using La₂O₃/CaO catalyst and the reactor model used was one-dimensional. For FT, the kinetic equations based on experiments in a pilot plant in RIII and National Iranian Oil Company (NIOC), with bifunctional Fe-HZSM5 catalyst (metal part: 100 Fe/5.4 Cu/7 K₂O/21SiO₂, acidic part: SiO₂/Al₂O₃ = 28) was used.
Figure 1. Two Consecutive Reactor OCM & FTS Membrane

2.2 Kinetic model OCM and FTS Membrane Reactor
A set of 10 step kinetic model of OCM, and 6 step kinetic model of FTS Membrane over La2O3/CaO catalyst describes the differential rates of formation for different species including gas phase and catalytic reactions. The following sets of stoichiometric equations were considered:

\[ \text{CH}_4 + 2\text{O}_2 \rightarrow \text{CO}_2 + 2\text{H}_2\text{O} \quad (3) \]

\[ \text{CH}_4 + \frac{1}{2}\text{O}_2 \rightarrow \text{C}_2\text{H}_6 + \text{H}_2\text{O} \quad (4) \]

\[ \text{CH}_4 + \text{O}_2 \rightarrow \text{CO} + \text{H}_2\text{O} + \text{H}_2 \quad (5) \]

\[ \text{CO} + \frac{1}{2}\text{O}_2 \rightarrow \text{CO}_2 \quad (6) \]

\[ \text{C}_2\text{H}_6 + \frac{1}{2}\text{O}_2 \rightarrow \text{C}_2\text{H}_4 + \text{H}_2\text{O} \quad (7) \]

\[ \text{C}_2\text{H}_4 + 2\text{O}_2 \rightarrow 2\text{CO} + 2\text{H}_2\text{O} \quad (8) \]

\[ \text{C}_2\text{H}_6 \rightarrow \text{C}_2\text{H}_4 + \text{H}_2 \quad (9) \]

\[ \text{C}_2\text{H}_4 + 2\text{H}_2\text{O} \rightarrow 2\text{CO} + 4\text{H}_2 \quad (10) \]

\[ \text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2 \quad (11) \]

\[ \text{CO}_2 + \text{H}_2 \rightarrow \text{CO} + \text{H}_2\text{O} \quad (12) \]

\[ \text{CO} + 3\text{H}_2 \rightarrow \frac{R_1}{R_2} \text{CH}_4 + \text{H}_2\text{O} \quad (13) \]

\[ \text{CO} + 4\text{H}_2 \rightarrow \frac{R_1}{R_2} \text{C}_2\text{H}_4 + 2\text{H}_2\text{O} \quad (14) \]

\[ 2\text{CO} + 5\text{H}_2 \rightarrow \frac{R_1}{R_2} \text{C}_2\text{H}_6 + 2\text{H}_2\text{O} \quad (15) \]

\[ 3\text{CO} + 7\text{H}_2 \rightarrow \frac{R_1}{R_2} \text{C}_3\text{H}_8 + 3\text{H}_2\text{O} \quad (16) \]

\[ 4\text{CO} + 9\text{H}_2 \rightarrow \frac{R_1}{R_2} n - \text{C}_n\text{H}_{10} + 4\text{H}_2\text{O} \]

\[ \text{CO} + \text{H}_2\text{O} \rightarrow \frac{R_1}{R_2} \text{CO}_2 + \text{H}_2 \quad (17) \]

Water-Gas-Shift Reaction

Sets of stoichiometric equations in OCM Reactor

Sets of stoichiometric equations in FTS Membrane Reactor

The reaction rates and Kinetic Parameter for each step are given below:

\[ r_j = \frac{k_{a, j}e^{-\frac{E_{a, j}}{RT}}P_{e, j}^{m_j}p_{O_2}^{n_j}}{(1 + k_{j, CO_2}e^{-\frac{E_{j, CO_2}}{RT}}p_{CO_2}^{n_j})} \quad \text{for} \quad j = 1, 3-6 \]

\[ r_i = k_i \exp\left(-\frac{E_i}{RT}\right) p_{CO}^{m_i} p_{H_2}^{n_i} (\text{mol kg}_{cat}^{-1} \text{ s}^{-1}) \]
\[ r_2 = \frac{k}{a_2 e^{-\frac{E_{a2}}{RT} P_{O2}}} \left( \frac{\Delta H_{ad,co2}}{RT} P_{O2} \right) P_{CH4} \]

\[ r_3 = k_{0.7} e^{-\frac{E_{a3}}{RT} P_{C2H6}} \]

\[ r_4 = k_{0.8} e^{-\frac{E_{a4}}{RT} P_{C2H6} P_{H2O}^{m_8} P_{H2O}^{m_9}} \]

\[ r_5 = k_{0.9} e^{-\frac{E_{a5}}{RT} P_{CO} P_{H2O}^{n_9} P_{H2O}^{n_9}} \]

\[ r_6 = k_{0.10} e^{-\frac{E_{a6}}{RT} P_{CO} P_{H2O}^{n_9} P_{H2O}^{n_9}} \]

Reaction rate for each step in OCM Reactor

Table 1. Kinetic parameters of OCM reactions

| Step | \( K_{a,j} \) (mol/g) | \( E_{a,j} \) (kJ/mol) | \( m_j \) | \( n_j \) | \( K_{a,so2} \) (Pa\(^{-1}\)) | \( \Delta H_{ad,so2} \) (KJ/mol) | \( K_{a,co2} \) (Pa\(^{-1}\)) | \( \Delta H_{ad,co2} \) (KJ/mol) |
|------|-----------------|-----------------|--------|--------|-----------------|-----------------|-----------------|-----------------|
| 1    | 0.2 x 10\(^{-5}\) | 48              | 0.24   | 0.76   | 0.25 x 10\(^{-12}\) | -175            | 0.23 x 10\(^{-11}\) | -124            |
| 2    | 23.2            | 182             | 1      | 0.4    | 0.83 x 10\(^{-13}\) | -186            |                 |                 |
| 3    | 0.52 x 10\(^{-6}\) | 68              | 0.57   | 0.85   | 0.36 x 10\(^{-13}\) | -187            |                 |                 |
| 4    | 0.11 x 10\(^{-3}\) | 104             | 1      | 0.55   | 0.4 x 10\(^{-12}\) | -168            |                 |                 |
| 5    | 0.17            | 157             | 0.95   | 0.37   | 0.45 x 10\(^{-12}\) | -166            |                 |                 |
| 6    | 0.06            | 166             | 1      | 0.96   | 0.16 x 10\(^{-12}\) | -211            |                 |                 |
| 7    | 1.2 x 10\(^{-7}\) | 226             |        |        |                 |                 |                 |                 |
| 8    | 9.3 x 10\(^{-3}\) | 300             | 0.97   | 0      |                 |                 |                 |                 |
| 9    | 0.19 x 10\(^{-3}\) | 173             | 1      | 1      |                 |                 |                 |                 |
| 10   | 0.26 x 10\(^{-1}\) | 220             | 1      | 1      |                 |                 |                 |                 |

Table 2. Kinetic parameters of FTS Membrane reactions

| Reaction | m    | n    | \( K_i \) | \( E_i \) |
|----------|------|------|-----------|-----------|
| 1        | -1.0889 | 1.5662 | 142583.8 | 83423.9 |
| 2        | 0.7622  | 0.0728 | 51.556   | 65018    |
| 3        | -0.5645 | 1.3155 | 24.717    | 49782    |
| 4        | 0.4051  | 0.6635 | 0.4632    | 34885.5  |
| 5        | 0.4728  | 1.1389 | 0.00474   | 27728.9  |
| 6        | 0.8204  | 0.5026 | 0.00832   | 25730.1  |
| 7        | 0.5850  | 0.5982 | 0.02316   | 23564.3  |
| 8        | 0.5742  | 0.710  | 410.667   | 58826.3  |

2.3 Mathematical modeling of OCM and FT reactor

2.3.1 Mathematical modeling of OCM and FT reactor

For OCM & FTS Membrane reactor a one-dimensional model was developed to simulate the heterogeneous reactions. The system has been modeled assuming the following assumptions [14]:

(a) Steady state.
(b) One-dimensional plug flow reactor packed with La2O3/CaO catalyst.
(c) Vapor phase is assumed to be ideal gas.

The mass and energy equations for the bulk gas phase can be written as:

$$\frac{-F_t}{A_c} \frac{dy_i}{dz} + a_v c_i k_{gi} (y_{is} - y_i) = 0$$  \hspace{1cm} (18)

$$\frac{-F_t}{A_c} c_{pg} \frac{dT}{dz} + a_v h_f (T_S - T) + \frac{\pi D_i}{A_c} U_{shell} (T_{shell} - T) = 0$$  \hspace{1cm} (19)

where, \(y_i\) and \(T\) are the gas-phase mole fraction and temperature, respectively. The boundary conditions for the bulk phase at \(z = 0\) are expressed by[15]:

\[y_i = y_{i,\text{in}}; \quad T = T_{\text{in}}\]

The mass and energy balance equations for the catalyst pellets can be formulated as:

$$a_v c_i k_{gi} (y_i - y_{is}) + \rho_B \tau_i = 0, i = 1, 2, 3, \ldots$$  \hspace{1cm} (20)

$$a_v h_f (T - T_s) + \rho_B \eta \sum \tau_i (-\Delta H_i) = 0$$  \hspace{1cm} (21)

where, \(y_s\) is and \(T_s\) are the mole fractions on the catalyst surface and solid phase temperature, respectively. Momentum balance for the reactor (Ergun’s equation)[16] is:

$$-\frac{dP}{dz} = \frac{\rho g u_f^2 (1-\varepsilon)}{\varepsilon^2} \left[ \frac{150(1-\varepsilon)}{\varepsilon \eta} + 1.75 \right]$$  \hspace{1cm} (22)

The net rates of formation of each component was given by \(\tau_i = \sum v_{ij} R_j\) where \(R_j\) represents rate of reaction \(j\) and \(v_{ij}\) is stoichiometric coefficient. The specific heat and heat of reactions are computed as a function of temperature[17]. The reaction rates were expressed as a function of partial pressure and temperature[18].

2.4 Thermodynamic Model Selection

In this study, the thermodynamic model is using Peng-Robinson[16]. The advantages and consideration of using this model in a simulation package because it is the most developed model in Aspen Hysys, High precision in a wide range of temperature and pressure, Special treatment for key components, Wide Data Bank of binary parameters[19].

2.5 Control Structure

In this study, 3 control structures were used[20]. The first structure used Proportional Integral (PI) controller, Proportional Integral Derivative (PID), and Model Predictive Control (MPC).

The following figure shows the structure of the PI, PID, and MPC controllers[21].
In Controlled Variable using : Temperature Inlet OCM Reactor, Percent Level Liquid in flash drum, Molar Flowrate Vapour Effluent, and Mol Fraction Ethylene Product by Manipulating Variable (Heat Duty Outlet Cooler, % Opening Valve Liquid Effluent, % Opening Valve Vapour Effluent, and Flowrate Molar Inlet in OCM Reactor)[20]. Disturbance give in CO2: H2 Ratio Inlet FTS Membrane Reactor, Molar Flowrate O2 (± 20%), and Temperature Inlet OCM Reactor.

2.6 Tuning Parameter MPC
Tuning is performed MPC parameters, by change (sample time), (prediction horizon), and (control horizon) in HYSYS, tuning is done by entering the parameter values that have been calculated using the non-adaptive DMC tuning strategy approach (Dougherty, 2003a) on the MPC Setup (Advanced) tab or parameter values obtained by trial error.

2.6.1 Sampling Procedure
The sample take and analyze in this study is a controlled variable response graph (CV). The controlled variable response graph was obtained after running the MPC program on HYSYS and tuning each controller.

2.6.2 Analysis Procedure
The sample analysis procedure is doing by calculating the Integral Absolute Error (IAE) value in the controlled variable response graph (CV) with the MPC controller that has been run. IAE values in MPC controllers are compared with IAE in PI, PID controllers. A controller with a smaller IAE value is the controller with better performance.

2.6.3 Calculation Method
1. Creating an empirical model
One of the most widely used identification methods to identify a dynamic empirical model in a process is the Process Reaction Curve (PRC). In PRC, appropriate parameters (dead time, time constant, and damping coefficient) can be determined with experimental response step data. The process identified is an open loop system, but in experimental testing, the closed loop system can also be identified. The stages in the process reaction curve include:

1. Steady state process condition
2. Process indicates a single step change in the input variable
3. Recording input and output responses until the process reaches steady state condition
4. Shows graphical calculations for the process reaction curve.

Graphical calculations are performed to determine the parameters in the first-order-with-dead-time (FOPDT) model. The model form is shown in equation (6) with X (s) as input and Y (s) as output.

\[
\frac{Y(s)}{X(s)} = \frac{K_p e^{-\theta s}}{rs + 1}
\]

(23)
The values determined from the graph are the change in input (δ), the steady state change in output (∆), and the time it takes for the output to reach 28% and 63% of its final value. The value obtained from the graph can be used to calculate the model parameters formulated in equation below:

\[ K_p = \frac{\Delta}{\delta}, \quad t_{63\%} = \theta + \tau \]

\[ t_{28\%} = \theta + \frac{\tau}{3}, \quad \theta = t_{63\%} - t_{28\%} \]

(24)

2. Tuning Parameter

MPC parameters are calculated using the DMC non-adaptive tuning strategy approach (Dougherty, 2003a) as follows:
a) The process dynamics approach of the controller output for pairs of measured process variables with the FOPDT model

\[ \frac{Y_r(s)}{U_s(s)} = K_{rs}e^{-\theta rs} \frac{\tau rs}{\tau rs + 1}, \quad r = 1, 2, ..., R ; \quad s = 1, 2, ..., S \]  

(25)

b) Choosing the closest sample time to:

\[ T_n = \text{Max} (0.1 \tau_n, 0.5O_n) \]

\[ T = \text{Min} (T_n), \quad r = r \quad 1, 2, ..., R ; \quad s = 1, 2, ..., S \]  

(27)

c) Calculating (prediction horizon):

\[ P = \text{Max} \left( \frac{\theta rs}{T} + K_{rs} \right) \]

\[ K_n = \frac{\theta rs}{T} + 1 \], \quad r = r \quad 1, 2, ..., R \quad s = 1, 2, ..., S \]  

(29)

d) Calculating (Control horizon):

calculated using the equation:

\[ M = \text{Max} \left( \frac{\tau rs}{T} + K_{rs} \right) \]  

(30)

3. Calculating Integral Absolute Error

IAE (Integral Absolute Error) Calculate Using Equation

\[ IAE = \int |SP(t) - CV(t)|dt \]  

(31)

In the controlled variable response graph (CV), the IAE is the absolute area of the difference between the set point graph area and the CV response graph area. The smaller the IAE, the closer the CV to the set point, thus indicating the better performance of the controller used. The calculation of the area under the graph is done using the Newton-Cotes Integration formula.

3. Research Procedure

The research method of this study consists of 9 Step. First step steady state simulation using the Aspen Hysys program and validating steady state simulation results if steady state simulation not match with literature simulation need to be repeated; Next is sizing stage and dynamic modeling; Step test on
each steam flow rate; Use of simulation programs with the addition of a Proportional Integral (PI) controller configuration, Proportional Integral Derivative (PID), Model Predictive Control (MPC); PI, PID, MPC controller tuning; Dynamic simulation program testing mode by providing disturbance; Determination of the optimum parameter is reviewed based on the Integral of The Absolute Value of The Error (IAE) method.

In System, the reflux drum level of the flash drum use Single Input Single Output (SISO) system. The SISO systems use conventional Proportional Integral Derivative (PID) controller. The MIMO controller is designed by creating the mass fractions of the ethylene products outlet OCM reactor, Heat Duty Outlet Cooler, % Opening Valve Liquid Effluent, % Opening Valve Vapour Effluent, and Flowrate Molar Inlet in OCM Reactor are manipulated variables and the change of feed O₂ is disturbance variables as in figure 6.

The next step is MPC Parameter Tuning uses the adaptive tuning method and Trial error. To get the First Order and Dead Time (FOPDT) to get matrix of Transfer Function 5x5. The dynamic system of the plant is Multiple Input Multi Output (MIMO) process, so that the plant needs MIMO controls system to keep the condition of steady state to the dynamic. The matrix of Transfer Function 5x5 is defined as equation (15) and equation below:

\[
\begin{bmatrix}
C_1 \\
C_2 \\
C_3 \\
C_4 \\
C_5 \\
\end{bmatrix} =
\begin{bmatrix}
G_{p,1.1} & G_{p,2.1} & G_{p,3.1} & G_{p,4.1} & G_{p,5.1} \\
G_{p,1.2} & G_{p,2.2} & G_{p,3.2} & G_{p,4.2} & G_{p,5.2} \\
G_{p,1.3} & G_{p,2.3} & G_{p,3.3} & G_{p,4.3} & G_{p,5.3} \\
G_{p,1.4} & G_{p,2.4} & G_{p,3.4} & G_{p,4.4} & G_{p,5.4} \\
G_{p,1.5} & G_{p,2.5} & G_{p,3.5} & G_{p,4.5} & G_{p,5.5} \\
\end{bmatrix}
+ \begin{bmatrix}
G_{d,1} \\
G_{d,2} \\
G_{d,3} \\
G_{d,4} \\
G_{d,5} \\
\end{bmatrix} F \quad (32)
\]

Tuning Adaptive and Trial error methods started by make several process variation and performing identification System, then tuning MPC parameter doing by make calculations based on equation 9-14 to get the parameter prediction horizon, control horizon, and control Interval.

Tuning MPC Parameter use Tuning Trial Error and Tuning non adaptive doing by make process variation in changing the output of the O₂ molar flow rate for the step response test and other controller outputs are varied at a certain value, resulting in model variations. These models have valve opening specifications in the initial and final conditions shown in the table 3&4.

| Table 3 % Valve Opening Spesification on each Model |
|---|---|---|
| Model | (%) Op Vlv | (%) Op Vlv Change |
| A | 50 | 52.5 |
| B | 46 | 53 |
| C | 52.8 | 53.8 |
| D | 52.1 | 53.9 |
| E | 50 | 54.5 |
| F | 45.8 | 45.6 |
Model A is the default testing model because it has the smallest change in the percent of valve opening. Model B is made based on model testing with a 7% difference in valve opening from its initial condition. Determination of the initial valve opening in this testing model is done by testing certain valve opening conditions until a stable system condition is obtained. At the opening conditions of 40% and 44%, the system becomes unstable which is indicated by the absence of products formed and the temperature of the reactor that has dropped beyond the specified CV limit. The valve opening that can still make the system stable is at 45% open, so that this B model is made larger than 45%, which is 46% in the initial condition.

Models C, D, E and F were made by determining the Ethylene product concentration set point, which is 0.2637. This value is based on the results of trial error on the PFR system simulation, because at the condition of achieving set point > 0.27 the system becomes more unstable. Then determined the initial conditions for the concentration of the product so that it has a difference of 2.5%, 5%, 10% and 20% to the predetermined set point. So the percentage referred to is not based on valve opening but based on the difference in the set point concentration of the initial and final product.

The performance of the controller is tested by changing the set point (SP) of product concentration and temperature at a certain value. SP changes made are classified into 7 scenarios. Scenario 1 is a product concentration that is determined in a low change range, from 0.2637 to 0.270293 and a temperature from 1210°C to 1240.25°F. While scenario 2 to 7 is a change in SP according to changes in product concentration and temperature in the PRC of each model. SP changes in each scenario are shown in Table 9.

### Table 4. Set point Change (SP) in every Scenario

| Scenario | Xp Concentration Change | Sp Temperature Change | Flowrate O2 |
|----------|-------------------------|-----------------------|-------------|
| 1        | 0.26                    | 0.27                  | 1210.00     |
| 2        | 0.26                    | 0.26                  | 1210.00     |
| 3        | 0.26                    | 0.28                  | 1210.00     |
| 4        | 0.26                    | 0.29                  | 1210.00     |
| 5        | 0.26                    | 0.30                  | 1210.00     |
| 6        | 0.26                    | 0.30                  | 1210.00     |
| 7        | 0.26                    | 0.32                  | 1210.00     |

Model B is made based on model testing with a 7% difference in valve opening from its initial condition. Determination of the initial valve opening in this testing model is done by testing certain valve opening conditions until a stable system condition is obtained. At the opening conditions of 40% and 44%, the system becomes unstable which is indicated by the absence of products formed and the temperature of the reactor that has dropped beyond the specified CV limit. The valve opening that can still make the system stable is at 45% open, so that this B model is made larger than 45%, which is 46% in the initial condition.

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### 4. Results and discussion

Tuning Parameter from PI, & PID controller Getting by autotuning in HYSYS. Table Below show Control Parameter for Kc, ti, td, ultimate gain, and ultimate period from each controller.

| Controlled Variable | Manipulated Variable | Kc | ti | td | Ultimate Gain | Ultimate Period |
|---------------------|----------------------|----|----|----|---------------|-----------------|
| TIC                 | Temperatur Inlet OCM  | 0.093 | 0.36 | 1.09 | 1.21          |
|                    | Reactor Heat Duty     |    |    |    |               |                 |
|                    | Keluruan Cooler       |    |    |    |               |                 |
| LIC                 | Percent Level Liquid  | 16  | 0.78 | 1.8 | 10.33         |
|                    | Liquid in flash drum  |    |    |    |               |                 |
|                    | Valve Liquid Effluent |    |    |    |               |                 |
| FIC                 | Vapour Molar Flowrate | 8.7 | 0.07 | 0.28 | 0.74          |
|                    | Effluent Valve Vapour |    |    |    |               |                 |
To obtain the best tuning parameters from control using a Model Predictive Control (MPC), it is necessary to know the effect of each manipulated variable on each controlled variable, so it is necessary to do a Step Test Response. Step Test Response is a method for obtaining the model used by MPC to make predictions. The step test in this simulation is carried out in dynamic mode by providing interference in the form of an input step on one of the manipulated variables by keeping the other manipulated variables constant.

Models from Step Test Response, tuning Parameter MPC shown in Table Bellow:

Table 6. Tuning Parameter Konventional PID Controller

| Nama | Controlled Variable | Manipulated Variable | Kc | t1 | tI | td | Ultimate Gain | Ultimate Period |
|------|---------------------|----------------------|----|----|----|----|---------------|-----------------|
| TIC  | Temperatur Inlet OCM Reactor | Heat Duty Keluaran Cooler | 0,573 | 2,29 | 0,17 | 1,26 | 1,04 |
| LIC  | Persent Level Liquid in flash drum | Valve Liquid Effluent | 0,77 | 23,7 | 1,71 | 1,69 | 10,8 |
| FIC  | Vapour Molar Flowrate Effluent | Valve Vapour Effluent | 0,06 | 1,25 | 0,09 | 0,13 | 0,57 |
| XIC  | Ethylene Product mol Fraction | Laju Alir Molar CO2 | 0,08 | 88,5 | 6,39 | 0,18 | 40,3 |
| PIC  | Pressure Outlet compressor | Heat Duty Kompressor | 0,53 | 0,61 | 0,04 | 1,17 | 0,28 |

Table 7. Tuning Parameter MPC using Update Step Response Test

| Parameter | Nilai |
|-----------|-------|
| P         | 94    |
| M         | 10    |
| Gamma-U   | 1     |
| Gamma-Y   | 1     |
| Ref. Trajectory | 1 |
| Control interval | 30 s |

| Nama       | Controlled Variable | Manipulated Variable | Kc | t1 | tI | td | Ultimate Gain | Ultimate Period |
|------------|---------------------|----------------------|----|----|----|----|---------------|-----------------|
| LIC        | Persent Level Liquid di flash drum | Valve Liquid Effluent | 0,77 | 23,7 | 0 | 1,69 | 10,8 |

Figure 7-16 compare each controller Performance
Figure 4. Response Vapour Molar Flowrate outlet flash drum (+20% O₂). The Value of settling time for PI,PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 13.0861, PID = 11.3944, and MPC = 10.1132. From the graph in Figure 8 above, it can be seen that the three controllers are able to maintain the controlled variable condition back to the set point, the three of them have a trend to increase, this is due to the increase in the composition of Methane in the gas component of the flash drum which has a large enough facor compressibility compared to the liquid component, so that the liquid component will decrease and gas will increase. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.

Figure 5. Response Vapour Molar Flowrate outlet flash drum (-20% O₂). The Value of settling time for PI,PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 4.3664, PID = 2.7487, and MPC = 1.5805. From the graph in Figure 9 above, it can be seen that the three controllers are able to keep the controlled variable condition back to the set point, the three of them have a downward trend, this is due to the decrease in the composition of Methane in the flash drum which has greater compressibility than the gas component. But there are some differences between the three, in the graph above it can be seen that the PID
controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.

**Figure 6.** Response Liquid Percent lvl outlet flash drum (+20% O₂). The Value of settling time for PI, PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 1.8694, PID = 0.8170, and MPC = 0.5926. From the graphs in Figure 10 above, it can be seen that the three controllers are able to keep the controlled variable conditions back to the set point, the three of them have a trend to increase first then constant, this is due to the increase in liquid composition in the flash drum, due to the reduced gas composition at the beginning. But then over time the molar flow rate of the gas components tends to increase and becomes constant so that the liquid component in the flash drum will increase and then become constant. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller overshoot than PI but bigger when compared to MPC.

**Figure 7.** Response Liquid Percent lvl outlet flash drum (-20% O₂). The Value of settling time for PI, PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 1.8694, PID = 0.8170, and MPC = 0.5926. From the graph in Figure 11 above, it can be seen that the three controllers are able to keep the controlled variable condition back to the set point, the three of them have a trend
to decrease first then constant, this is due to the decrease in liquid composition in the flash drum, due to the gas composition increasing at the beginning but then over time the molar flow rate of the gas component tends to decrease and becomes constant so that the liquid component in the flash drum will decrease and then become constant. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller overshoot than PI but bigger when compared to MPC.

**Figure 8.** Response Pressure Outlet compressor (+20% O₂) The Value of settling time for PI, PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 5.3595, PID = 0.0014, and MPC = 2.7187. From the two graphs in Figure 12 above, it can be seen that the three controllers are able to keep the controlled variable condition back to the set point, the three of them have a trend to go up and down first then decrease, this is because the increase in the composition of O₂ has a fairly large compressibility factor compared to other components in it. feed reactor OCM such as Methane, CO, Ethane, and H₂. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.
Figure 9. Response Pressure Outlet compressor (+20% O₂) The Value of settling time for PI,PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 3.5730, PID = 0.0009, and MPC = 1.8125. From the graph in Figure 13 above, it can be seen that the three controllers are able to maintain the controlled variable condition back to the set point, the three of them have a trend to decrease first then increase, this is due to the decrease in the composition of O₂ which has a large enough compressibility factor compared to Methane, CO, Ethane, and H₂. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.

Figure 10. Response Temperature Outlet cooler (+20% O₂) The Value of settling time for PI,PID and MPC Controller = 1 Hour. The Value of Integral Absolute Error (IAE) for PI = 28.8158, PID = 21.2751, and MPC = 12.6361. From the two graphs in Figure 14 above, it can be seen that the three controllers are able to keep the controlled variable conditions back to the set point, the three of them have a trend to decrease first then increase, this is due to the decrease in the composition of O₂, which causes ethylene products which have a fairly low heat capacity decreased. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.
The Value of settling time for PI, PID and MPC Controller = 1 Hour. The Value of Integral Absolute Error (IAE) for PI = 19.2105, PID = 14.1834, and MPC = 8.4241. From the two graphs in Figure 15 above, it can be seen that the three controllers are able to maintain the controlled variable condition back to the set point, the three of them have a trend to decrease first then increase, this is because the composition of the ethylene product that comes out of the OCM reactor has a fairly low heat capacity. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.

The Value of settling time for PI, PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 0.0046, PID = 0.0027, and MPC = 0.0018. From the graph in Figure 16 above, it can be seen that the three controllers are able to keep the controlled variable conditions back to the set point, the three of them have a trend to increase first then decrease, this is because the composition of the O2 flow rate will show the conversion to ethylene products with the component flow rate slightly if the flow rate of the O2 component is increased, the conversion of ethylene products will decrease due to the formation of higher hydrocarbon groups such as propylene (C-
3), C-4, to gologan C-5. But there are some differences between the three, in the graph above it can be seen that the PID controller has a much smaller settling time and overshoot than PI but bigger when compared to MPC.

![Response Ethylene Prod mol fraction (+20% O₂)](image)

**Figure 13.** Response Ethylene Prod mol fraction (+20% O₂) The Value of settling time for PI,PID and MPC Controller = 2 Hour. The Value of Integral Absolute Error (IAE) for PI = 0.0030, PID = 0.0018, and MPC = 0.0012. From the graph in Figure 17 above, it can be seen that the three controllers are able to maintain the controlled variable condition back to the set point, the three of them have a trend to decrease first then increase, this is because the composition of the O₂ flow rate will show the conversion to ethylene products with the component flow rate a little if the flow rate of the O₂ component initially decreases, the ethylene conversion obtained is not maximal because of the lack of O₂ groups needed to react with the carbon groups to form hydrocarbons. But there are some differences between the three, in the graph above it can be seen that the PID controller has a settling time and overshoot which is much smaller than PI but bigger when compared to MPC.

Strategies in Optimizing the MPC Controller is to use Tuning non-adaptive and trial error to be able to determine the calculation algorithm and update the new MPC tuning parameters to be used on MPC controllers that have updated the step response based on model in table 8 and Set Point change in table 9.

Based on average IAE in each model IAE in model A = 0.551660184, IAE in model B = 0.404068572, IAE in model C = 0.253975135, IAE in model D = 0.266042162, IAE in model E = 0.25650074, IAE in model F = 0.257624685, showing that model C is best optimization model that can be applied in every scenario by giving minimum IAE compared to another model. Optimization use Tuning MPC parameter in model C have tuning parameter below:

**Table 8.** Tuning Parameter MPC Optimization using Tuning non-adaptive and trial error

| Parameter     | Nilai |
|---------------|-------|
| P             | 100   |
| M             | 55    |
| Gamma-U       | 1     |
| Gamma-Y       | 1     |
| Ref. Trajectory| 1   |
| Control interval | 15 s |
| Nama   | Controlled Variable | Manipulated Variable | Kc  | t_i | t_d | Ultimate Gain | Ultimate Period |
|--------|---------------------|----------------------|-----|-----|-----|--------------|----------------|
| LIC    | Percent Level       | Liquid di            | 0,77| 23,7| -   | 1,69         | 10,8           |

Detailed calculation algorithm use model C

\[
\begin{bmatrix}
F_{LC} \\
L_{IC} \\
P_{LC} \\
X_{LC}
\end{bmatrix}
= \begin{bmatrix}
40.8 e^{-0.06s} & 21.16e^{-0.38s} & 59.12e^{-0.065s} & 20.4e^{-0.057s} & 0.063e^{-0.243s} \\
4.507s + 1 & 6.02s + 1 & 3.718s + 1 & 7.81s + 1 & 5.517s + 1 \\
52.8e^{-0.067s} & 19.28e^{-0.087s} & 84.8e^{-0.042s} & 74e^{-0.017s} & 0.051e^{-0.345s} \\
9.333s + 1 & 9.313s + 1 & 3.525s + 1 & 3.7s + 1 & 4.788s + 1 \\
43.2e^{-0.036s} & 9.48e^{-0.418s} & 298.8e^{-143s} & 317.6e^{-0.01s} & 0.021e^{-0.232s} \\
9.848s + 1 & 6.948s + 1 & 9.79s + 1 & 5.606s + 1 & 6.985s + 1 \\
43.2e^{-0.258s} & 21.5e^{-0.3518s} & 78.4e^{-0.005s} & 6.8e^{-0.325s} & 0.061e^{-0.285s} \\
4.875s + 1 & 5.848s + 1 & 3.745s + 1 & 3.175s + 1 & 4.748s + 1 \\
54^{-0.2s} & 21.472e^{-0.367s} & 60e^{-0.388s} & 0.055e^{-0.006s} & 0.026e^{-2.183s} \\
5.2s + 1 & 6.197s + 1 & 4.528s + 1 & 5.528s + 1 & 10.05s + 1
\end{bmatrix}
\]

5. Conclusion
Ethylene conversion control in the Oxidative and Coupling Methane reactor system using 5 control variables can be done with three Proportional Integral (PI) control structures, Proportional Integral Derivative (PID), and Predictive Control (MPC) models with MPC controller configuration using 1 PID Controller in percent lvl liquid, and another variable using MPC Controller although by Overall, MPC controller is not yet able to control the system Reactor with optimum Product ethylene reaction based on IAE values, however, the MPC controller is better able to maintain system stability than the PI controller.

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Symbols Used

\( y_i \) [-] Gas phase mol fraction

\( A_C \) [m²] Cross section area of tube

\( A_{\text{shell}} \) [m²] Cross section area of shell

\( a_v \) [m³m⁻³] Specific surface area of catalyst pellet

\( c_p \) [Kj mol⁻¹ K] Specific heat

\( C_t \) [mol m⁻³] Total concentration

\( C_j \) [mol m⁻³] Concentration of species \( j \)

\( d_t \) [m] Tube diameter

\( d_p \) [m] Pellet diameter

\( D_i \) [m] Tube inside diameter

\( D_{ro} \) [m] Tube outside diameter

\( F_t \) [mol/s] Total molar rate

\( h_f \) [W/m²K] Gas-catalyst heat transfer coefficient

\( \Delta H_i \) [kj/mol] Heat of reaction

\( k_{gi} \) [mol/s] Mass transfer coefficient between Gas and solid phase for component \( i \)

\( P_{\text{H}i}^{\text{sh}} \) [Bar] Shell side pressure

\( P_{\text{H}i}^f \) [bar] Tube side pressure

\( P \) [mol/m¹s²Pa⁰.⁵] Permeability of hydrogen through Pd–Ag layer

\( r_i \) [mol g⁻¹s] Rate of formation of reaction

\( R_i \) [M] Inner radius of Pd–Ag layer

\( R_O \) [M] Outer radius of Pd–Ag layer

\( R_e \) [-] Reynolds number

\( T \) [K] temperature

\( T_{\text{ex}} \) [K] External temperature

\( T_{\text{shell}} \) [K] Temperature of coolant stream, infixed bed reactor

\( U_S \) [m/s] Superficial velocity

\( U \) [Wm⁻²K] Overall heat transfer coefficient

\( U_{\text{shell}} \) [Wm⁻²K] Overall heat transfer coefficient between coolant and process streams
Greek symbols

- $\varepsilon_b$ [-] Catalyst bed porosity
- $\rho_b$ [g/m$^3$] Density of catalyst in the bed
- $\rho_g$ [kg/m$^3$] Density of system gas
- $\alpha_{H}$ Mol m$^{-1}$ s$^{-1}$ pa$^{-0.5}$ hydrogen permeation rate constant
- $\eta$ [-] Catalyst effectiveness factor
- $\psi$ [-] Shape factor

Abbreviation

- FOPDT First-order plus dead time
- IAE Integral absolute error
- MIMO Multiple input multiple Output
- MPC Model predictive control
- SISO Single input single output
- PI Proportional Integral
- PID Proportional Integral Derivatives

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