



A Comparative Study between Design and Parameter Adjustment for Profit Maximization of Low-Density Polyethylene (LDPE) Production in High-Pressure Tubular Reactor

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ABSTRACT

Low density polyethylene (LDPE) market becomes increasingly competitive and profit margins are tightening, manufacturers must create solutions to optimize profit in LDPE high pressure tubular reactors. Thus, in this study, the optimization of LDPE was proposed and conducted by improving the design and parameter adjustment of the LDPE tubular reactor. A mathematical model was developed and validated with industrial data by using MATLAB R2021. Input parameter study was carried out to investigate the effect of initiator concentration (CI), monomer concentration (CM), and solvent concentration (CS) to the ethylene conversion rate, reaction temperature rate, and final product grade, respectively. The CM was identified as the most significant parameter to influence LDPE polymerization process. CM increment results in the highest reaction temperature peak, which was originating from 249.58 to 299.21oC. The highest MFI value was also obtained when the CM was increased from 0.01954 to 0.01979 mol/cm³. Then, a comparative study between design and parameter adjustment for profit maximization in LDPE High Pressure Tubular Reactor was conducted. With the profit of RM166.83 million/year, compared to RM106.83 million/year, double reaction zones demonstrates that it has much better ethylene conversion rate compared to single reaction zone with optimization.

Keywords:

Low density Polyethylene;
Polymerization; Tubular Reactor;
Modeling

1. Introduction

Low Density Polyethylene or known as LDPE is a very essential commodity polymer with a lot of industrial applications. LDPE is known for its exceptional electrical and impacts qualities, as well as its chemical and moisture resistance. LDPE is widely utilised in the production of plastic containers, computer hardware packaging, toys, tubing, and processing equipment around the world. Despite this, the most prevalent application is in plastic bags. LDPE consumption has been claimed to be on

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the rise around the world due to its unique qualities [1]. With a compound annual growth rate (CAGR) of 19.4 %, the global low-density polyethylene market is expected to grow from \$56.6 billion in 2020 to \$67.56 billion in 2021 [2]. The increase is primarily due to companies reorganising their operations and recovering from the impact of COVID-19, which had previously resulted in restrictive containment measures such as social distancing, remote working, and the closure of commercial activities, all of which created operational challenges. At a CAGR of 3%, the market is estimated to reach \$76.05 billion in 2025 [2].

Tubular reactors are commonly used in LDPE industry to generate branching low density polyethylene. It is carried out in an exothermic polymerization process with pressure ranges from 1300 to 3000 atm, while the temperature climbs from 50°C to 330°C. According to Agrawal *et al.*, [3], a typical commercial reactor has multiple reactions, heating, and cooling zones, as well as intermediate additions of initiators, monomers, and solvents, resulting in significantly different polymerization conditions in each zone. This reactor's single-pass ethylene conversion is reported to be 20-35 percent.

According to Azmi and Aziz [4], a typical high-pressure LDPE tubular reactor is built out of a very long spiral-wrapped jacketed metallic pipe with a total length of 500 to 3,300 metres. The TR is made up of a sequence of straight portions with 180-degree bends connecting them. The inner diameter is between 60 and 80 mm, whereas the outer diameter is between 160 and 180 mm. To withstand the severe working conditions needed in the polymerisation of ethylene, a thick wall layer is applied to the reactor tube. The present industrial high-pressure TR is used on length-to-diameter ratios (L/D) ranging from 1 to 6 tens of thousands of times. There are various reactions, as well as heating and cooling zones, in the TR. The temperature of the reaction mixture is controlled in the pre-heating and cooling zones by cycling steam or liquid water through separate jacket zones [4]. The schematic diagram of an industrial LDPE-TR is shown in Figure 1.

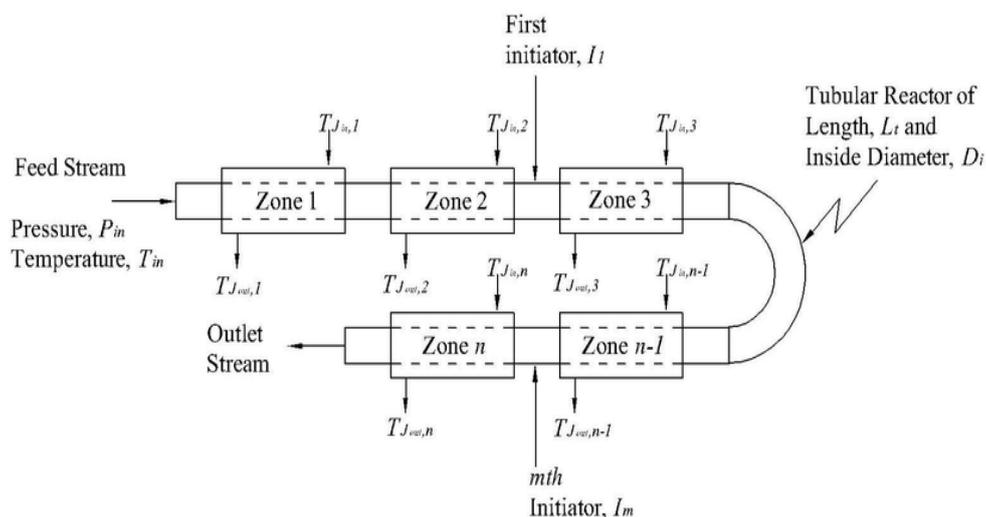


Fig. 1. Schematic diagram of an industrial LDPE tubular reactor [4]

The industry has become more competitive, and market profit margins have shrunk. Therefore, manufacturers must focus on the solutions to maximize the profit in LDPE high pressure tubular reactor to fulfil the market demand. For a process engineer, a mathematical model is an essential tool. It enables the investigation of the effects of various design and operational variables on production and product quality safely and cost-effectively [5,6]. From the past researchers, many works have been carried out. Azmi *et al.*, [4] used the steady-state assumption for parameter

adjustment through a simplified mathematical model. Agrawal *et al.*, [3] conducted optimization study through multi objective optimization on LDPE production. Erdeghem *et al.*, [7] focused on increasing LDPE productivity while lowering investment costs. Buchelli *et al.*, [8] and Fries *et al.*, [9] focused on fouling study inside the reactor wall. Pladis *et al.*, [10] proposed a computational model for predicting the viscoelastic behaviour of LDPE generated in high pressure tubular reactor. Numerous studies have been reported by previous researchers. However, none of the studies report a comparative between design adjustment versus parameter adjustment in their work. For this reason, in this study, a mathematical model will be developed and validated using actual industry data prior to the comparative study.

2. Methodology

2.1 Mathematical Model Development and Data Validation

In this study, a mathematical model was developed and validated with previous work by Azmi *et al.*, [11].

2.1.1 Kinetic mechanisms

The kinetic mechanisms considered in this study is listed in Table 1. Overall, 13 reaction mechanisms were considered in this study.

Peroxide Initiation	$I_k \xrightarrow{f_{ik}, k_{ik}} 2R(0) \quad k = 1, 2$
Monomer Thermal Initiation	$3M \xrightarrow{k_{mi}} R(1) + R(2)$
Propagation	$R(m) + M \xrightarrow{k_p} R(m + 1)$
Thermal Degradation	$R(m) \xrightarrow{k_{tdt}} P(m) + R(0)$
Chain Transfer to Polymer	$R(n) + P(m) \xrightarrow{m k_{trp}} P(n) + R(m)$
Backbiting	$R(m) \xrightarrow{k_{bb}} Rm$
β -scission of Tertiary Radical	$R(m) \xrightarrow{k_{\beta}} P(m) + R(0)$
Oxygen Initiation	$O_2 + M \xrightarrow{k_o} 2R(0)$
Generation of Inert	$O_2 + R(m) \xrightarrow{f_o k_o} X$
Termination by Combination	$R(n) + R(m) \xrightarrow{k_{tc}} P(n + m)$
Chain Transfer to Monomer	$R(m) + M \xrightarrow{k_{trm}} P(m) + R(1)$
Chain Transfer to Transfer Agent	$R(m) + S \xrightarrow{k_{trs}} P(m) + R(0)$
β -scission of Secondary Radical	$R(m) \xrightarrow{k_{\beta_1}} P(m) + R(0)$

Where:

I = Initiator; R = Peroxide; M = Monomer; S = Chain transfer agent; P = Reactor pressure; P(m) = Polymer molecule of chain length m, (m = 1, ..., ∞); R(m) = Living radical of chain length, m; f_{ik} = Initiation efficiency of fictitious initiator k, (k = 1, 2); f_o = Efficiency of inert generation by oxygen

2.1.2 Model assumptions and equation for tubular reactor

The following were the assumptions used in this study:

- i. Assumption of ideal plug flow with no axial mixing.
- ii. The pressure inside the reactor is kept constant throughout the reactor because the pressure drop in each zone is less than 10%, which is negligible [12]
- iii. According to Donati *et al.*, [13] plant studies, there is no pulse valve effect in the model.
- iv. As suggested by Ehrlich and Mortimer [14], the reaction mixture is assumed to be in a homogeneous phase.
- v. According to Chen *et al.*, [15] the heat capacity of a reaction mixture is defined as the sum of the heat capacities of the pure components.
- vi. The model's independent variable was the axial length (z) of the tubular reactor, and the ordinary differential equations (ODE) were integrated along the reactor length.
- vii. Volumetric flow rate and temperature of jacket fluid is kept constant [15,16]
- viii. The reaction mechanism is chosen and listed in Table 1.
- ix. Single feed and initiator injection point where the axial location of feed and initiator at z = 0.0 mm.
- x. Parameters Used in Tubular Reactor
- xi. The model equations used for this study were listed in Table 2.

Table 2
 Model equation for tubular reactor

Description	Equation	
Overall Mass Balance	$\frac{dv}{dz} = -\frac{v}{\rho} \frac{d\rho}{dz}$	(1)
Initiator Balance	$V \frac{dC_{I_m}}{dz} = \left(-2fK_d C_{I_m} - C_{I_m} \frac{dv}{dz} \right)$	(2)
	$\therefore \text{for } m = 1,2$	
Monomer Balance	$V \frac{dC_M}{dz} = \left(2K_{th} C_M^3 - K_{trm} C_M \lambda_0 - C_M \frac{dv}{dz} \right)$	(3)
Solvent Balance	$V \frac{dC_S}{dz} = \left(-K_{trs} C_S \lambda_0 - C_S \frac{dv}{dz} \right)$	(4)

where:

C_{I_m} = concentration of initiator, mol/l; C_M = monomer concentration, mol/l; C_S = solvent concentration, mol/l; C_p = specific heat of reactant mixture, cal/g·K; D = inside diameter of reactor, cm; I = initiator M monomer; P_l = dead polymer with chain length l; P_k = dead polymer with chain length k; R_{in} = primary initiator radical; R_1 = radical of chain length l; R_k = radical of chain length k; Re = Reynolds number; S = solvent

2.2 Design versus Parameter Adjustment Comparison Study

For the comparison study, design and parameter adjustment of the LDPE tubular reactor was conducted. For parameter adjustment, significant adjustment on parameters along the LDPE High Pressure TR such as concentration of initiator, solvent, monomer, and inlet temperature were adjusted to obtain maximum monomer conversion while satisfying the tolerance for MFI and reactor temperature. Meanwhile, for design adjustment, a comparison between single reaction zone with optimization and double reaction zones were conducted and the profit maximization for both single reaction zone with optimization and double reaction zones were calculated

accordingly. The profit calculations were determined using Eq. (5) [11], whereas the cost of the product price, raw materials, and utility were listed in Table 3.

$$P = (42.89 \times 10^{-3} F_M (X_M/100)) - \left(2.68 \times 10^{-3} CP + \left(9.15 \times 10^{-3} F_M (X_M/100) + 33.66 \times 10^{-3} F_S + 1.01 (F_{I,1} + F_{I,2} + \dots + F_{I,n}) \right) \right) \quad (5)$$

Table 3

Cost of the product price, raw materials, and utility cost

Unit Price	Revenue/ Cost (Million RM/year) assuming a stream factor of 0.9 (i.e. 328.5 days of production per year)	
Product price [17]:		
LDPE 6.40 RM/kg	$Rev = 42.89 \times 10^{-3} F_M (X_M/100)$	(2)
Cost of Raw Materials [17]:		
Ethylene, F_M 4.06 RM/kg	$cost_{eth} = 9.15 \times 10^{-3} F_M (X_M/100)$	(3)
Solvent, F_S 4.27 RM/kg	$cost_S = 33.66 \times 10^{-3} F_S$	(4)
Initiators, F_I 128 RM/kg	$cost_{mit} = 1.01 (F_{I,1} + F_{I,2} + F_{I,3} + F_{I,4})$	(5)
Utility Cost [18]:		
Electricity RM 0.38/kW/h	$cost_{elec} = 2.68 \times 10^{-3} CP$	(6)

3. Results and Discussions

The results of the modelling and optimization are presented and discussed in this chapter. In order to maximize profit in LDPE tubular reactor, a comparison between the developed model and industrial data is carried out. The simulation of the model is solved in MATLAB R2021 and the results are presented in Figure 2 to Figure 5 and Table 4 to Table 8. The tube length of 1200 m, reactor inner diameter of 0.05 m, and constant reactor pressure of 2000 atm are chosen as the reference condition in this simulation.

3.1 Effect of Parameter Adjustments of LDPE Tubular Reactor on Monomer Conversion

In this section, the effect of parameter adjustments of single reaction zone of LDPE TR on several properties such as monomer conversion (XM), Melt Flow Index (MFI) etc. were compared with the industrial data. Inlet temperature (Tin), initiator concentration (CI), monomer concentration (CM), and solvent concentration (CS) across the tubular reactor were selected as manipulative variables.

First, adjustments on initiator concentration, CI were made to compare the changes on monomer conversion and other properties. The industrial data produces 9.6145% monomer conversion (XM) using 3.0180×10^{-6} mol/cm³ of initiator concentration. Then, the value of initiator concentration, CI was increased to 3.0430×10^{-6} mol/cm³ and decreased to 3.01196×10^{-6} mol/cm³ to observe the changes on LDPE's properties especially its monomer conversion while satisfying its MFI and reactor temperature.

For CI = 3.0430×10^{-6} mol/cm³ and CI = 3.01196×10^{-6} mol/cm³, the ethylene conversions were 10.3862% and 9.4545% respectively. Judging from Table 4, both initiator concentration and ethylene conversion values were directly proportional. This is true since higher initiator concentration promotes greater ethylene conversion and therefore increasing the peak temperature. This is due to exothermic nature of ethylene polymerization [4].

Table 4
 Comparison table for adjustment of initiator concentration, CI

	Industrial Data	Increase CI (mol/cm ³)	Decrease CI (mol/cm ³)
Monomer Conversion, XM (%)	9.6145	10.3862	9.4545
MFI (g/10.min)	6.5993	6.9962	6.5072
Reactor Temperature, T (°C)	249.58	257.91	247.86
Initiator concentration, CI (mol/cm ³)	3.0180e-06	3.0430e-06	3.01196e-06

Next, adjustments on value of monomer concentration were made to compare the changes on monomer conversion and other properties. The industrial data gave 9.6145% monomer conversion, XM with 0.01954 mol/cm³ of monomer concentration. Then, the value of monomer concentration, CM was increased to 0.01979 mol/cm³ and decreased to 0.01866 mol/cm³ to observe the changes on LDPE's properties especially its monomer conversion while keeping its MFI and reactor temperature at a safe level. For CM = 0.01979 mol/cm³ and CM = 0.01866 mol/cm³, the ethylene conversions were 14.0513% and 6.0415% respectively.

Judging from Table 5, both monomer concentration and ethylene conversion values were directly proportional. This is true, since higher monomer concentration promotes greater ethylene conversion, whilst lower monomer concentration reduces the ethylene conversions.

Table 5
 Comparison table for adjustment of monomer concentration, CM

	Industrial Data	Increase CM (mol/cm ³)	Decrease CM (mol/cm ³)
Monomer Conversion, XM (%)	9.6145	14.0513	6.0415
MFI (g/10.min)	6.5993	9.7709	5.0134
Reactor Temperature, T (°C)	249.58	299.21	208.81
Monomer concentration, CM (mol/cm ³)	0.01954	0.01979	0.01866

Adjustments on value of solvent concentration were also made to compare the changes on monomer conversion and other properties. The industrial data gave 9.6145% monomer conversion, XM with 1.4477e-04 mol/cm³ of solvent concentration. Then, the value of solvent concentration, CS was increased to 1.4868e-04 mol/cm³ and decreased to 1.3015e-04 mol/cm³ to observe the changes on LDPE's properties especially its monomer conversion while satisfying its MFI and reactor temperature. However, only small amount of solvent concentration, CS can be adjusted since it will greatly effect on melt flow index. For CS = 1.4868e-04 mol/cm³ and CS = 1.3015e-04 mol/cm³, the ethylene conversions were 9.6146 % and 9.6159 % respectively.

Table 6
 Comparison table for adjustment of solvent concentration, CS

	Industrial Data	Increase CS (mol/cm ³)	Decrease CS (mol/cm ³)
Monomer Conversion, XM (%)	9.6145	9.6146	9.6157
MFI (g/10.min)	6.5993	6.9245	5.4841
Reactor Temperature, T (°C)	249.58	249.58	249.59
Solvent concentration, CS (mol/cm ³)	1.4477e-04	1.4868e-04	1.3015e-04

From Table 6, an increase to the solvent concentration increases the MFI and vice versa. As mentioned in Grau *et al.*, [19] research, in radical polymerization, solvent impact is usually negligible, however in the case of ethylene polymerization, solvent appears to have a significant role, particularly to the polymer's MFI.

3.2 Effect of Design Adjustments of LDPE Tubular Reactor on Monomer Conversion

In this section, single reaction zone with optimization and double reaction zones were compared for the design adjustments. Figure 2 and Figure 3 indicates only 1 reaction zone involve in a single reaction zone with optimization. In this reaction zone, an increment of temperature is observed due to the highly exothermic rise of the polymerization reaction which generates a free radical reaction between the feed in the initiator and the monomer. Monomer conversion (XM) also indicates a fast escalation as shown in Figure 3.

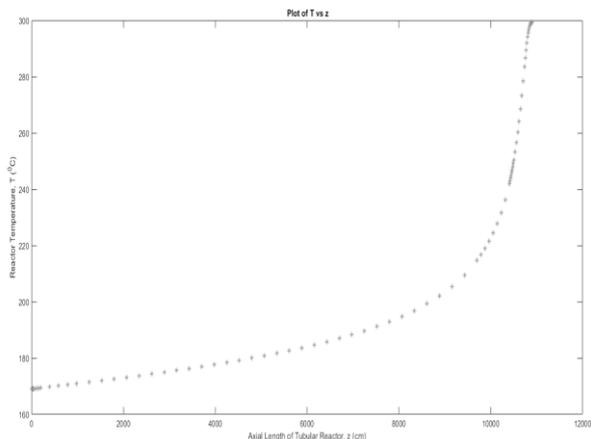


Fig. 2. Reactor temperature, t profile of industrial data for single reaction zone with optimization

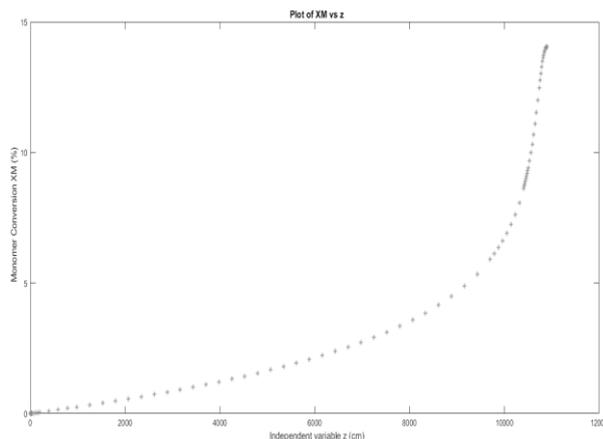


Fig. 3. Reactor monomer conversion, XM profile of industrial data for single reaction zone with optimization

Figure 4 and Figure 5 depicts the temperature and monomer conversion profile of the LDPE tubular reactor for two reaction zones, which reveals that there are four zones in total, 2 reactions and 2 cooling zones. The extremely exothermic rise of the polymerization reaction, which generates a free radical interaction between the feed in the initiator and the monomer, causes a rapid temperature peak in the reaction zones. Monomer conversion (XM) also shows a rapid escalation, and this temperature peak shows that practically all the initiator has been used. XM remains constant until another initiator is injected in the next reaction zone since no reaction has occurred.

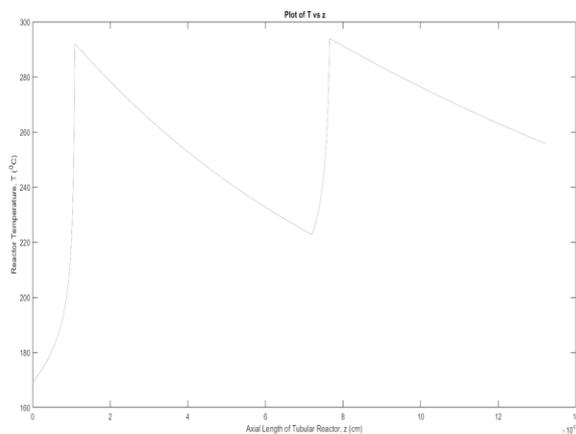


Fig. 4. Reactor temperature, T profile of industrial data for double reaction zones

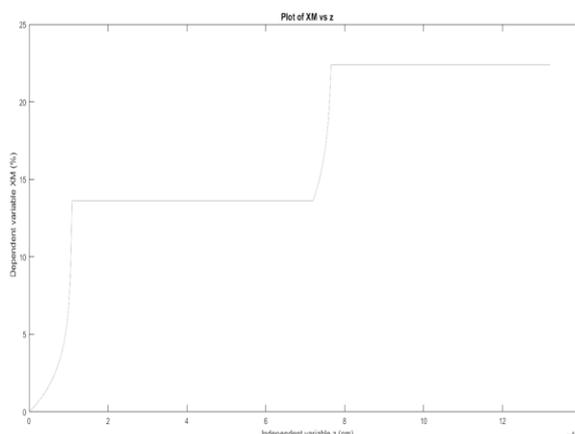


Fig. 5. Reactor monomer conversion, XM profile of industrial data for double reaction zones

In the second zone, the reaction mixture is cooled down to the optimal temperature level for the half-life decomposition of the second initiator. The efficiency of initiator disassociation is largely reliant on temperature, and if the temperature surpasses its half-life temperature, it will be ineffective [20]. Because of this inefficiency, the initiation reaction and propagation rate will be slowed, lowering the monomer conversion value.

Using a counter-current heat exchanger, the temperature of the reaction mixture is steadily lowered in the cooling zone. The goal of this constant cooling procedure is to slow the creation of a polymer-rich layer on the tubular reactor's inner wall.

By referring to the Table 7, the monomer conversion for double reaction zones was higher than single reaction zone with optimization with monomer conversions of 22.3972% and 14.0153% respectively. However, the reactor temperature for double reaction zones was lower than single reaction zone with optimization with reactor temperature of 299.21°C and 255.82°C respectively. The results demonstrates that by adding the number of reaction zones greatly improved the monomer conversion rate. Despite, the increment of reactor temperature needs to be alerted as to avoid fluctuation in the polymerization process. In the worst-case situation, if the temperature level exceeds 300°C, the radicals will be ruined, and no free-radical polymerization will occur [21].

Table 7

Comparison table for single reaction zone with optimization and double reaction zones

	Single Reaction Zone with Optimization	Double Reaction Zones
Monomer Conversion, XM (%)	14.0513	22.3972
MFI (g/10.min)	9.7709	5.3277
Reactor Temperature, T (°C)	299.21	255.8172
Inlet Temperature, Tin (°C)	169.0033	169.0033
First Initiator concentration, CI1 (mol/cm ³)	3.0180e-06	3.0978e-06
Second Initiator concentration, CI2 (mol/cm ³)	-	1.92980e-06
Monomer concentration, CM (mol/cm ³)	0.01979	0.01954
Solvent concentration, CS (mol/cm ³)	1.4477e-04	1.4477e-04

3.3 Profit Maximization Calculation for Single Reaction Zone with Optimization and Double Reaction Zone

In this section, profit, revenue, cost of material, and cost of electrical used were calculated for Single Reaction Zone with Optimization and Double Reaction Zones through the development of the mathematical model in MATLAB. The results were tabulated and summarized in the Table 8 below. Furthermore, only utility and material costs were calculated. The cost for additional equipment such as reactor was not included, for simplification purpose.

Table 8

Profit maximization for single reaction zone with optimization and double reaction zones

	Single Reaction Zone with Optimization	Double Reaction Zones
Monomer Conversion, XM (%)	14.0513	22.3972
MFI(g/10min)	9.7709	5.3277
Revenue (million) RM/year	195.02	306.86
Material Cost (million RM/year)	80.37	126.47
Electric Cost (million RM/year)	8.57	13.56
Profit (million RM/year)	106.08	166.83

From Table 8, even though the material and electric costs for Double Reaction Zones were higher than the costs for Single Reaction Zone with Optimization, it gave higher revenue and profit for

Double Reaction Zones than the Single Reaction Zone with Optimization. The results proved that with the addition of reaction zones, it greatly improved the ethylene conversions rate, revenue, and profit of the LDPE High Pressure Tubular Reactor.

4. Conclusion

From the input parameters effect studies, initiator concentration (CI), monomer concentration (CM), and solvent concentration (CS) influence the ethylene conversion rate, reaction temperature rate, and final product grade. The highest monomer conversion rate, at 14.0513 was observed when the CM was increased from 0.01954 to 0.01979 mol/cm³. This CM increment also results in the highest reaction temperature peak, which was originating from 249.58 to 299.21°C. The highest MFI value was also obtained when the CM was increased from 0.01954 to 0.01979 mol/cm³. The lowest monomer conversion, reaction temperature peak and MFI were obtained when the CM was decreased from 0.01954 to 0.01866 mol/cm³. The CM was identified as the most significant parameter to influence LDPE polymerization process. For design adjustment, comparison between single reaction zone with optimization and double reaction zones were conducted. With the profit of RM166.83 million/year, compared to RM106.83 million/year, double reaction zones demonstrates that it has much better ethylene conversion rate compared to single reaction zone with optimization. It was proven that adjusting the design of LDPE tubular reactor by adding extra reaction and cooling zone improved monomer conversion, thus increasing the annual profit, even though it requires much expensive raw materials and electricity costs. Higher revenue in double reaction zones counteract the much higher raw materials and electricity costs. For future research, it is recommended to calculate material construction costs and need to be incorporated into MATLAB since it was not included in this study.

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