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Design of A PFR For the Production of 1,000,000 Tons Per Year of Ethyl Acetate from Esterification of Acetic Acid and Ethyl Alcohol

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Abstract: The research considered the design or size of a plug flow reactor for the production of 1,000,000 tons of ethyl acetate per year from the esterification of acetic acid and ethyl alcohol in the presence of an acid catalyst. The PFR design models for volume, height, diameter, space time, space velocity, quantity of heat generated, quantity of heat generated per unit volume of the reactor as well as the temperature effect models were developed using the conservation principle of mass and energy at steady state operation of the reactor. The developed models were simulated using MATLAB at initial feed and operating temperature of 299.830k and 343.15k respectively and fractional conversion variation between 0 to 0.95 at an interval of 0.05. At a maximum conversion of 0.95, the PFR size specification for volume, height, diameter, space time, space velocity, quantity of heat generated and the quantity of heat generated per unit volume of the reactor was 15.2774m^3, 4.2691m, 2.1346m, 2.9956sec., 0.3338sec^{-1}, 12821.5800j/s and 839.2536j/sm^3 respectively. The effect of the operating parameters on the performance or functional parameters of the reactor are presented in profiles as shown in figure 2 to 12 and the profile behaviour or trend were in agreement with process behaviour of PFR steady state operation in various literatures. The research have shown that in order to ensure sustainability and continuous production of ethyl acetate to meet the global demand of the economic and viable product, the plug flow reactor have demonstrated a good performance characteristics as a reacting media for the esterification process especially in the energy efficiency of the process as well as the product yield.

Keywords: Esterification, ethyl acetate, plug flow reactor, design, MATLAB Simulation

I. Introduction

Ethyl acetate is an organic solvent and a chemical intermediate with the molecular formula $C_4H_8O_2$ and molecular weight of 88.10g/mol. It is a colorless liquid with a pungent smell and a non-toxic, non-hygroscopic volatile liquid and occurs moderately as a polar solvent (Bijay & Hiren, 2011). It has a wide range of industrial applications such as production of pharmaceuticals, solvents, inks, plastics, synthetic fruits and chemicals that can be applied domestically and industrially (Grodowska & Parzewki, 2010; Karan, 2017; Nagamalleswara, 2015). Traditionally, ethyl acetate production is via catalytic esterification of acetic acid and ethyl alcohol (Calvar *et al*, 2007; Evelien *et al*, 2014; Sykes, 1986; Hasanoglu *et al*, 2009). Chemical reactions usually occur in chemical reactors and esterification reaction can occur in reactors like the plug flow reactor (Ni & Meunier, 2007), continuous stirred tank reactor (Tang *et al*, 2016), Microwave reactors (Umrigar *et al*, 2022; Baraka *et al*, 2023), Membrane reactors (Ghahremani *et al*, 2021) and fixed bed reactors (Son *et al*, 2023). In this article, the plug flow reactor (PFR) also known as the tubular reactor (TR) or the packed bed reactor (PBR) is considered as the reacting media for the esterification reaction. The PFR is characterized with uniform composition, no mixing or back flow and the reactant species moves along at the same speed, unvarying product quality, cheap maintenance, energy efficiency, high conversion rate and suitable for large capacity processes (Fogler, 2006). Generally, the design of chemical engineering equipment involves the application of the fundamental principle of mass and energy for the reactor size determination (Wosu & Ezeh, 2024; Ojong *et al*, 2024; Wosu *et al*, 2024; Wosu *et al*, 2023).

The importance and applications of ethyl acetate have prompted several research in the field and thus; Hilmioglu (2022) stated that ethyl acetate is produced in batch reactor using sulfo succinic acid as a catalyst and highlighted the advantage of this technology the processes that obey the Le Chatelier principle which causes a shift or forward esterification reaction as a result of the excess reactant involved in the process. Ding *et al* (2012) and Nurhayati *et al* (2017) also stated that sulphuric acid can be utilized as catalyst during esterification process for ethyl acetate production while Ikhazuangbe & Oni in 2015 stated that ethyl acetate can also undergo hydrolysis in the presence of sodium hydroxide and the experiment showed that the reaction rate depends on the feed concentration while the rate constant is a function of the reaction time. Abdulaziz *et al* (2023) utilized the Aspen HYSYS as a tool for sensitivity analysis of flow reactor performances during esterification process. Modern research have shown that ethyl acetate can be a substitute for fuel (green premium) for vehicles and power generators (Heuser et al, 2019). This article delves into the design of a PFR for ethyl acetate production as a tool to enhance sustainability and effective production to meet the ever growing global demand of the economic and viable product.

II. Materials and Methods

2.1 Materials

The materials utilized in the research are computer set, data obtained from journals, textbooks and the simulation tool used is MATLAB.

2.2 Methods

The methodology adopted in this research is quantitative and the data used were obtained from thermodynamic properties of the reactant species and products, literature data, and calculated/derived data and the following procedures were sequentially adopted;

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2.2.1 Development of the Reaction Kinetic Models

The Kinetic model of the esterification reaction is obtained from the reaction chemistry of the process in equation (1)

$$\text{Acetic acid + Ethyl alcohol} \xrightarrow{k_1} \text{Ethyl acetate + Water} \quad (1)$$

Equation (1) can be expressed molecularly as;

$$CH_3COOH + CH_3OH \xrightarrow{K_1} CH_3COOCH_3 + H_2O \quad (2)$$

Symbolically, equation (2) can be expressed as;

$$A + B \xrightarrow{k_1} C + D \quad (3)$$

where A represents acetic acid, B is ethyl alcohol, C is ethyl acetate, D is water and k_1 represents kinetic rate constant which is an indication that the reaction process is temperature dependent and the process condition is exothermal. The depleting rate of the reactant species is related to the rate constant, fractional conversion, initial concentration of the limiting reactant, temperature, activation energy as shown in equation (4)

$$-r_i = k_o e^{-E/RT} C_{io}(1 - x_i) \quad (4)$$

2.2.2 Development of the PFR Design/Sizing Model

Consider the schematic representation of a plug flow reactor with mass and heat effect

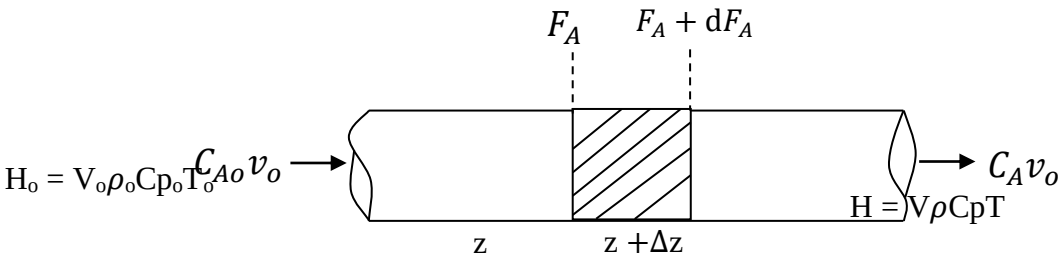

Figure 1: PFR Schematic with Mass and Heat Effect

The design and temperature effect model of the PFR is developed the application of the conservation principle of mass and energy thus;

$$\begin{bmatrix}\text{Rate of Accumulation of Material Within the Volume}\end{bmatrix} = \begin{bmatrix}\text{Rate of input of feed into the Volume}\end{bmatrix} - \begin{bmatrix}\text{Rate of Outflow of feed from the Volume}\end{bmatrix} - \begin{bmatrix}\text{Rate of } \textit{depletion of} \text{ feed due to chemical reaction}\end{bmatrix} \quad (4)$$

The terms in equation (4) can be defined, substituted and simplified at steady state to yield the PFR performance model for volume, height, diameter, space time, space velocity, quantity of heat generated as well as the quantity of heat generated per unit volume of the reactor thus;

$$V_R = v_o \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RT} C_{io}(1-x_i)} \quad (5)$$

$$H_R = \left[\frac{25V_o}{\pi} \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RT} C_{io}(1-x_i)}\right]^{\frac{1}{3}} \quad (6)$$

$$D_R = \frac{\left[\frac{25V_o}{\pi} \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RT} C_{io}(1-x_i)}\right]^{\frac{1}{3}}}{25} \quad (7)$$

$$\tau_{PFR} = \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RT} C_{io}(1-x_i} \quad (8)$$

$$S_V = \frac{1}{\int_0^{x_A} \frac{dx_A}{k_o e^{-E/RT} C_{io}(1-x_i)}} \quad (9)$$

$$Q = \Delta H_R F_{Ao} x_A \quad (10)$$

$$q = \frac{\Delta H_R F_{Ao} x_A}{V_R} \quad (11)$$

The temperature effect model of the PFR in Figure 1 developed using the conservation principle of energy balance in equation (12)

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$$\begin{bmatrix}\text{Rate of inflow of energy into the differential volume of reactor}\end{bmatrix} = \begin{bmatrix}\text{Rate of outflow of energy from the differrential volume of reactor}\end{bmatrix} - \begin{bmatrix}\text{Rate of depletion of energy within the differential volume of reactor}\end{bmatrix} + \begin{bmatrix}\text{Rate of energy added into the differential volume of reactor}\end{bmatrix} \quad (12)$$

The terms in equation (12) can be defined, substituted and simplified to yield the temperature effect model thus;

$$\frac{dT}{dZ} = \frac{1}{u\rho C_p}(\Delta H_R)(-r_i) \quad (13)$$

Data for Evaluation

The data for evaluation in this research are the properties/thermodynamic data and data obtained from literatures as presented in table 1 and 2 respectively.

Table 1: Properties/ Thermodynamic Data

Data/Parameter	Values	Description
ρ_A	1050Kg/m^3	Density of acetic acid
ρ_B	789Kg/m^3	Density of ethyl alcohol
ρ_C	902Kg/m^3	Density of ethyl acetate
ρ_D	997Kg/m^3	Density of water
P_o	101325Kg/m^3	Initial pressure
R	8314Nmmol^{-1}K^{-1}	Gas constant

Table 2 Data Obtained from Literature

Data	Values	Description	References
T	343.15K	Operating temperature of the reactor	Nagamalleswara*et al*., 2015
r_i	5.28×10^7mol/m^3	Reaction rate	Nagamalleswara et al, 2015
E	59.403kJ/mol	Activation energy	Nagamalleswara et al, 2015

III. Results and Discussion

The results and discussion of the PFR design for the production of ethyl acetate from the esterification of acetic acid and ethyl alcohol is presented in Table 3 and Figure 2 to 10.

Table 4: Design Results showing Fractional Conversion, Temperature, Reactor Volume, Height, Diameter, Space Time, Space Velocity, Quantity of Heat Generated and the Quantity of Heat Generated per unit Volume of the Reactor

X_A	T(K)	V_R(m3)	H_R (m)	DR(m)	τ(s)	S_V (S^{-1})	Q(J/s)	q(J/sm3)
0.05	299.830	0.042	0.599	0.299	0.008	120.512	674.820	15945.818
0.15	299.830	0.142	0.897	0.449	0.028	35.942	2024.460	14267.311
0.25	299.830	0.268	1.109	0.555	0.053	19.028	3374.100	12588.804
0.35	299.830	0.433	1.302	0.651	0.085	11.779	4723.740	10910.297
0.45	299.830	0.658	1.496	0.748	0.129	7.752	6073.380	9231.789
0.55	299.830	0.983	1.711	0.855	0.193	5.190	7423.020	7553.282
0.65	299.830	1.493	1.967	0.983	0.293	3.415	8772.660	5874.775
0.75	299.830	2.412	2.307	1.154	0.473	2.114	10122.300	4196.268
0.85	299.830	4.556	2.852	1.426	0.893	1.119	11471.940	2517.761
0.95	299.830	15.277	4.269	2.135	2.996	0.334	12821.580	839.253

Table 4 shows the design result of the PFR for esterification process. Here, the MATLAB simulation was performed at initial feed and operating temperature of 299.830k and 343.15k and varying fractional conversion from 0 to 0.95 at an interval of 0.05 and the process behaviour showed that the reactor volume, height, diameter, space time and quantity of heat generated increases with an increase in the fractional conversion while the space velocity and the quantity of heat generated per unit volume of the reactor

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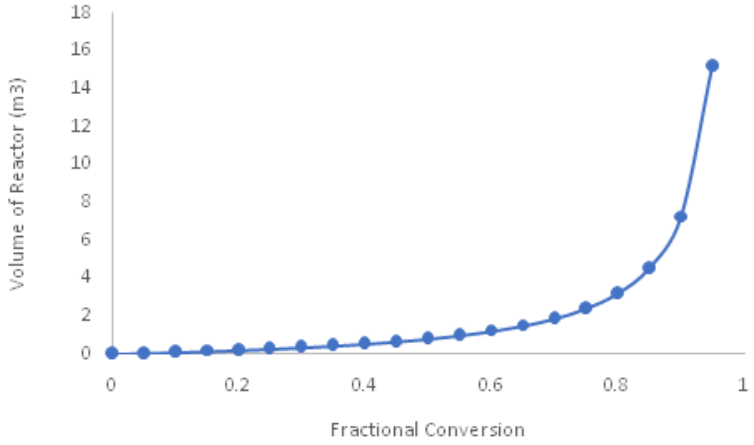
decreases as the fractional conversion increases. At a maximum conversion of 0.95, the reactor volume, height, diameter, space time, space velocity, quantity of heat generated and the quantity of heat generated per unit volume of the reactor are 15.277m^3, 4.269m, 2.135m, 2.996seconds 0.334sec^{-1}, 12821.580j/s, 839.254j/sm^3 respectively.

3.1 Results of PFR Design Parameters Process Behaviour

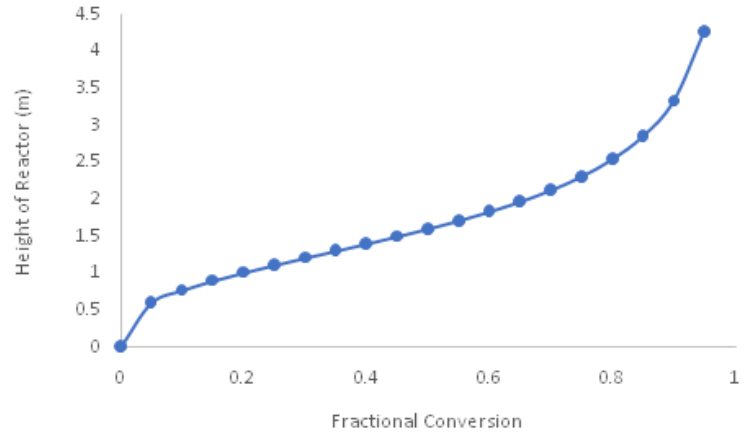
The effect or variation of fractional conversion and temperature on the performance parameters of the PFR is presented below.



Profile of PFR Volume (V_R) and Fractional Conversion (X_A)

Figure 2: Profile of PFR Volume (V_R) and Fractional Conversion (X_A)

Figure 2 is a profile or relationship between the PFR volume and fractional conversion during esterification process obtained from the MATLAB simulation. The result showed that at initial feed and operating temperature of 299.830k and 343.15k respectively with changes in fractional conversion from 0 to 0.95 at an interval of 0.05, the reactor volume increases exponentially as the fractional conversion increases. At a maximum conversion of 0.95, the PFR maximum volume for yearly production of ethyl acetate was 15.2774m^3. The exponential relationship between the PFR volume and fractional conversion is dependent on the reaction kinetic scheme of the esterification process, mass transfer effect, design assumptions made, the effect of temperature, pressure and concentration of the reactant species.



Profile of PFR Height (H_R) and Fractional Conversion (X_A)

Figure 3: Profile of PFR Height (H_R) and Fractional Conversion (X_A)

Figure 3 is a relationship between the PFR height and fractional conversion during the esterification process for ethyl acetate production. According to the plot, an initial linear increase of the reactor height was observed at fractional conversion change from 0 to 0.05, this increase became exponential at fractional conversion above this range. At maximum conversion of 0.95, the maximum height of the reactor at maximum volume was 4.2691m, the profile behaviour between the PFR height and fractional conversion as obtained from the MATLAB simulation depends on certain factors such as the reaction kinetic scheme or rate law of the esterification process, mass transfer effect,design assumptions and effect of temperature, concentration and pressure of the reactant species.

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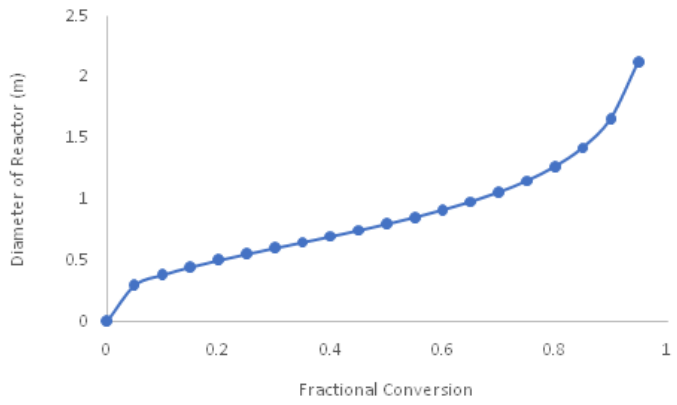
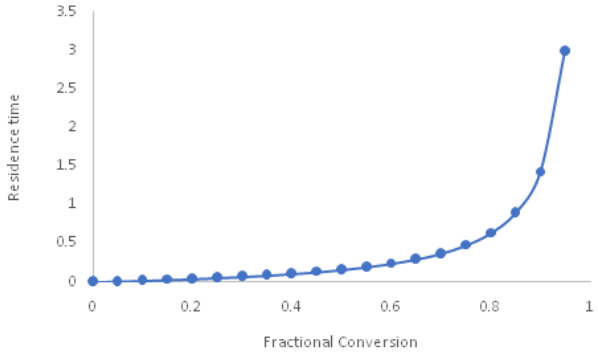


Profile of PFR Diameter (D_R) and Fractional Conversion (X_A)

Figure 4: Profile of PFR Diameter (D_R) and Fractional Conversion (X_A)

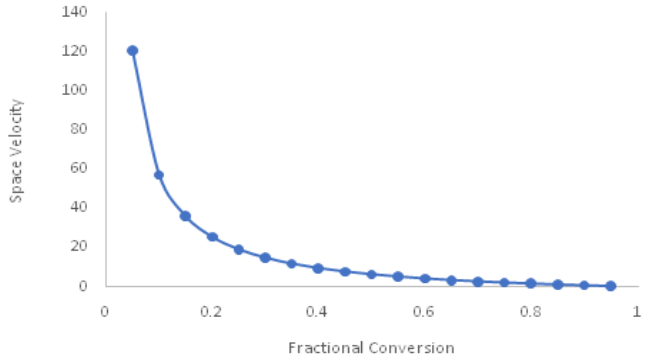
Figure 4 is the behavioural relationship between the PFR diameter and fractional conversion gotten from the MATLAB simulation of the esterification process for ethyl acetate production. Just like the PFR height, an initial linear increase of the reactor diameter was observed at fractional conversion change from 0 to 0.05, this change became exponential at higher conversion above this range. At a maximum conversion of 0.95, the PFR diameter was 2.1346m. it is important to note that this profile behaviour is influenced by certain factors such as the rate law of the esterification reaction, mass transfer effect, feed parameters and design assumptions made during the process.



Profile of PFR Space Time (τ) and Fractional Conversion (X_A)

Figure 5: Profile of PFR Space Time (τ) and Fractional Conversion (X_A)

Figure 5 is the profile relationship of the PFR space time also known as the residence time and fractional conversion during the esterification process for ethyl acetate production. The MATLAB simulation profile showed that there is an exponential increase in space time as the fractional conversion increases. At a maximum conversion of 0.95, the space time recorded was 2.9956sec. this is the maximum time spent by the reactant species in the reactor during the esterification process and this time is greatly influenced by the rate constant or the pre-exponential factor of the reaction kinetic scheme.



Profile of Space Velocity (S_V) and Fractional Conversion (X_A)

Figure 6: Profile of PFR Space Velocity (S_V) and Fractional Conversion (X_A)

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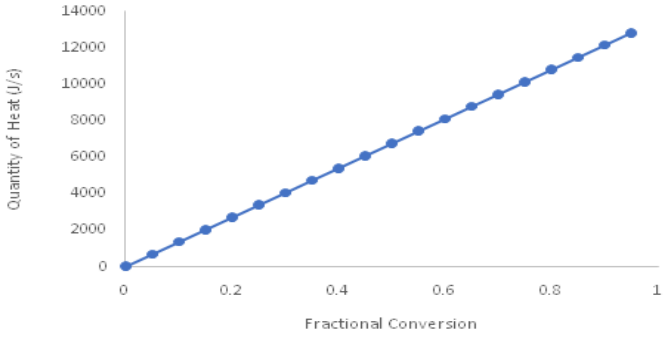
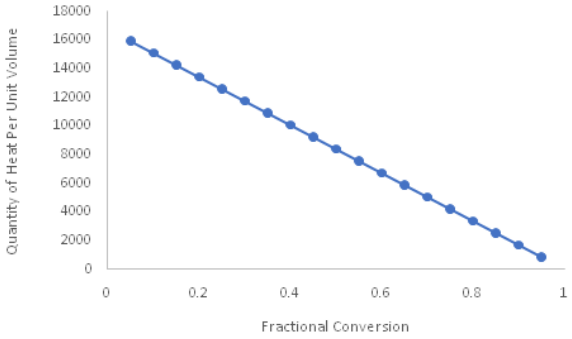
Figure 6 is the MATLAB simulation result showing the profile behaviour of PFR space velocity and fractional conversion during esterification process for ethyl acetate production. The space velocity is mathematically defined as the reciprocal of space time and this mathematical relationship is demonstrated in the profile as the space velocity exhibited an exponential decrease as the fractional conversion increases. At maximum fractional conversion of 0.95, the space velocity recorded was 0.3338sec^{-1}. This profile behaviour is greatly influenced by the rate constant or frequency factor from the esterification reaction kinetics.



Profile of the PFR Quantity of Heat Generated (Q) and Fractional Conversion (X_A)

Figure 7: Profile of the PFR Quantity of Heat Generated (Q) and Fractional Conversion (X_A)

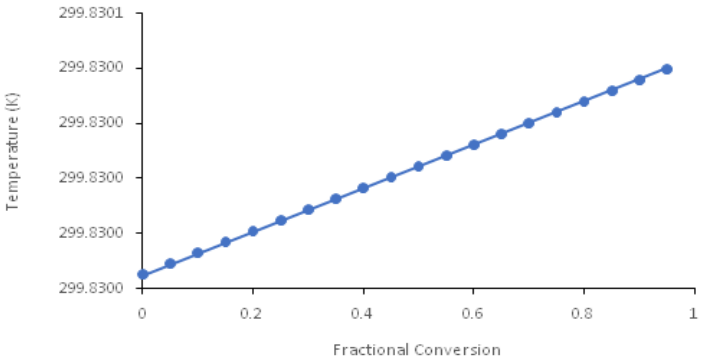
Figure 7 shows that there is a linear increase relationship of the quantity of heat generated as the fractional conversion increases during the esterification process in the PFR. The relationship simply means that the quantity of heat generated during the reaction process is directly proportional to the fractional conversion of the reactant species. At a maximum conversion of 0.95, the quantity of heat generated was 12821.5800j/s. This relationship was as a result of the effect of reaction rate, feed rate, heat transfer and the process operating condition.



Profile of the Quantity of Heat Generated per unit Volume of the PFR (q) and Fractional Conversion (X_A)

Figure 8: Profile of the Quantity of Heat Generated per unit Volume of the PFR (q) and Fractional Conversion (X_A)

Figure 8 shows that the quantity of heat generated per unit volume of the PFR decreases linearly as the fractional conversion increases during the esterification process for ethyl acetate production. According to the plot, at a maximum fractional conversion of 0.95, the quantity of heat generated per unit volume of the PFR decreases to 839.2536j/sm^3. This profile behaviour is greatly influenced by heat transfer effect and operating condition of the process.



Profile of PFR Temperature (T) and the Fractional Conversion (X_A)

Figure 9: Profile of PFR Temperature (T) and the Fractional Conversion (X_A)

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Figure 9 that at initial feed and operating temperature of 299.830k and 343.15k respectively with varying fractional conversion from 0 to 0.95 at an interval of 0.05, the PFR operates effectively during the esterification reaction for ethyl acetate production. This is an indication that when the initial feed and operating temperature is within its specific range for esterification reaction, the process will be greatly favoured and this will enhance optimum yield of the target product as well as the energy efficiency of the process.

IV. Conclusion

1,000,000 tons of ethyl acetate was produced from the esterification of acetic acid and ethyl alcohol in a Plug flow reactor (PFR). The reaction media (PFR) was designed by applying the conservation principle of mass and energy over the reactor during the steady state process. The performance model of the PFR mass and energy balance were simulated using MATLAB at initial feed and operating temperature of 299.830k and 343.15k varying fractional conversion from 0 to 0.95 at 0.05 intervals to give the reactor functional parameters. At a maximum conversion of 0.95, the PFR design or size specification for volume, height, diameter, space time, space velocity, quantity of heat generated and the quantity of heat generated per unit volume of the reactor was 15.2774m^3, 4.2691m, 2.1346m, 2.9956sec., 0.3338sec^{-1}, 12821.5800j/s and 839.2536j/sm^3 respectively. The resultant effect of the fractional conversion and temperature on the PFR functional parameters were presented in figure 2 to figure 9. The researched showed that the PFR is a suitable reaction media for esterification process and its design will enhance the optimum production of ethyl acetate and its sustainability to meet its global demand.

Nomenclature

Symbol	Definition	Unit
ΔH_R	Change in enthalpy of reactants	J/mol
A	Acetic acid	-
B	Ethyl alcohol	-
C	Ethyl acetate	-
C_i	Initial concentration of species	mol/m^3
Cp	Specific heat capacity	J/mol
D	Process water	-
D_R	Diameter of the reactor	M
E	Activation Energy	J/mol
F_A	Initial molar flow rate	mol/S
H_i	Enthalpy of species	J/mol
H_R	Height of the Reactor	M
K_o	Pre-exponential factor	S^{-1}
Q	Quantity of Heat generated	J/S
Q	Quantity of heat generated per reactor volume	J/Sm^3
R	Gas constant	$Nmmol^{-1}k^{-1}$
r_A	Reaction rate of species	$mol/m^3/s$
S_V	Space velocity	Sec^{-1}
T	Operating Temperature	Kelvin
T_c	Temperature of coolant	K
T_o	Initial or fed temperature	K
UAc	Heat transfer coefficient	Kg/m^2SK
V_i	Fractional conversion	Dimensionless
V_o	Volumetric flow rate	m^3/S
V_R	Volume of the Reactor	m^3
ρ_i	Density of species	Kg/m^3
τ	Space time	Seconds

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