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Design Assessment of Flow Reactors for the Production of 1,000,000 tons per year of Ethyl Acetate from Esterification Reaction of Acetic Acid and Ethyl Alcohol

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Abstract The economic importance of ethyl acetate as a feed material utilized in industries for the production of plastics, solvents, rubber, inks and other useful chemicals utilized domestically and industrially necessitated this research as it considered the design assessment of flow reactors (Continuous Stirred Tank Reactor and Plug Flow Reactor) for the production of 1,000,000 tons per year of ethyl acetate from the esterification of acetic acid and ethyl alcohol in the presence of an acid catalyst. The flow reactors design models were developed from the first principle of mass and energy balance and the steady state performance models were simulated in MATLAB at same initial feed and operating temperature of 298.15 k and 343.15 k with fractional conversion range of $X_A \geq 0 \leq 0.95$. The design assessment of the flow reactors performance was based on the yield of ethyl acetate and energy efficiency of the process. At maximum conversion of 0.95, the CSTR and PFR volume was 80.880 m^3 and 15.277 m^3 respectively with a percentage difference of 34.1 % while the quantity of heat generated per unit volume of the CSTR and PFR was 158.526 j/sm^3 and 839.254 j/sm^3 respectively with a percentage difference of 34.1 %. The comparative analysis of the design results showed that more yield of ethyl acetate is produced in the CSTR as shown in the reactor volume while the PFR performed better in terms of energy efficiency and conservation as shown in the quantity of heat generated. The study showed that both reactors (CSTR and PFR) are suitable reaction media for esterification process but the choice of reactor depends on the designer's need.

Keywords: Ethyl Acetate, CSTR, PFR, Design, MATLAB Simulation

I. Introduction

Ethyl acetate is an organic solvent and one of the simplest carboxylate esters. It is a colorless liquid with a pungent smell [1] and starting or feed material used in process industries for the production of plastics, solvents, inks, plasticizers, rubber and chemicals that can be applied domestically and industrially [2,3,4]. Commercially, ethyl acetate is produced from the catalytic esterification of acetic acid and ethyl alcohol [5,6,7,*8*]. Esterification reaction usually occur in flow reactors such as continuous stirred tank reactor and plug flow reactors [9,10,11] microwave and membrane reactors [12,13,14] and fixed bed reactors [15]. In this research article, the comparative analysis or assessment of flow reactors (CSTR and PFR) design for ethyl acetate production from the esterification of acetic acid and ethyl alcohol is considered. The performance model of the flow reactors just like other chemical engineering equipment were developed from the first principle of mass and energy conservation [16,17,18,19,20,21]. The developed models were simulated using MATLAB at same initial feed and operating condition and the design results of the flow reactors were analysed and compared on the basis of ethyl acetate yield and energy efficiency of the processes. The global demand of ethyl

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acetate has necessitated several research on its production and applications in a bid to ensure sustainability and thus; [22,23] researched on the production of ethyl acetate using H_2SO_4 acid as a catalyst for the esterification process. [24], used sulfo succinic acid catalyst in batch manufacture of ethyl acetate and stated the advantage of this technology over other technologies that involves forward or shift reaction due the Le Chatelier principle. [25], researched on the determination of reaction rate and rate constant from the hydrolysis of ethyl acetate using NaOH as a catalyst and stated that the reaction rate depends on feed concentration while the rate constant is dependent on the residence time during the process. Research has shown that ethyl acetate is a vital source of fuel for power and electric generation [26]. This article becomes highly imperative as it seeks to analyze and compare the performance of the CSTR and PFR for effective production of ethyl acetate.

II. Materials and Methods

A. Materials

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

B. Methods

The methodology adopted in this research is quantitative and qualitative or analytical, 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

i. 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)

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

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;

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

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)

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

ii. Development of the CSTR and PFR Design/Sizing Models

The design or performance model of the flow reactors is gotten from the application of mass and energy balance over the reactors. Consider the schematic representation of a continuous stirred tank reactor with feed stream, product stream and heat effect as shown in Figure 1

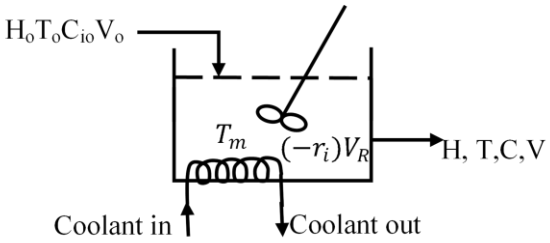

Figure 1: CSTR with Feed Stream, Product Stream and Heat Effect

The mass and energy balance models for the CSTR design were developed using the following assumptions.

i. The feed assumes a uniform composition throughout the reactor
ii. The reacting mixture is well stirred
iii. The composition of the exit stream is the same as that within the reactor
iv. Shaft work by the impeller or stirrer is negligible
v. Constant density
vi. The temperature within the reactor is kept at a constant value by the heat exchange medium

The design models of the CSTR and PFR in terms of its volume, height, diameter, space time and space velocity can be obtained by applying the principle of material balance stated as follows

$$\begin{bmatrix}\text{Rate of accumulation of material within the volume}\end{bmatrix} = \begin{bmatrix}\text{Rate of input of feed into the volume}\end{bmatrix} - \begin{bmatrix}\text{Rate of outflow of feed from the voume}\end{bmatrix} - \begin{bmatrix}\text{Rate of depletion of feed due to chemical reaction}\end{bmatrix} \quad (5)$$

For the CSTR, the terms in equation (5) can be defined, substituted and simplified at steady state operation to yield the following CSTR functional parameters thus;

$$V_{R(CSTR)} = \frac{F_{io}x_i}{k_o e^{-E/RT} C_{io}(1-x_i)} \quad (6)$$

$$H_{R(CSTR)} = \left[\frac{16F_{io}x_i}{\pi K_o e^{-E/RT} C_{io}(1-x_i)}\right]^{\frac{1}{3}} \quad (7)$$

$$D_{R(CSTR)} = \frac{\left[\frac{16F_{io}x_i}{\pi K_o e^{-E/RT} C_{io}(1-x_i)}\right]^{\frac{1}{3}}}{2} \quad (8)$$

$$\tau_{CSTR} = \frac{x_i}{K_o e^{-E/RT}(1-x_i)} \quad (9)$$

$$S_{V(CSTR)} = \frac{K_o e^{-E/RT}(1-x_i)}{x_i} \quad (10)$$

Generally, the quantity of heat generated per unit volume of reactors is mathematically given as;

$$Q = \Delta H_R F_{io} x_i \quad (11)$$

$$q = \frac{\Delta H_R F_{io} x_i}{V_R} \quad (12)$$

The energy balance equation of the CSTR and PFR for the exothermic esterification process can be obtained by applying the principles of conservation of energy given as;

$$\begin{bmatrix}\text{Rate of accumulation of heat within the volume}\end{bmatrix} = \begin{bmatrix}\text{Rate of Input of heat to the volume}\end{bmatrix} - \begin{bmatrix}\text{Rate of Output of heat from the volume}\end{bmatrix} - \begin{bmatrix}\text{Rate of depletion of heat due to chemical reaction}\end{bmatrix} - \begin{bmatrix}\text{Rate of heat removal to the surrounding}\end{bmatrix} + \begin{bmatrix}\text{Shaft work done by the stirrer}\end{bmatrix} \quad (13)$$

For the CSTR, terms in equation (13) can be defined, substituted and simplified at steady state to give the temperature effect model of the reactor thus;

$$T = \frac{\tau\Delta H_R r_i v_o + UA_c T_c + \rho v_o c_p T_o}{\rho v_o C_p + UA_c} \quad (14)$$

Consider the schematic representation of a plug flow reactor with mass and heat effect as shown in Figure 2.

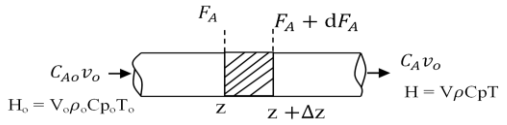

Figure 2: PFR Schematic with Mass and Heat Effect

The design and temperature effect models of the PFR were developed from the application of the conservation principle of mass and energy in equation (5) and (13) respectively

For the PFR, the terms in equation (5) 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;

$$V_{R(PFR)} = v_o \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RTC_{io}(1-x_i)}} \quad (15)$$

$$H_{R(PFR)} = \left[\frac{25v_o}{\pi}\int_0^{x_A} \frac{dx_A}{k_o e^{-E/RTC_{io}(1-x_i)}}\right]^{\frac{1}{3}} \quad (16)$$

$$D_{R(PFR)} = \frac{\left[\frac{25v_o}{\pi}\int_0^{x_A} \frac{dx_A}{k_o e^{-E/RTC_{io}(1-x_i)}}\right]^{\frac{1}{3}}}{25} \quad (17)$$

$$\tau_{PFR} = \int_0^{x_A} \frac{dx_A}{k_o e^{-E/RTC_{io}(1-x_i} \quad (18)$$

$$S_{V(PFR)} = \frac{1}{\int_0^{x_A} \frac{dx_A}{k_o e^{-E/RTC_{io}(1-x_{i)}}}} \quad (19)$$

The temperature effect model of the PFR in Figure 2 is developed using the conservation principle of energy balance in equation (13)

For the PFR, the terms in equation (13) can be defined, substituted and simplified to yield the temperature effect model thus;

$$\frac{dT}{dZ} = \frac{1}{u\rho C_p}(\Delta H_R)(-r_i) \quad (20)$$

iii. Data for Evaluation

The data for evaluation in this research are the properties/thermodynamic data and data obtained from literatures as presented in Tables 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	[4]
r_i	5.28×10^7mol/m^3	Reaction rate	[4]
E	59.403kJ/mol	Activation energy	[4]

III. Results and Discussion

The results of the flow reactor (CSTR and PFR) design assessment for ethyl acetate production are presented in Table 3 and Figure 3 to 10 below.

Table 3: Design Results of CSTR and PFR Volume, Height, Diameter, Space Time, Space Velocity, Quantity of Heat Generated and the Quantity of Heat Generated per unit Volume of the Reactors at 95% Fractional Conversion

Reactor Design Parameters (Unit)	@ 95% Fractional Conversion		Difference (%)
	CSTR	PFR	
Volume (m^3)	80.880	15.277	34.1
Height (m)	7.441	4.269	13.5
Diameter	3.270	2.135	10.5
Space Time (s)	15.859	2.996	34.1
Space Velocity (s^{-1})	0.063	0.334	34.1
Quantity of Heat Generated (J/s)	12821.580	12821.580	0.00
Quantity of Heat Generated per unit volume of the Reactor (J/sm^3)	158.526	839.254	34.1

Table 3 shows the design assessment or performance analysis of flow reactors (CSTR and PFR) during ethyl acetate production from the esterification reaction of acetic acid and ethyl alcohol. The MATLAB simulation of the flow reactors models was done at same initial feed and operating condition. At maximum conversion of 0.95, the CSTR and PFR volume was $80.880m^3$ and $15.277m^3$ respectively with a percentage difference of 34.1% while the quantity of heat generated per unit volume of the CSTR and PFR was $152.526j/sm^3$ and $839.254j/sm^3$ respectively with a percentage difference of 34.1%. the analysis of design results showed that more yield of ethyl acetate is produced in the CSTR as shown in the volume while the PFR performed better in terms of energy efficiency of the process as indicated in the quantity of heat generated per unit volume of the reactor.

i. Profile of CSTR and PFR Volume (V_R) and Fractional Conversion (X_A

Figure 3 is a graphical relationship between the CSTR and PFR volume and fractional conversion obtained from the MATLAB simulation of both reactors at same initial feed and operating temperature of 298.15k and 343.15k with varying fractional conversion of X_A $\geq 0 \leq 0.95$ at an interval of 0.05. According to the profile, the volume of both reactors increases exponentially as the fractional conversion increases. However, at maximum conversion of 0.95, the volume of the CSTR and PFR was 80.880m3 and 15.277m3 respectively. This result showed that more yield of ethyl acetate is produced in the CSTR compared to that of the PFR during the esterification reaction.

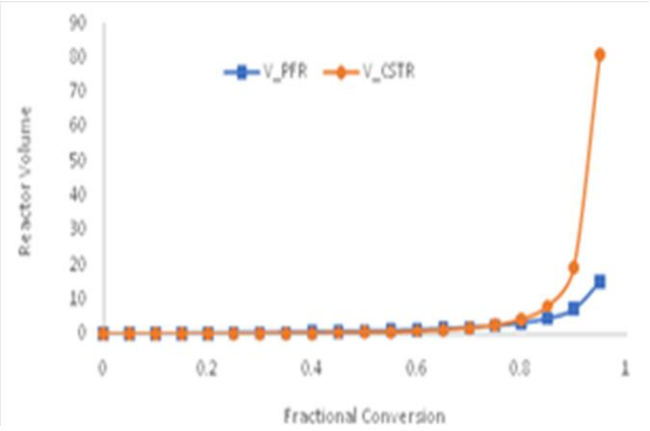

Figure 3: Profile of CSTR and PFR Volume (V_R) and Fractional Conversion (X_A)

ii. Profile of CSTR and PFR Height (H_R) and Fractional Conversion (X_A)

Figure 4 is a profile relationship of CSTR and PFR height and fractional conversion obtained from the MATLAB simulation of reactors performance models for height during the esterification process. The simulation was performed at same initial feed and operating temperature of 298.15k and 343.15k with fractional conversion variation of XA $\geq 0 \leq 0.95$ at an interval of 0.05. From the profile behaviour, the flow reactors height was increased exponentially as the fractional conversion increases and at a maximum conversion of 0.95, the height of the CSTR and PFR design was 7.441m and 4.369m respectively with a percentage difference of 13.5%. This result is mathematically just since the reactor volume is also a function of its height.

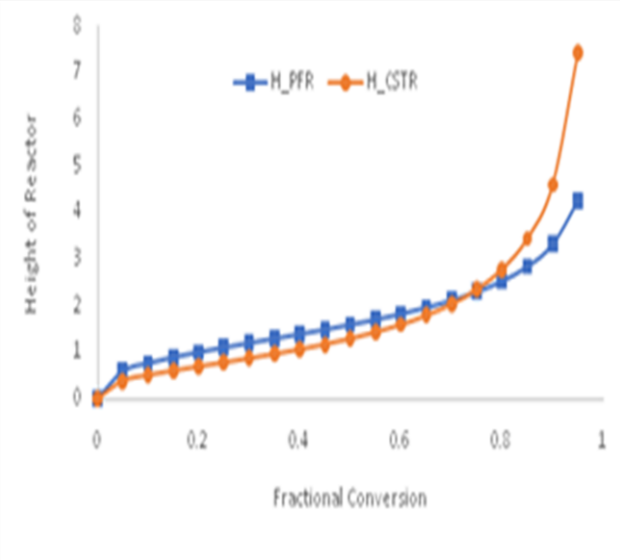

Figure 4: Profile of CSTR and PFR Height (H_R) and Fractional Conversion (X_A)

iii. Profile of CSTR and PFR Diameter (D_R) and Fractional Conversion (X_A)

Figure 5 shows a variation of the CSTR and PFR diameter with fractional conversion for the production of ethyl acetate from the esterification of acetic acid and ethyl alcohol. This profile was obtained from the MATLAB simulation of the steady state performance model of the reactors diameter at same initial feed and operating temperature of 298.15k and 343.15k with fractional conversion variation of XA $\geq 0 \leq 0.95$ at an interval of 0.05. According to the plot, the diameter of both reactors was increased exponentially as the fractional conversion increases in both reactors. At a maximum fractional conversion of 0.95, the CSTR and PFR fractional conversion was 3.270m and 2.135m respectively with a percentage difference of 10.5%. This is justified by the high significant difference between both reactors volume.

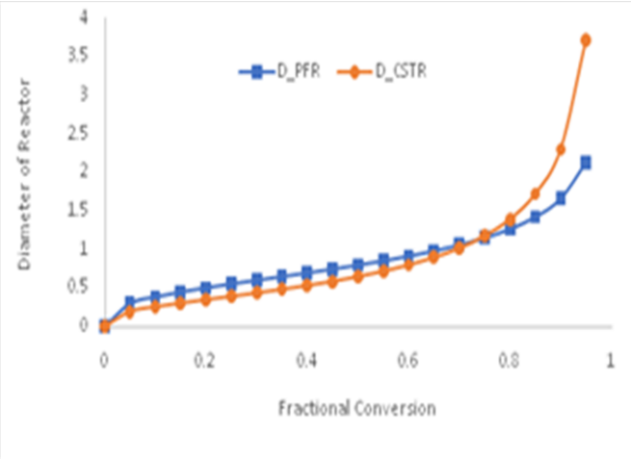

Figure 5: Profile of CSTR and PFR (D_R) and Fractional Conversion (X_A)

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

Figure 6 shows a slow exponential increase of the CSTR and PFR space time at a fractional conversion below 0.75. At higher fractional conversion above 0.75, the exponential increase of the space time taken became rapid and more significantly in the CSTR. This profile was obtained from the MATLAB simulation of the space time steady state model for the esterification process. At a maximum fractional conversion of 0.95, the CSTR and the PFR space time values taken was 15.859seconds and 2.996seconds respectively with a significant percentage difference of 34.1%. This significant difference between the CSTR and PFR space time is as a result of the continuous operation associated with the CSTR.

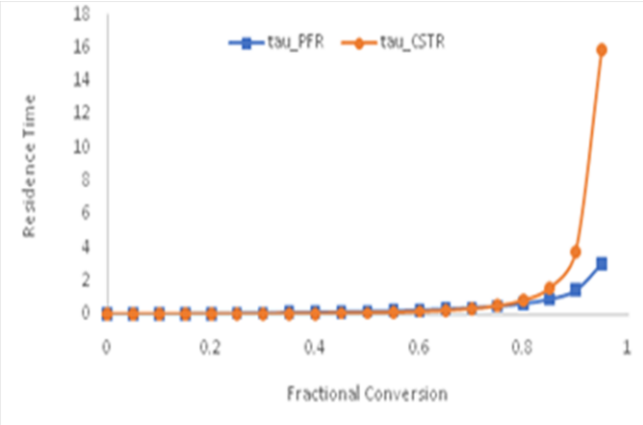

Figure 6: Profile of the CSTR and PFR Space Time (τ) and Fractional Conversion (X_A)

v. Profile of the CSTR and PFR Space Velocity (S_V) and Fractional Conversion (X_A)

Figure 7 shows the CSTR and PFR space velocity variation with fractional conversion during the ethyl acetate production in both reactors. This profile was developed from the MATLAB simulation of the steady state performance models of the reactors space velocity at same initial feed and operating temperature of 298.15k and 343.15k with change in fractional conversion of XA $\geq 0 \leq 0.95$ at 0.05 intervals. According to the plot, the space velocity of the reactors decreases exponentially as the fractional conversion increases. At higher fractional conversion above 0.9, the space velocity value in both reactors tends towards negative infinity. This profile behaviour is justified by the mathematical relationship between the space time and the space velocity. At a maximum conversion of 0.95, the CSTR and PFR space velocity was $0.063s^{-1}$ and $0.334s^{-1}$ respectively with a percentage difference of 34.1%. Here, the space velocity of the PFR is higher because of its configuration for fast reaction within a short residence time compared to that of the CSTR.

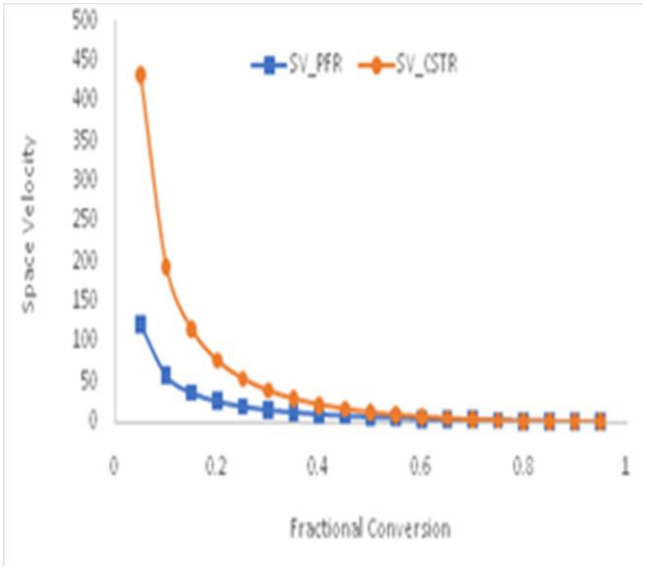

Figure 7: Profile of the CSTR and PFR Space Velocity (S_V) and Fractional Conversion (X_A)

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

Figure 8 shows there is linear increase in the CSTR and PFR quantity of heat generated as the fractional conversion increases during the esterification process in both reactors. This profile relationship was obtained was developed from the MATLAB simulation of the reactors quantity of heat generated models at the same initial feed and operating temperature of 298.15k and 343.15k with fractional conversion variation of XA $\geq 0 \leq 0.95$ at 0.05 intervals. The plot showed that there is no variation in the quantity of heat generated in both reactors and at a maximum conversion of 0.95 the quantity of heat generated in the CSTR and PFR was 12821.580J/s.

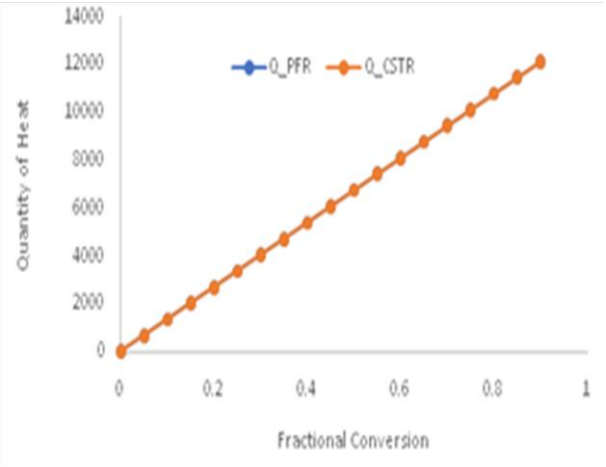

Figure 8: Profile of the CSTR and PFR Quantity of Heat Generated (Q) and Fractional Conversion (X_A)

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

Figure 9 shows the relationship between the CSTR and PFR quantity of heat generated per unit volume of the reactors and fractional conversion during the esterification reaction process. This profile was developed from the MATLAB simulation of the process using the same operating condition in both reactors. According to the profile, the quantity of heat generated per unit volume of both reactors decreases exponentially as the fractional conversion increases. At a maximum conversion of 0.95, the quantity of heat generated per unit volume of the CSTR and PFR was 158.526j/sm3 and 839.254j/sm3 respectively with a percentage difference of 34.1%.This results showed that the PFR design is better in terms of energy efficiency of the process than the CSTR.

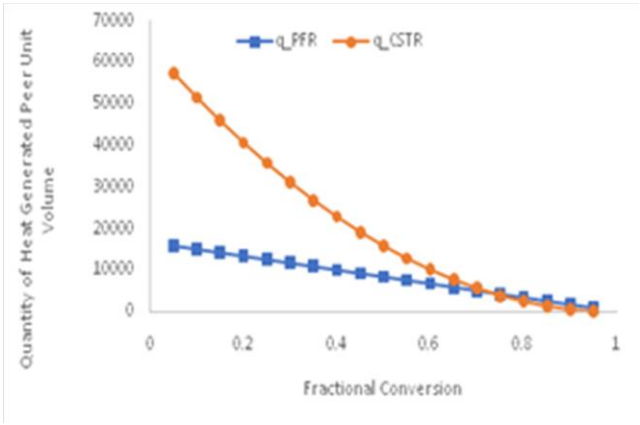

Figure 9: Profile of the Quantity of Heat Generated per unit Volume of the CSTR and PFR (q) with Fractional Conversion

viii. Profile of the CSTR and PFR Temperature and the Fractional Conversion

Figure 10 is the graphical relationship between the CSTR and PFR temperature and the fractional conversion during the esterification process for ethyl acetate production. This profile was gotten from the MATLAB simulation of the flow reactors temperature effect models at same initial feed and operating temperature of 298.15k and 343.15k respectively with changes in the fractional conversion from 0 to 0.95 at 0.05 intervals. According to the profile, the changes or variation in fractional conversion during the process has no significant effect on the operating temperature of the reactors. This is based on the fact that the process temperature was under control or the process was performed at standard operating temperature within the range for esterification. However, temperature above or below the standard specification will results to side reactions, loss of materials and formation of low quality or purity of ethyl acetate.

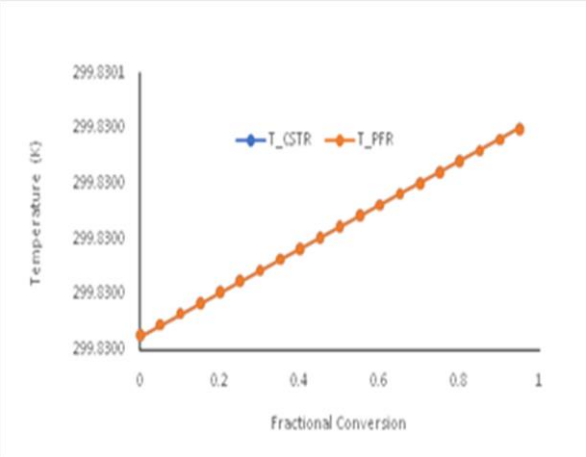

Figure 10: Profile of the CSTR and PFR Temperature and Fractional Conversion

IV. Conclusion

The research considered the application of the conservation principle of mass and energy in the design of flow reactors (CSTR and PFR) for ethyl acetate production from the esterification of acetic acid and ethyl alcohol. The steady state design models of the flow reactors were simulated using MATLAB at same feed and operating temperature of 298.15k and 343.15k with fractional conversion variation range of XA $\geq 0 \leq 0.95$ at 0.05 intervals. The reactor designs showed that at a maximum fractional conversion of 0.95, the volume of the CSTR and PFR was $80.880m^3$ and $15.277m^3$ with a percentage difference of 34.1% while the quantity of heat generated per unit volume of the CSTR and PFR was $158.526j/sm^3$ and $839.254j/sm^3$ with a percentage difference of 34.1%. The comparative analysis or assessment of both reactor design for the esterification process showed that more yield of ethyl acetate is produced in the CSTR as indicated by the reactor volume while in terms of energy efficiency, the PFR was more effective as indicated by the quantity of heat generated in the reactor. This research has shown that both the CSTR and the PFR are suitable reaction media for esterification process and the choice of either of the reactor is dependent on the designer's need.

Nomenclature

Symbol	Definition	Unit
ΔH_R	Charge 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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