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PREDICTIVE PSEUDO-STEADY-STATE MECHANISTIC MODEL FOR CHAIN REACTIONS IN PETROCHEMICAL FURNACE REACTORS

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ABSTRACT

The research predicts the application of pseudo steady-state mechanism to chain reactions. Process chain reactions taking place in a petrochemical furnace reactor generates many intermediates species, and the material balance process are written in the kinetic balance to account for the contributory effects of the intermediates species than assuming negligible concentrations in the kinetic balance process. A set of material balance model on the number of molecules cum intermediates species resulting from the free radicals' mechanism of the initiation, propagation, and termination all taking place in the furnace reactor were developed; and resolved simultaneously with energy/temperature balance of the radiative-convective zones of the petrochemical furnace reactor. A mat-lab simulation process applying the petrochemical plant process data as boundary conditions, gave plot profiles of the molecules (ethylene, hydrogen, methane, and butane) and active intermediates of (methyl, ethyl, and hydrogen) and a clear flat plateau which mainly depicts applicability of pseudo steady-state mechanism in kinetic studies. The models predict the following results ethane cracking 5.57%. ethylene formation 14.6%, temperature effects 0.03% and pressure drop 3.5 % which is very adequate for the petrochemical furnace reactor operations industrially. Finally, the essence of the work is to demonstrate how intermediates species formed in reactions process should be incorporated in any given material balance model to account for the overall kinetic studies rather than neglecting as zero contributions in any process chemistry.

Contribution/Originality: The research contributes to chemical engineering view point a pseudo steady state mechanism incorporates radical species of petrochemical chain reactions in writing the material balance to account for the overall effects in kinetic studies. While, chemistry assumes radical species are spectator ions assumed to be zero in any reactions kinetics.

1. INTRODUCTION

Thermal cracking processes of hydrocarbons have been of great interest to chemist even before the petroleum industry existed. The industry of producing chemicals from petroleum started when chemist produced alcohol from ethylene and propylene via thermal cracking.

Hence, cracking of hydrocarbon thermally generates vital products as C_2H_4, C_4H_{10}, methane, and hydrogen to chemical and petrochemical industries as basic hydrocarbon feed stocks [1].

Cracking operations in petrochemical furnace reactor may involve dehydrogenation, polymerization, isomerization, alkylation and other reactions carried out at high temperatures and low pressures of 1100K and

pressure of 560 Pascal Wordu and Akinola [2]; Rase [3] sometimes catalyst is added to control the chemical reactions which occur during the process, with the goal of promoting the development of specific molecules [4].

The thermodynamic properties such as standard heat of formation ($\Delta H_{f,298}$) specific heat capacity(C_P) and kinetic data for chemical species i of the reactions process are given in Table 1 and 2. Boris [5]; Kutepov, et al. [6] posited destructive petroleum processing applying thermal cracking of hydrocarbons as proceeding by the free-radical mechanism which involves three stages of chain initiation, chain propagation, and chain termination processes. The structural stereochemistry of the compounds undergoing chain reaction in the furnace reactor are shown in Figure 1 .

Chain propagation (free-radical reactions) the unsaturated groups of atoms or ions which contain one or more unpaired electrons, that is, which do have their full complement of electrons, and do not decompose instantaneously to more stable species, are called free radicals. As a result of an incomplete valence shell, free radicals are highly reactive, and the reactions involving them proceed at a high rate. On colliding with the molecules of the feed stock, they form a product plus yet other free radicals, and this process, once initiated, can be repeated over and over, making the reaction self-propagating. The life time of free radicals is very short, being of the order of 10^{-3} -10^{-4} $^-$s. The entire series of reactions that follows the production of the very reactive intermediates, or free radicals, is called a chain reaction. The bulk of the reaction product results precisely from a chain reaction via the free radicals rather than from the rupture of the carbon chain. The reactions involving free radicals are as follows:

1.   The substitution reaction

$$CH_3 + C_2H_6 - CH_4 + C2H_5$$
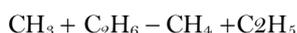
2.   The decomposition of free radicals with the formation of   unsaturated molecules plus more free radicals

$$CH_3CH_2CHCH_3 - CH_2 = CHCH_3 + CH_3$$

3.   The addition of radicals in multiple bonds

$$CH_3 + C2H_4 - C_3H_7$$

4.   Isomerism (presumably, it proceeds via an intermediate cyclic state)
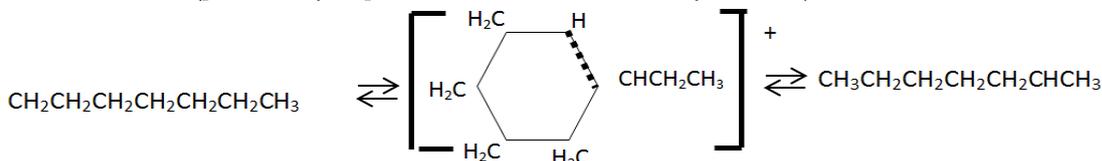
Chain termination at equilibrium, the probability of a free radical meeting another free radical becomes comparable with the probability of their collision with the molecules of the feedstock. The interaction of two free radicals leads to what is termed chain termination with the formation of a stable hydrocarbon:

$$CH_2 = CH + CH = CH_2 \; - CH_2 = CH - CH = C_2$$
$$CH_2 = CH + H - CH_2 = CH_2$$
$$C_2H_5 + H - C_2H_4 + H_2$$
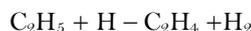
Figure 1 chain reactions scheme.

Hence, Pseudo Steady-State mechanism (PSS) is applied in species concentrations balancing process of the intermediate species generated due to chain reactions in the petrochemical furnace reactor.

Therefore, the research introduced PSS-mechanistic approach as the underlying kinetic principles accounting for the intermediates species effects in chain reaction process of furnace reactor.

2. MATERIALS AND METHOD

2.1. Materials

The materials for the analytical technique research are obtained from industrial petrochemical plant as shown in Table 1 and 2.

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Table-1. Properties values at 25°c and atmospheric pressure.

Properties	Values
Molecular weight	30.07
Specific gravity	01.04
Density of liquid and B.Pt (Kg/m³ and k)	34.10, 546.49
Absolute viscosity (Centipoises)	0.095
Specific heat (cp) in J/kgk	171
Thermal conductivity (W/M°c)	0.017
Latent heat of vaporization at B.Pt (J/kg)	488000
Flammable	yes
Heat of combustion (KJ/kg)	51800

Sources: Perry and Green [7]; Smith, et al. [8].

2.1.1. Petrochemical Furnace Reactor Parameters

Table-2. Industrial plant data petrochemical plant, Rivers State-Nigeria.

S/N	Reactor Parameters	Values
1	C_{P_A}	75.409
2	T	1100K
3	C_{P_B}	133.84
4	C_{P_C}	865.75
5	$C_{P_i} = C_{P_A} + C_{P_B} + C_{P_C}$	924.18
6	ρ_i	0.0859
7	q_C	$7.56 x 10^7$
8	q_R	$4 x 10^4$
9	q	$7.60 x 10^7$
10	$A = 2\pi R L$	47771.4
11	$(-\Delta H_r)$	$51872.2 \frac{KJ}{KMol}$
12	$\omega = \frac{q}{\rho_i C_{P_i}}$	$7.97 x 10^5$
13	$j = \frac{-\Delta H_r}{\rho_i C_{P_i}}$	653.4
14	Total Pressure (P$_T$)	560KPa
15	Mass flux (G)	112.4kg/m²s
16	Reactor temperature (T$_R$)	1100K
17.	Pressure drop (ΔP)	0.45KPa
18	Reactor length (z)	152m
19.	Tube thickness (Δd)	0.01m
20	Tube thickness (Δd)	0.01m
21	Thermal conductivity of reactor tube (K)	11.8W/MK

Source: [EPCL Nigeria Petrochemical Reactor Plant]; Wordu and Akinola [2]; Wordu and Ojong [9].

2.2. Methods

2.2.1. Development of Kinetic Model – PSS-Mechanism Applied Fogler [1]

The concept of PSS-mechanism applied in material balance of intermediate species formed in depleting of feed in reactors is quite innovative as previous balances considers intermediate species as zero ie having no effects on material balance in reaction process.

The research made use of the concept in the development of the material balances for the chain reactions taking place in the petrochemical furnace reactors.

Therefore, the general formula for the saturated hydrocarbon cracking to ethylene as desired product accompanied by active intermediates and molecular hydrogen and methane gas are given by the chemical Equation 1.

$$C_nH_{2n+2} \xrightarrow{k_1} C_{(n-m)}H_{2m+2} + C_mH_{2m} \qquad (1)$$
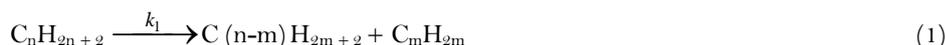
Where, n > m and is the number of atoms

The research adopted process chemistry where the pyrolysis of ethane is irreversible first-order chemical reaction process with a temperature of 1100k given below:

$$C_2H_{6\,(g)} \xrightarrow{k_1} C_2H_{4_{(g)}} + H_{2_{(g)}} \qquad (2)$$

The yield of ethylene can be as high as 65-70%.

The chemical process Equation 2 is a multiple and complex reaction taking place in the petrochemical furnace reactor. The recognized sequence of initiation, propagation and termination are the fundamental basis of researching on the chain reactions mechanism of a given petrochemical plant operations.

A little insight into initiation is the process of generating the active intermediate with which to kick-start entire process; propagation or chain transfer is the process of interaction of active intermediate with the reactant or product to produce another active intermediate, and finally, deactivation of the active intermediate to form products.

Therefore, the research adopts kinetic balance of the chain reactions concepts elucidated and can be applied to the thermal decomposition of ethane to ethylene, methane, and butane and hydrogen gas.

The model development applies the *PSS-mechanism* to derive the rate laws for the formation of ethylene, and material balance for furnace reactor operating pyrolytically at temperature of 1100K, plant pressure of 560 Pascal and feed rate of 55,000Kg/hr.

Typical kinetic process taking place in the plug flow furnace reactor maintain the reaction sequence stated as follows:

2.2.1.1. Initiation

The starting material feed component having to kick-start the pyro lytic process by decomposing to generate the much needed methyl radical with an unpaired electron ready for transfer process.

$$C_2H_6 \xrightarrow{k_1} 2CH_3^{\circ} \qquad (3)$$

Equation 5 expresses the ethane feed cracking to generate the needed radicals to initiate the process.

2.2.1.2. Propagation

The methyl radical with an unpaired electron undergoes chain transfer by having to interact with the feed or product to produce another active intermediates ethyl and hydrogen radicals, and products methane, ethylene, butane and hydrogen gas;

$$CH_3^\bullet + C_2H_6 \xrightarrow{K_2} CH_4 + C_2H_5^\bullet \qquad (4)$$

$$C_2H_5^\circ \xrightarrow{k_3} C_2H_4 + H^\circ \qquad (5)$$

$$H^\circ + C_2H_6 \xrightarrow{k_4} C_2H_5^\circ + H_2 \qquad (6)$$

Equations 4, 5 and 6 shows the stepwise kinetic models for the chain reactions taking place in the petrochemical furnace reactor with Equation 5 predominant of ethylene formation and hydrogen radical generation.

2.2.1.3. Termination

$$2C_2H_5^\circ \xrightarrow{k_5} C_4H_{10} \qquad \textbf{(7)}$$

Equation 7 reaction is deactivation of the active intermediate to produce the desired products of ethylene, methane, butane, and hydrogen gas, bringing the entire chain reactions into abrupt end. The rate equations expressing the three kinetic processes are:

$$r_{1, C2H6} = - K_{1, C2H6} C_2H_6 \qquad (8)$$

$$K_1 = K_{1, C2H6}$$

$$r_{2, C2H6} = - K_2 CH_3^* C_2H_6 \qquad (9)$$

$$r_{3, C2H4} = K_3 C_2H_5^* \qquad (10)$$

$$r_{4, C2H6} = - K_4 H^* C_2H_6 \qquad (11)$$

$$r_{5, C2H5} = - K_{5, C2H5*} \left[C_2H_5^* \right]^2 \qquad (12a)$$

Equation 12 decimate to equation 12b

$$K_5 = K_{5, C2H5*} \qquad (12b)$$

Subsequently, Equations 8, 9, 10, 11, 12a and 12b are the equivalent rate equations for the kinetics taking place in the reactor. While the superscripts * indicates radical species.

2.3. Rate Laws

PSS-mechanism *balance Intermediate Species*.

The rate of depletion leading to formation of ethylene desired reaction 5, given by the Equation 13

$$r_{3, C_2H_4} = k_3 \left[C_2H_5^\circ \right] \qquad (13)$$

Equation 13 gives rise to the net rate of reaction for the active intermediates CH_3^*, $C_2H_5^*$, H^*

Net rate of ethyl radical:

$$\left(\frac{dC_{C_2H_5^\circ}}{d\tau} \right) = r_{2,C_2H_5^\circ} + r_{3,C_2H_5^\circ} + r_{4,C_2H_5^\circ} + r_{5,C_2H_5^\circ} = 0 \qquad (14)$$

Equation 14 shows the coupling of ethyl radical component balance.

And reaction stoichiometric coefficients of the entire process given as;

$$r_{2,C_2H_5^\circ} = -r_{2,C_2H_4}, \ r_{3,C_2H_5^\circ} = -r_{3,C_2H_4}, \ r_{4,C_2H_5^\circ} = -r_{4,C_2H_4},$$

Coupling the rate laws yields the actual material or mole balance for the entire kinetics taking place in the furnace reactor:

$$\left(\frac{dC_{C_2H_5^\circ}}{d\tau}\right) = -r_{2,C_2H_6} - r_{3,C_2H_4} - r_{4,C_2H_6} + r_{5,C_2H_5^\circ} = 0 \tag{15}$$

$$\left(\frac{dC_{H^\circ}}{d\tau}\right) = r_{3,H^\circ} + r_{4,H^\circ} = r_{3,C_2H_4} + r_{4,C_2H_6} = 0 \tag{16}$$

$$\left(\frac{dC_{CH_3^\circ}}{d\tau}\right) = r_{1,CH_3^\circ} + r_{2,CH_3^\circ} = -r_{1,C_2H_6} + r_{2,C_2H_6} = 0 \tag{17}$$

Substituting the concentrations into the elementary Equation 17 and Simplifying equation (17) by substituting the rate equations (-r₁) and (-r₂) yields Equation 18.

$$2k_1[C_2H_6] - k_2[CH_3^\circ][C_2H_6] = 0 \tag{18}$$

Equation 18 is obtained by simplifying Equation 17 .

Solving for the concentration of the free radical, $[CH_3^\circ]$

$$[CH_3^\circ] = \frac{2k_1}{k_2} \tag{19}$$

Adding Equations 15 and 16 yields.

$$-r_{2,C_2H_6} + r_{5,C_2H_5^\circ} = 0 \tag{20}$$

Substituting for concentrations in the rate laws

$$k_2[CH_3^\circ][C_2H_6] - k_5[C_2H_5^\circ]^2 = 0 \tag{21}$$

Solving for $[C_2H_5^\circ]$ gives us

$$[C_2H_5^\circ] = \left\{\frac{k_2}{k_5}[CH_3^\circ][C_2H_6]\right\}^{\frac{1}{2}} = \left\{\frac{2k_1}{k_5}[C_2H_6]\right\}^{\frac{1}{2}} \tag{22}$$

Substituting for $[C_2H_5^\circ]$ in Equation 10 yields the rate formation of ethylene

$$r_{C_2H_4} = k_3[C_2H_5^{\cdot}] = k_3 \left(\frac{2k_1}{k_5}\right)^{\frac{1}{2}} [C_2H_6]^{\frac{1}{2}} \qquad (23)$$

Next we write the rate of $H^{\cdot}$ formation in Equation 16 in terms of concentration

$$k_3[C_2H_5^{\cdot}] - k_4[H^{\cdot}][C_2H_6] = 0$$

Using Equation 22 to substitute for $[C_2H_5^{\cdot}]$ gives the concentration of the hydrogen radical

$$[H^{\cdot}] = \frac{k_3}{k_4}\left(\frac{2k_1}{k_5}\right)^{\frac{1}{2}} [C_2H_6]^{-\frac{1}{2}} \qquad (24)$$

The rate of disappearance of ethane is

$$r_{C_2H_6} = -k_1[C_2H_6] - k_2[CH_3^{\cdot}][C_2H_6] - k_4[H^{\cdot}][C_2H_6] \qquad (25)$$

Substituting Equations 19 and 24 yields Equation 26

$$r_{C_2H_6} = -\left\{3k_1[C_2H_6] + k_3[C_2H_6]^{\frac{1}{2}}\left(\frac{2k_1}{k_5}\right)^{\frac{1}{2}}\right\} \qquad (26)$$

Thus the various rates expressions for the remaining intermediates hydrogen, methyl, and ethyl radicals gives Equations 27 to 32 below:

$$r_{CH_3^{\cdot}} = -2k_1[C_2H_6] \qquad (27)$$

$$r_{CH_4} = 2k_1[C_2H_6] \qquad (28)$$

$$r_{H^{\cdot}} = k_3[C_2H_6]^{\frac{1}{2}}\left\{\sqrt{\frac{2k_1}{k_5}} - \sqrt{\frac{k_5}{2k_1}}\right\} \qquad (29)$$

$$r_{H_2} = \sqrt{\frac{2k_1}{k_5}}[C_2H_6] \qquad (30)$$

$$r_{C_4H_{10}} = k_1[C_2H_6] \qquad (31)$$

$$r_{[C_2H_5^{\cdot}]} = 0 \qquad (32)$$

2.4. Temperature Effects

$$v\frac{dT}{dz} = \varphi e^{-\frac{E}{RT}}C_A + \omega \qquad (33)$$

Where $\boldsymbol{\varphi = -jk_0}$

Equation 33 is the energy/temperature effect along the furnace reactor for ethane cracking.

$$q \ = 20 + GC_{PA}\left(\frac{K}{h_i\Delta d}\right)\left(T_{FG} - T_i\right) \tag{34}$$

While Equation 34 represents the quantity of heat q in the reactor

Pressure drop

$$\frac{dp_{Tot.}}{dZ} = - \ \frac{2fG^2}{1000\rho d} \tag{35}$$

Equation 35 represents the pressure effects in the furnace reactor.

3. SOLUTION TECHNIQUES PROCESS – MODELS FOR ITERATIVE PROCESS

Let, $1 = C_2H_6$, $2 = CH_3^*$, $3 = CH_4$, $4 = C_2H_5^*$, $5 = C_2H_4$, $6 = H^*$, $7 = H_2$,

$8 = C_4H_{10}$

$$r_1 = -k_1 y_1 - k_2 y_1 y_2 - k_4 y_1 y_6 \tag{36}$$

$$r_2 = 2k_1 y_1 - k_2 y_1 y_2 \tag{37}$$

$$r_3 = k_2 y_1 y_2 \tag{38}$$

$$r_4 = k_2 y_1 y_2 - k_3 y_4 + k_4 y_1 y_6 - k_5 y_4^2 \tag{39}$$

$$r_5 = k_3 y_4 \tag{40}$$

$$r_6 = k_3 y_4 - k_4 y_1 y_6 \tag{41}$$

$$r_7 = k_4 y_1 y_6 \tag{42}$$

$$r_8 = \frac{1}{2} k_5 y_4^2 \tag{43}$$

Equations 36 to 43 are expressed in mass fractions forms for mat-lab simulations.

$$\left[C_2H_5^\circ\right] = \left\{\frac{k_2}{k_5}\left[CH_3^\circ\right]\left[C_2H_6\right]\right\}^{\frac{1}{2}} = \left\{\frac{2k_1}{k_5}\left[C_2H_6\right]\right\}^{\frac{1}{2}} \tag{44}$$

$$\left[H^\circ\right] = \frac{k_3}{k_4}\left(\frac{2k_1}{k_5}\right)^{\frac{1}{2}}\left[C_2H_6\right]^{-\frac{1}{2}} \tag{45}$$

$$\left[CH_3^\circ\right] = \frac{2k_1}{k_2} \tag{46}$$

Equations 44, 45 and 46 are the active radicals balance in the chain reactions mechanism taking place in the petrochemical furnace reactor (plug flow reactor).

4. RESULTS AND DISCUSSIONS

The results and discussions of the conceptual research are conjunctively taken using the plot profiles of figures 1 to 7. The complex cracking rate laws of component species cum energy balances of radiation-convective zones and pressure effects were evaluated simultaneously to predict the *PSS-mechanism* on the chain reactions of the ethane cracking in the petrochemical furnace reactor.

The application of *PSS-mechanism* accounts for the balances of the three short-lived intermediate species which are so reactive that they never accumulate in large quantities and are difficult to detect in the cracking process.

While, the molecular hydrogen formed was routed to the methanol blending plant for *(CO+H₂) mixture* as synthesis gas feed for fertilizer production.

The butane gas which is the heavier stuff of the cracked gases majorly contributes to the coke formations on the cracking furnace, and also serves for blending with propane gas in some determined percentages for domestic gas purpose.
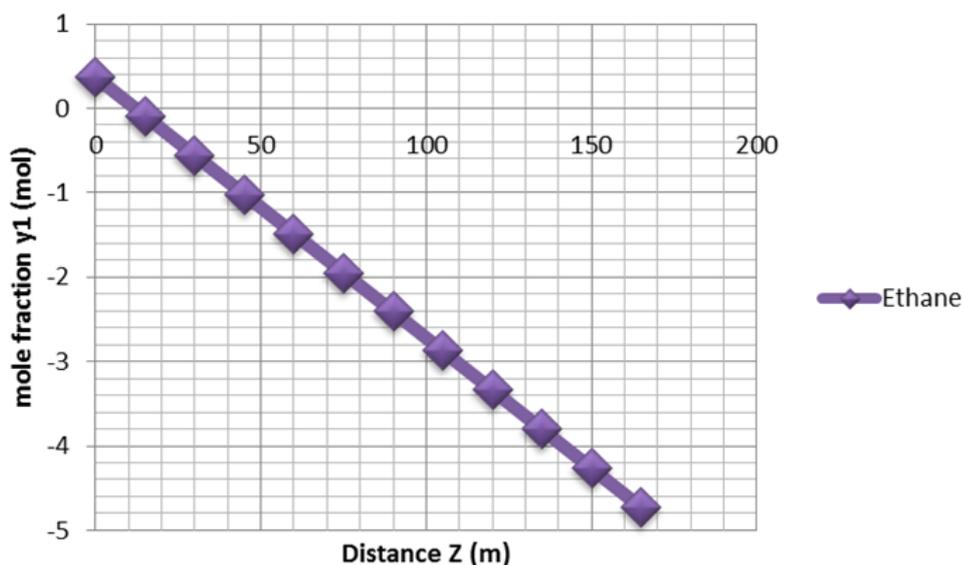
Figure-1. Mole fraction of ethane depletion with distance z.

Figure 1 exhibits the mole fraction-for the ethane feed depletion in the furnace reactor.
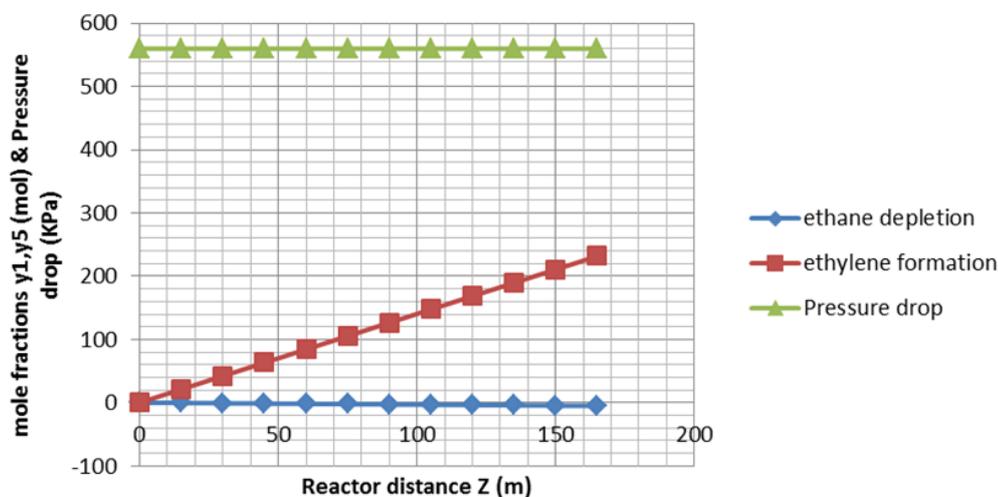
Figure 2 Composite plots of mole fractions and pressure drop with reactor distance z.

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Figure 2 depicts a composite plot of ethane depletion, ethylene formation and pressure drop along the length of the reactor. From the plot as ethane is cracked in the furnace reactor ethylene increases to a maximum point, while pressure drop is fairly constant.
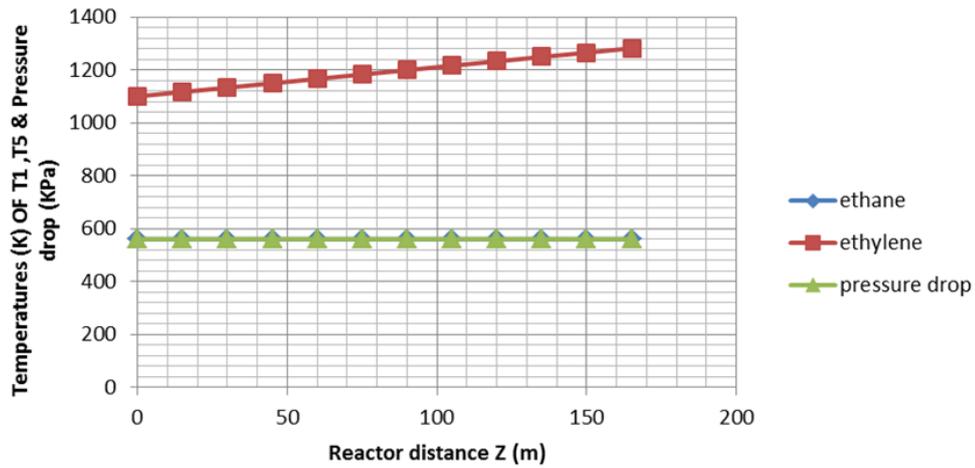
Figure-3. Comparisons plot of temperatures T_1, T_5 and pressure drop.

Figure 3 exhibits a progressive formation ethylene at a reactor pressure of operations.
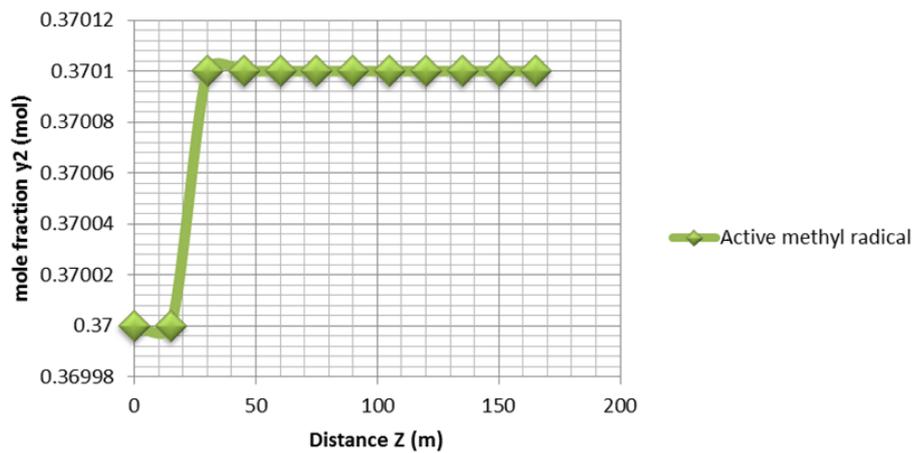
Figure-4. Methyl radical variations along reactor length (m).
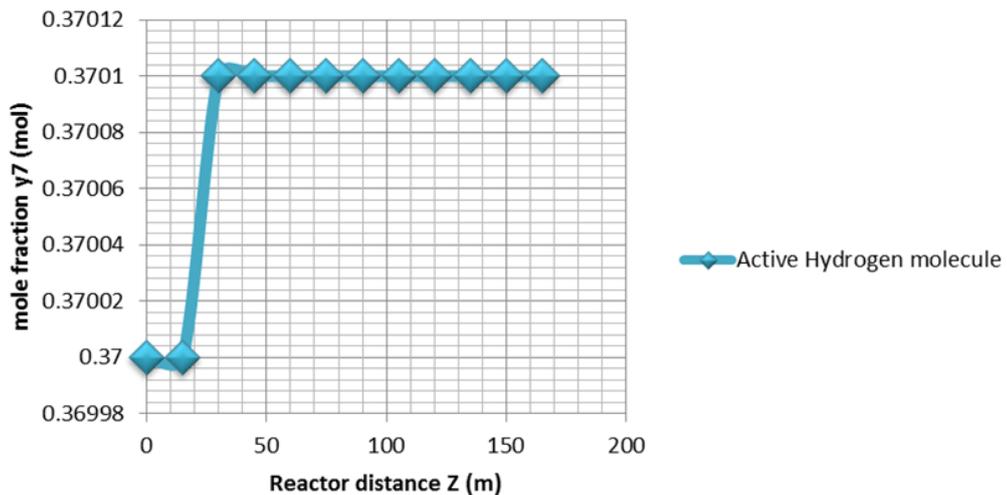
Figure-5. Active H_2 molecule profile along reactor length (m).

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Figure 4 and 5 predict the *PSS-mechanism* compliance of the research, it from initial concentration to a maximum then maintains a steady state and become sufficiently constant. which implies that at this point the change in the concentration as a function of distance z in the reactor is a constant zero, *i.e* $\dfrac{dci}{dz} = 0$ which is the clear essence of studying for *PSS-mechanism* of chain reactions, which is where the intermediate species CH_3*, H^+ effects are accounted for in the material balances.
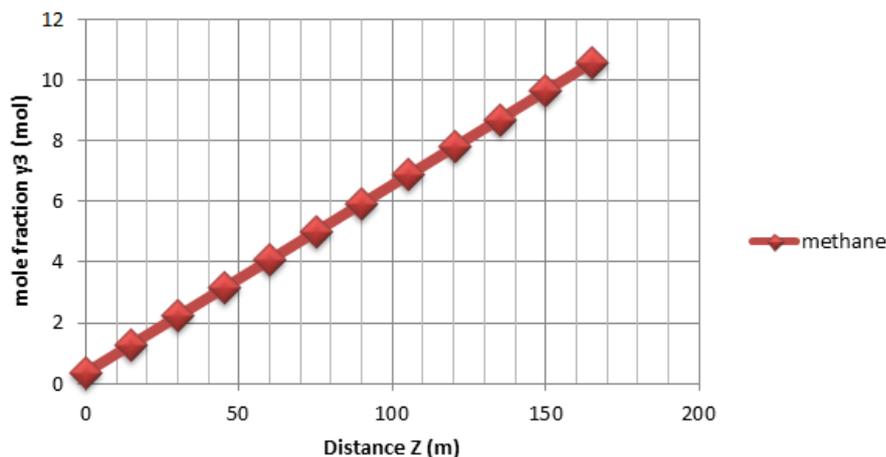
Figure-6. Plot of mole fraction of methane versus reactor distance.
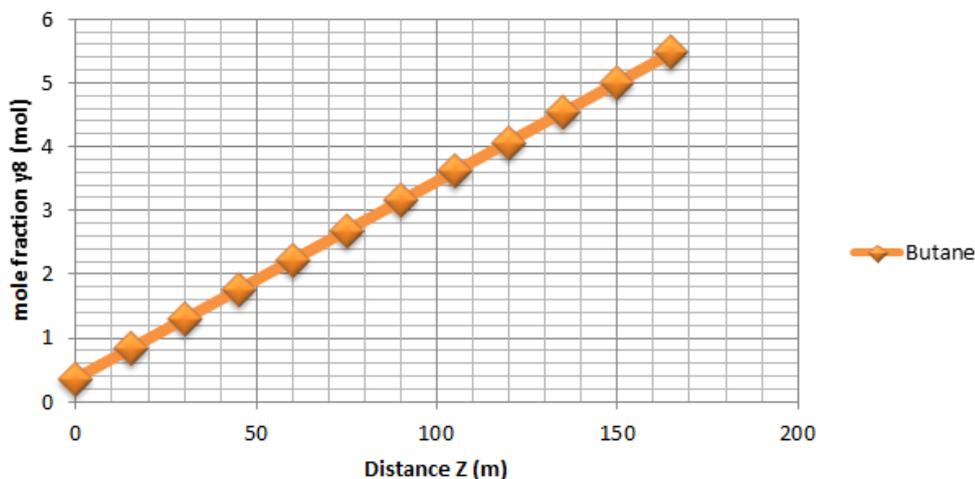
Figure-7. Mole fractions with distance for Butane formation.

Figures 6 and 7 profiles for methane and butane formation are expected as cracking process continues these two gases becomes molecularly favored by the chain reactions process. The aggregate formation of methane is routed to the ammonia plant for fertilizer production, while, butane serves as major feed for (LPG) liquefied petroleum gas for domestic purposes.

5. CONCLUSION

The research shows that the chemical reaction engineering concepts of pseudo steady state mechanism can be applied to chain reactions processes in the material balance at steady state process to account for the radical formations in the depleting or cracking of ethane feed or any other feed cracking processes in a reactor.

NOMENCLATURE

A	-	Cross-sectional area of reactor tube, (m²)
C_A	-	Concentration of ethane, (kgmol/m³)
C_{Pi}	-	Specific heat capacity of component i, $KJ/Kgmol.k$
D	-	Pipe internal diameter, (m).
f	-	Fanning friction factor
G	-	Mass flux, $\left(Kg/m^2.S\right)$
h	-	Step size change, (- -)
L	-	Length of reactor tube, (m)
K	-	Reaction Rate constant (Sec⁻¹)
n_A	-	No. of moles of A.
M_{wt}	-	Molecular weight of ethane $\dfrac{kg}{kgmol}$
F_T	-	Total gas flow at any point, C j
F_{AO}	-	Inlet molar fined rate of ethane, $\dfrac{kgmol}{sec}$
F_{TO}	-	Total inlet flow rate
F_A	-	Molar flow rate of ethane at any point, $\dfrac{kg}{sec}$
F_B	-	Molar flow rate of ethylene at any point, $\dfrac{kgmol}{sec}$
Fc	-	Molar flow rate of hydrogen at any point $\dfrac{kgmol}{sec}$
P	-	Total pressure at any point, (KPa)
P_o	-	Inlet total pressure, (KPa)
Q	-	Heat input from furnace $\dfrac{KJ}{sm}$ (of reactor length)
r_A	-	Specific reaction rate $\left(\dfrac{Kgmol\ ethane}{m^3.5}\right)$
R	-	Universal gas constant = 8.314kpa. $\dfrac{m^3}{Kgmol.K}$
Re	-	Reynolds Number, $\left(\int UD\ _g\ (\text{--})\right)$

$\boldsymbol{\mu}$

T	-	Absolute Temperature, (K),
T_o	-	Inlet Temperature, (K)
T_{out}	-	Outlet temperature, (K)
U	-	Superficial velocity, (m/s)
V_R	-	Reactor volume, (m³)
X_A	-	Mole fraction of ethane converted to products. i.e conversion of Ethane
X_{AD}	-	Inlet mole fraction of ethane
$X_{A\ MAX}$	-	Maximum conversion of ethane
Y_A	-	Mole fraction of ethane
Z	-	Length measured from reactor inlet, (m)
β	-	Heat-transfer surface area per unit volume of the reactor, (m²/m³)
ΔH^0_{298}	-	Heat of formation, (kg/kgmol)
ΔH_R	-	Heat of pyrolysis reaction, (KJ/kgmol)
Δp	-	Pressure drop through the reactor, (Psia)

Greek Symbols

Σ_A	=	Expansion factor for ethane for conversion
γ	=	range of 0.0-0.65
t	=	Residence time, (S)
$\int$	=	Density of the mixture @ any point in the reactor, (kg/m³) = $\dfrac{M_{WA}*\int}{RT\ (1+8x)}$
γ	=	Viscosity of ethane at inlet condition g = 2.4 x 10⁻⁵ (kg/m.s).

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