

Dynamic Simulation of Industrial Reformer Reactors

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ABSTRACT

This research focused on mechanistic kinetic model for the upgrading of the low component fractions petroleum naphtha (generic) of Nigerian reformer reactors. Four main constituent lumps were considered in the synthesis of the rate equations that describe the mass action velocity for the depletion processes of the feed in the reactors. The reactors under study is a continuous catalyst reforming stacked reactors, one reactor on top of the other with gravity mobile flow of catalyst from reactor 1, 2 and 3. The plant design brings to the barest minimum the deactivation of catalyst by the precursors of the reactions. Energy and component mass balance approach were adopted to develop the dynamic model for the research. The models were parameterized by benchmarking against industrial reformer plant data. The equations are non-linear intractable partial differential equations describing the dynamics of the reactors are very stiff to solve. The resolution of the model equations depends on the obtaining the values of the activation energies for the four lumps chemical species reactions. A constrained optimization which considers the minimization of the sum of the squares of difference of the objective function was carried out by integrating the differential equations numerically. The dynamic equations were converted into a set of ordinary differential equations by applying numerical method of lines (MOL) of finite difference approximation of the spatial derivatives, and the equations integrated with respect to time. The dynamic equations were resolved using the mat-lab ode solver-23. The comparison of deviations of dynamic model with the steady state model results showed naphthene 2.2 percent, paraffin 0 percent, aromatic -1.6 percent and temperature 0.13 percent, which are consistent with the steady state values of naphthene 3.7 percent, paraffin 0.6 percent, aromatics 1.2 percent and temperature 1.03 percent. A sensitivity analysis carried out on the open loop of the reactor system showed that the reactor temperature, pressure and activity of the catalyst are the major variables for striking a balance economically.

Keywords: *Dynamic Simulation, Continuous-Catalyst-Stacked-Reactor, Constrain optimization, Four-Lump Kinetics.*

1. INTRODUCTION

Global environmental legislation banned the use of lead compound as an additive for boosting antiknock properties of premium fuel. Coupled with this stricter environmental regulation, there has been a consistent increase in the demand for higher fuel efficiency standards of engines. These require the use of higher compression ratio in engines, and therefore motor fuel with an even greater octane number. These considerations have continually forced scientist and the refineries toward research producing high-octane-number products from the catalytic reformer reactors. This can be achieved by reforming the petroleum naphtha under more severe conditions maintaining optimal process parameters.

Gasoline is a valuable finished product of petroleum at about 19th Century. The demand became great that many new oil fields were discovered and fundamental improvements in refining methods developed through research. Therefore, gasoline served as the fuel for internal combustion engines and is mostly rated by its

volatility and antiknock qualities. Volatility is a measure of the ease of vaporization of gasoline in the carburetor while the measure of antiknock performance of gasoline is known as its octane number.

Tetra-ethyl-lead (H_2CC) Pb_4 compound was the most important additive used in gasoline blending processes. It greatly increases the octane number but has been found to contaminate the exhaust gases with poisonous lead oxides and renders refiners impotent if contacted carelessly. This primarily led to research of upgrading low petroleum fractions of which catalytic reforming processes was developed.

Catalytic reforming is a refining process that uses adequate operating conditions and appropriate catalysts to convert low quality naphtha in the presence of hydrogen into high grade motor fuels, as well as high yields of aromatic hydrocarbons and liquefied petroleum gas. There are a number of reforming processes in use but the major differences between them are in the composition of the catalyst used in the reactor and the methods of regenerating the catalyst. The feed for catalytic reforming

is naphtha (generic). The boiling range of the feed varies according to the process and yields needed.

The catalytic reforming process is one process that converts gasoline-boiling range low-octane hydrocarbons from paraffin to iso-paraffin and to naphthene, the naphthene are changed to aromatics without changing carbon numbers in the molecule. These yields, that is iso-paraffin and aromatics are high-octane gasoline components required to fuel automobiles. The choice of certain process parameters affect the performance of the reactors such a dehydrogenation are endothermic and very fast which cause an immediate drop in the reactor temperature. Preferably, the adiabatic stacked plug flow reactor is frequently used with intermediate heat exchangers between the reactors.

The research was based on fundamental principles of reaction engineering and reactor hydrodynamics. The model shall predict the dynamics and performances, particularly the product yield compositions for a wide variety of designs and operating conditions. The research focused on a gas-solid reaction system i.e heterogeneous catalytic reaction. The modeling of the complex chemical reactions occurring on the surface of the catalyst is the most intricate part of the overall modeling effort. Appropriate kinetic modeling of these reactions are imperative to achieving the desired prediction accuracy of the model by assuming a homogeneous phase..

A good number of different approaches of varying degrees of sophistication have been developed in the past to model the reforming chemistry. Literature show that Smith (1959), Krane et al (1960), Schulman and Sinfelt (1981), Kmak, (1972), Kuo and Wei (1969), Marin and Froment (1982), Ramage (1987), De Pauw and Froment (1974), were the very earlier researchers that delved into the study of naphtha (gasoline) catalytic reforming. In order to reduce the complexity of the model to a manageable level the number of chemical components are assigned to a smaller set of kinetic lumps, each composed of chemical species grouped together. The first significant attempt at de-lumping petroleum naphtha into different constituents was attempted by Smith (1959). He considered naphtha feed to consist of three basic components namely paraffins, naphthenes, and aromatics. Also Smith (1959) kinetic catalytic analysis evidently assumed a homogenous pattern, when in essence the process is heterogeneous and constant volume operation. He also assumed that three hydrocarbon classes has the same number of carbon atoms.

Bommannan et al (1989) employed the simplified model of Smith (1959) in some recent reformer modeling work. In a more extensive attempt to model reforming reactions of whole naphtha, Krane et al (1960). recognized the presence of various carbon numbers from C_6 to C_{10} as

well as the difference between paraffins, different components.

Kmak (1972), presented the first endeavour to incorporate the catalytic nature of the reactions by deriving a reaction scheme with Hougen-Watson Langumuir–Hinshelwood type of kinetics. Rate equations derived from this type explicitly account for the interaction of the chemical species with the catalyst. In another notable effort, Ramage, et al (1987), developed a detailed kinetic model based on extensive studies of an industrial pilot-plant reactor. The Kmak (1972) model was later refined by Marin and Froment (1982), who presented the reaction network for the whole naphtha fractions from C_5 to C_{10} . The network includes 23 pseudo components and used Hougen-Watson-type rate equations. Marin and Froment, (1982) and Van Trimpont et al (1988), also conducted separate studies on C_6 and C_7 a carbon-number fractions, respectively and developed the corresponding Hougen-Watson-type rate equations. Various possible reaction paths way and mechanisms, were systematically evaluated before choosing the one that best fit the experimental data on a laboratory scale reactor.

The lumping criterion carried out by Kuo and Wei (1969), was used to defining kinetic lumps for the complex reaction system. Kuo and Wei (1969), incorporated the reaction pathways and rate equations of Marin and Froment, (1982) and Van Trimpont (1988), mechanistic insight to a maximum extent within the model. It is therefore expected that the model got a better prediction capabilities.

In a more extensive attempt to model reforming reactions of whole naphtha, Krane et al (1960), recognized the presence of various carbon numbers from C_6 to C_{10} as well as the difference between paraffins, different components. Kmak (1972), presented the first endeavour to incorporate the catalytic nature of the reactions by deriving a reaction scheme with Hougen-Watson Langumuir–Hinshelwood type of kinetics. Rate equations derived from this type explicitly account for the interaction of the chemical species with the catalyst.

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before choosing the one that best fits the experimental data on a laboratory scale reactor.

Taskar and Riggs (1997) catalytic model assumed a quasi-state, mole and energy balance equations, constant catalyst activity, axial and radial dispersion effects were negligible. The uncertainty associated with diffusion effects in the catalyst pellets was lumped into kinetic rate parameters. The steady state variation of the chemical species and temperature were integrated through each reactor bed.

Wordu and Oboho, (2003) recognized the three chemical species paraffins, naphthenes and aromatics. They further adopted a material balance approach with respect to naphthenes, paraffins, aromatics, hydrogen gas and enthalpy changes for the four reactions taking place with respect to space-time. They further assumed that for an industrial plug flow reactor operating adiabatically, quantity of heat input q and heat output q is equal to zero. The results obtained were in agreement with the industrial plant data obtained from Nigerian reformer reactor, Alesa-Elеме, Port Harcourt.

Ferchneider and Mege (2004) investigated on fixed bed reactors with a single fluid phase for reaction processes catalyzed by a solid phase. They made use of a one-dimensional model and separately resolved the equation of conservation of mass and energy, and momentum. The calculation was based on the multi-dimensional model and the simultaneous resolution of the local conservation equations. The reactors they assumed to investigate was distinguished by their annular geometry and radial circulation of the Feedstock. The flow of streams in the reactors was assumed to be axsymmetric. They further carried out calculation of the hydrodynamics (mean velocities, pressure), thermal and mass fields

(concentration of each species) serves to identify the influence of the internal components in two industrial reactors geometries. Finally, the quantity of coke formed and deposited on the catalyst surface, was calculated by the model, and reveals potential areas of weakness in the research.

Hu and Zhu (2004), presented molecular modeling and optimization for the naphtha catalytic reforming process; the molecular modeling approach also focuses on producing high octane number gasoline by reforming reactions in three sequencing fixed bed reactors. They made use of the naphtha consisting of molecules from C_5 to C_{10} including paraffin iso-paraffin, naphthene and aromatic. The molecular kinetic network consisted of paraffin cracking, naphthene side-chain cracking, aromatic side-chain cracking, ring opening, ring closure, paraffin isomerizations, dehydrogenation and hydrogenation. Based on this reaction network molecular model for catalytic reforming was formulated. A process optimization was performed for feed temperature and pressure under constraints such as benzene content, aromatic content, and RON (Research Octane Number) limitations. This more recent innovation on catalytic reforming of low boiling point naphtha Feed fraction gave a final product with the highest profit with respect to benzene and aromatic contents and RON value. The application of molecular modeling to optimization in the process control gave significant benefits. Kubo (2004) studied on the analysis of catalytic reforming

2. PROCESS AND CHEMISTRY

The process and chemistry are complicated reactions occurring in series-parallel to each other, forming the rather complicated overall reaction scheme (Weismann, 1983) indicated in figure 1.

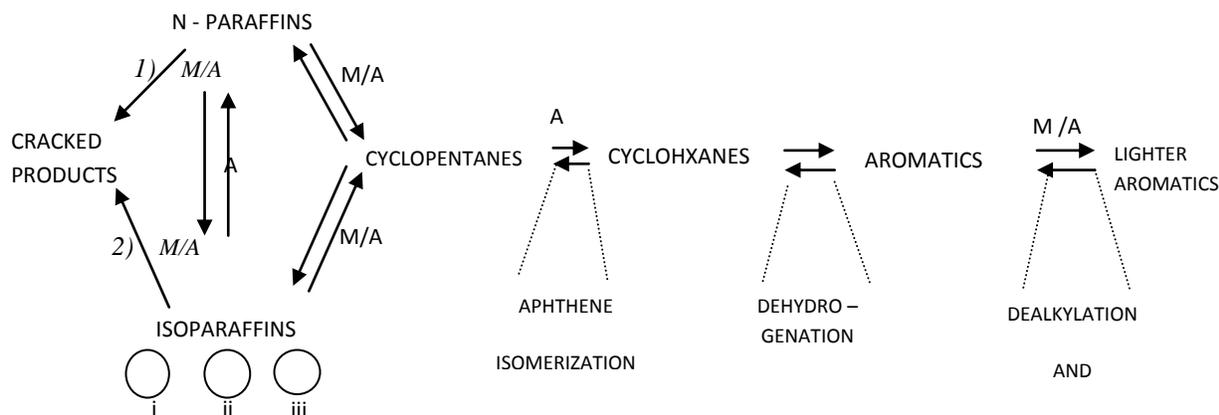


Fig.1: Overall Feed conversion scheme in the reactors

Predominant active sites: A = acid, M = metal. *i* = hydrocracking and demethylation (m); *ii* = paraffin isomerization; *iii* = dehydrocyclization. Gates, Katzer, Schuit, (1979), gave the main reforming reactions.

3. MATERIALS AND METHODS

Appropriate data were determined by estimation using relevant mathematical models and some were obtained

from literature. The kinetic parameter materials for applications in the dynamic model resolution are presented in Tables 1, 2, 3, 4 and 5.

Table 1: Kinetic Parameters A_0 , K_p , K_f , K_c and ΔH_R for the reactions

Reactions 1, 2, 3 & 4	Parameters		
	Pre-exponential constant, A_0	Equilibrium Constant, K_p	Heat of Reaction, ΔH_R KJ/Kmol of H_2 liberated
Conversion of naphthenes to Aromatics	1.19×10^8	$1.0404 \times 10^6 \exp(46.15 - 212700/RT)$	70928
Conversion of paraffins to naphthenes	4.170×10^{13}	$9.869 \times 10^3 \exp(36950/RT - 712)$	- 44185
Naphthenes cracking	4.5881×10^{18}	-	- 51860
Paraffins cracking	4.5881×10^{18}	-	- 51860

Table 2: Equilibrium Constants, K_p

Reactions	Parameters
1, 2, 3 and 4	Equilibrium constant, K_p
Conversion of naphthenes to aromatics	$1.0404 \times 10^6 \exp\left(\frac{46.15 - 212700}{RT}\right)$
Conversion of paraffins to naphthene	$9.869 \times 10^3 \exp\left(\frac{36950}{RT} - 712\right)$
Naphthenes hydrocracking	-
Paraffins hydrocracking	-

Table 3: Heat of Reactions, ΔH_R

Reactions	Parameters
1, 2, 3 and 4	Heat of Reactions, ΔH_R , kJ/Kmol of H_2 liberated
Conversion of naphthenes to aromatics	70928
Conversion of paraffins to naphthene	- 44185
Naphthenes hydrocracking	- 51860
Paraffins hydrocracking	- 51860

Table 4: Arrhenius Constants or Pre-exponential Constants, A_0

Reactions	Parameters
1, 2, 3 and 4	Arrhenius Constants, A_0
Conversion of naphthenes to aromatics	1.19×10^8
Conversion of paraffins to naphthene	4.170×10^{13}
Naphthenes hydrocracking	4.5881×10^{18}
Paraffins hydrocracking	4.5881×10^{18}

Table 5: Steady State Model Predictions and Plant data Reformate Yields, Mole %

Components	Plant Data	Model Predictions
Naphthenes, N	0.12	0.1156

Aromatics, A	0.53	0.5365
Paraffins, P	0.35	0.3479
hydrogen, H ₂	8.0	8.0
Temperature, T	780 K	772 K

4. REACTION RATE EQUATION

The reaction rate equation gives an expression for the rate of transformation or depletion of a reactant or formation of desired product in a given chemical process. The rate of transformation, conversion or disappearance of a reactant *i* to form the product *j* through the reforming reactions 1 to 4, $(-r_{i,j})_{1 \rightarrow 4}$, (feed converted per unit Fresh Feed per unit time) in the reformer reactor maintaining a first-order elementary reactions is given by:

$$(-r_{i,j}) = K_{i,j} y_i^n \phi_{i,j} \quad (1)$$

w.r.t. mole fraction of reaction species

Since catalyst deactivation is negligible, the function $\phi_{i,j}$ is therefore dropped in the rate model. The model is then written w.r.t partial pressure of the components in the vapour phase in the reformer reactors.

$$(-r_{i,j}) = K_{i,j} P_i^n \quad (2)$$

w.r.t. rate of depletion of feed component *i*,

we have,

$$(-r_{i,j}) = \left(\frac{dN_i}{dt} \right)_{1 \rightarrow 4} = K_{i,j} P_i^n \quad (3)$$

where,

$K_{i,j}$ = rate constant for the reaction process *i,j* (s⁻¹)

$(-r_{i,j})$ = rate of reaction

y_i^n = mass fraction of the Feed component *i*

N_i = mole fraction of the feed components *i*

n = order of reaction *i,j*

$\phi_{i,j}$ = deactivation function

But, since catalyst deactivation is negligible, the function $\phi_{i,j}$ is therefore dropped or eliminated in the rate model. The model is then w.r.t partial pressures of the components in the vapour phase of the reaction mixtures.

The dependence of the reaction rate constant on temperature is given by the popular Arrhenius equation as:

$$K_{i,j} = A_{oi,j} e^{-E_{i,j}/RT} \quad (4)$$

where,

$K_{i,j}$ = Rate constant for the four reactions

$A_{oi,j}$ = Frequency or pre-exponential factor or constant.

$E_{i,j}$ = Activation energy of the reactions

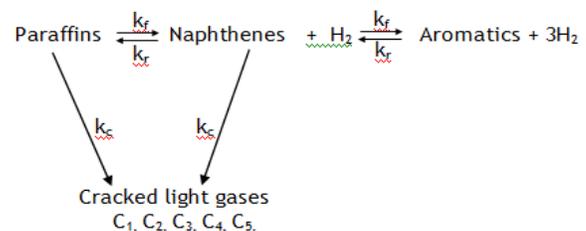
R = Universal gas constant

T = Absolute K

The introduction of this expression into the model equation makes the model equation intractable non-linear equations and difficult to solve for the reaction rate constants $K_{i,j}$ analytically. Therefore appropriate numerical method will be applied.

5. KINETIC LUMPS

Oboho, (2005) posited that the reactions taking place in the reactors follow Lumping Scheme kinetics as shown in Figure 2. below:



Lumping model of reforming reactions posited by Oboho, (2005)

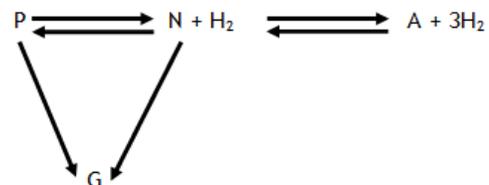


Figure 2. Composite Four-lump reforming reactions Oboho, (2005)

P – Paraffins, N - Naphthenes, A - Aromatics, H₂ - hydrogen, G – Gases, K_c - rate constant for hydro-cracking, K_f - rate of forward reaction, K_r - rate of reverse reaction.

Four fundamental reactions are identified in this unique lumping scheme kinetics. The derivation of the rate equations from the scheme is very crucial for research. The rate equations will be derived with respect to the components of the feed naphtha which are Paraffins, Naphthenes, Aromatics, Hydrogen and Cracked Gases. These are the constituents of the lumping scheme considered for the research. Smith,(1959), and Oboho, (2005) also recognized four major reactions associated with the rate equations respectively. They are:

Dehydrogenation of naphthenes to form aromatics and hydrogen

Hydrogenation of aromatics to form paraffins and vice versa

Hydro-cracking of naphthenes to gases

Hydrocracking of paraffins to gases and Temperature effects

6. DYNAMIC MODEL

The following derivations are essential for the study of a typical plug flow reactor system. The equations serve as the reactor equation for a reformer reactor. Mathematically, the plug flow reactor equation is stated thus,

$$\frac{V}{F_{AO}} = \int \frac{1}{(-R_A)} d\alpha \quad (5)$$

But,

$$F_{AO} = v_o C_{AO} \quad \text{i.e.} \quad \frac{m^3}{\text{sec}} \cdot \frac{\text{moles}}{m^3} \quad \text{in terms of concentration} \quad (6)$$

Substituting equation (6) into (5), to obtain,

$$\frac{V}{v_o C_{AO}} = \int \frac{1}{(-R_A)} d\alpha \quad (7)$$

Since,

$$\frac{V}{v_o} = \tau = C_{AO} \int \frac{d\alpha_A}{(-R_A)} \quad (8)$$

Differentiating equation (8) we have,

$$d\tau = C_{AO} \frac{d\alpha_A}{(-R_A)} \quad (9)$$

Equation (8) rearranges to,

$$\frac{d\alpha_A}{d\tau} = \frac{(-R_A)}{C_{AO}} \quad (10)$$

Equation (10) is the reactor equation in differential form for a plug flow regime which is required for the research.

Furthermore, considering the fact that the research focuses on the simulation of an existing reformer process reactor, appropriate model equations which takes into account the complex chemical species involved in the depletion process is paramount. Therefore, the reactor equation above was differentiated to obtain a summary equation (10) in differential form which when rearranged and on substitution of the $(-r_i)$ terms for the major component species i of the depletion processes and the energy or temperature effects will form the main fabric of a system of non linear partial differential model equations for the simulation of the reactor.

Consequently, stating the material balance for element of volume of a reformer reactor, using the figure 3 below, we have,

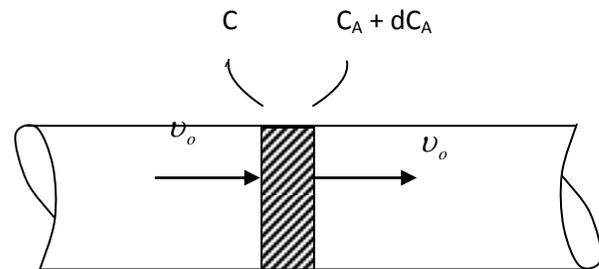


Figure 3: material balance for element of volume of a reformer reactor

Mathematically,

$$v_o C_A - v_o (C_A + dC_A) - (-r_A) dV = 0 \quad (11)$$

$$-v_o dC_A - (-r_A) dV = 0 \quad (12)$$

Rearranging equation (11) we have,

$$\frac{dV}{v_o} = - \frac{dC_A}{-r_A} \quad (13)$$

But,

$$d\tau = - \left(\frac{dC_A}{-r} \right) \quad (14)$$

where,

$$\tau = \frac{V}{v_o} \quad (15)$$

Differentiating equation (15) we have,

$$d\tau = \frac{dV}{v_o} \quad (16)$$

$$\frac{dC_A}{d\tau} = -(-r_A) \quad \text{or} \quad -\frac{dC_A}{d\tau} = (-r_A) \quad (17)$$

Equation (17) can be used to substitute the values of the $(-r_i)$ to obtain the various component balance for the reactions with respect to number of moles of species in the reactors. e.g

$$\frac{dN_{\text{N}}}{d\tau} = K_f P_N - \frac{K_{f1}}{K_{p1}} P_A P_{H_2}^3 + K_{p2} P_N P_{H_2} - \frac{K_{f2}}{K_{p2}} P_P + K_3 \frac{P_N}{P_T} \quad (18)$$

Where, the right hand side of equation (18) is the required kinetic model balance for the species with respect to naphthene.

7. BOUNDARY CONDITIONS FOR ELEMENT OF VOLUME OF A REFORMER REACTOR

Stating the material balance equation with respect to feed components naphthenes N_N , considering an element of volume per unit time, elemental volume of a plug flow reactor is applied to derive the material balance equation.

Thus,

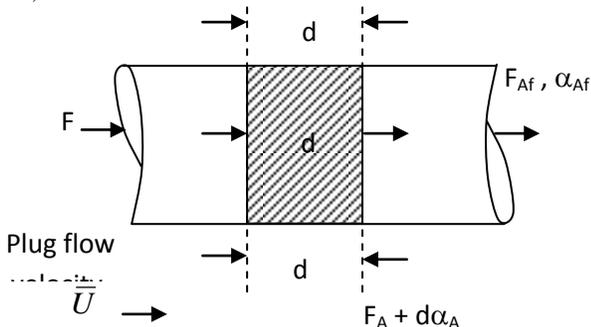


Fig. 4: A cross-section of the reactor depicting inflow of Feed and outflow of products

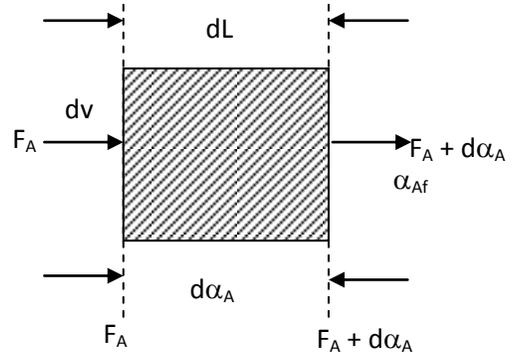


Fig. 5 : Differential volume element of the reactor for the conversion of the Feed

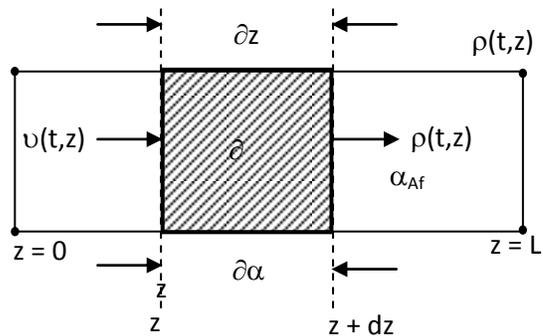


Fig. 6 : Boundary conditions for the reformer reactor model equations

8. REFORMER REACTOR MODEL (RRM)

The continuity and energy balance equations are principally used to develop mathematical models applied for the study of the reforming of the hydrocarbons components and temperatures effects of the process. The reforming reactors is modeled as adiabatic plug flow reactor operated in series and / or configured in series. The dynamic studies of the process plant using developed models will serve as major focus for this research of the reformer reactors.

9. MATERIAL/ COMPONENT MASS BALANCE

The process which the low hydrocarbons components (C_5, C_6, C_7, C_8) are transformed and/or upgraded into products (stable aromatics) will be monitored by the application of the law of conservation of mass, energy and momentum on a differential volume element of the reformer reactors of the Nigerian Petroleum Refinery located at Alesa-Elеме. But, the law of conservation of momentum has no relevance in this research; hence it is not emphasized in

this work. Therefore, the law of conservation of mass and energy prevail throughout the research.

The law of conservation of mass for reacting component is applied to a differential element of a reactor shown as this:

$$\begin{aligned}
 \left[\begin{array}{l} \text{Rate of flow of} \\ \text{component } i \text{ into} \\ \text{different element} \end{array} \right] &= \left[\begin{array}{l} \text{Rate of flow of} \\ \text{component } i \text{ out} \\ \text{different element} \end{array} \right] + \left[\begin{array}{l} \text{Rate of flow of} \\ \text{component } i \text{ due to} \\ \text{chemical reaction in} \\ \text{different element} \end{array} \right] \\
 &+ \left[\begin{array}{l} \text{Rate of accumulation of} \\ \text{component } i \text{ within} \\ \text{different element} \end{array} \right]
 \end{aligned} \tag{19}$$

Rate of flow of component i into differential volume element via boundary at L i.e. inlet of reactor (Kg s⁻¹)

$$= F_A \cdot \rho_i$$

Rate of flow of component i out of differential volume element via boundary at (L + dL) (Kg s⁻¹)

$$= F_{AO} (\rho_i + d\rho_i)$$

Rate of change of component i due to chemical reaction in differential volume element (Kgs⁻¹)

$$= (-r_i) \rho_i dv \equiv (-r_i) \rho_i AdL$$

Rate of accumulation of component i in differential volume element (Kgs⁻¹)

$$= \frac{\partial}{\partial t} (\rho_i AdL)$$

Bringing these terms together gives the material balance for the research work.

$$F_{AO} \rho_i = F_{AO} \rho_i + \partial \rho_i + (-r_i) \rho_i AdL + A \frac{\partial \rho_i}{\partial t} dL \tag{20}$$

Rearranging equation (5) gives

$$-A \frac{\partial \rho_i}{\partial t} - F_{AO} \frac{\partial \rho_i}{\partial L} = (-r_i) \rho_i A \tag{21}$$

but,

$$y_i = \frac{\rho_i}{\rho_{Feed}} \tag{22}$$

and,

$$U_F = \frac{F_{AO}}{A} = \frac{v_o}{A} \tag{23}$$

where,

U_F = Feed rate or superficial velocity of the Feed (moles/s)

v_o = Volumetric flow rate (moles/s)

A = Cross sectional area of the reactor

Substituting equation (23) into equation (21) gives

$$-\frac{\partial y_i}{\partial t} - U_F \frac{\partial y_i}{\partial L} = (-r_i) \tag{24}$$

$$\text{But, } U_F = \frac{L}{\tau} \tag{25}$$

$$\partial L = U_F \cdot \partial \tau \quad (26)$$

$$-\frac{\partial y_i}{\partial t} - U_F \cdot \frac{\partial y_i}{U_F \cdot \partial \tau} = (-r_i) \quad (27)$$

$$\frac{\partial y_i}{\partial t} + \frac{\partial y_i}{\partial \tau} = -(-r_i) \quad (28)$$

$$\frac{\partial y_i}{\partial t} + \frac{\partial y_i}{\partial \tau} + (-r_i) = 0 \quad (29)$$

$$-\frac{\partial y_i}{\partial t} - \frac{\partial y_i}{\partial \tau} = (-r_i) \quad (30)$$

The above equation is the dynamic model equation for the reformer reactors describing the fractional conversion of

reactant i or the yield of product j along the three reactors in series.

Subsequently, the dynamic model equations will be as follow:

$$-\frac{\partial y_i}{\partial t} - \frac{\partial y_i}{\partial \tau} = (-r_i) \quad (31)$$

$$\frac{\partial y_i}{\partial t} = -\frac{\partial y_i}{\partial \tau} - (-r_i) \quad (32)$$

10. ENERGY EQUATION

The temperature distribution along the reactor can be monitored by application of the law of conservation of energy on a differential volume element of the reactor as stated below:

$$\left[\begin{array}{l} \text{Rate of heat flow} \\ \text{into differential} \\ \text{volume element} \end{array} \right] = \left[\begin{array}{l} \text{Rate of heat flow out} \\ \text{of differential volume} \\ \text{element} \end{array} \right] + \left[\begin{array}{l} \text{Rate of heat absorbed / released} \\ \text{due to chemical reaction in} \\ \text{different volume element} \end{array} \right] + \left[\begin{array}{l} \text{Rate of heat accumulation} \\ \text{in differential volume} \\ \text{element} \end{array} \right] \quad (33)$$

The terms in equation (24) for a differential volume element of the reactor are as stated as follows:

$$\begin{aligned} \text{Rate of heat flow into differential volume} &= N_T C_p \rho T \\ \text{element (KJ/s)} & \\ \text{Rate of heat flow out of differential} &= N_T C_p \rho (T + dT) \\ \text{volume element (KJ/s)} & \\ \text{Rate of heat absorbed / released due to chemical} &= (\Delta H_r) (-r_i)_{1 \rightarrow 4} = \\ \text{reaction in different volume element (KJ/s)} &= \sum_{i=4}^n ((\Delta H_r) (-r_i)) \rho A dz \end{aligned}$$

where, n is the number of the reforming reactions.

$$\text{Rate of heat accumulation in differential volume} = \frac{\partial}{\partial t} (A \rho C_p dz) \\ \text{element (KJ/s)}$$

$$\begin{aligned}
 N_T C_p \rho T &= N_T C_p \rho (T + dT) + (\Delta H_r)(-r_i)_{1 \rightarrow 4} AC_p \rho T dz \\
 &+ \frac{\partial}{\partial t} (AC_p \rho T) dz
 \end{aligned} \tag{34}$$

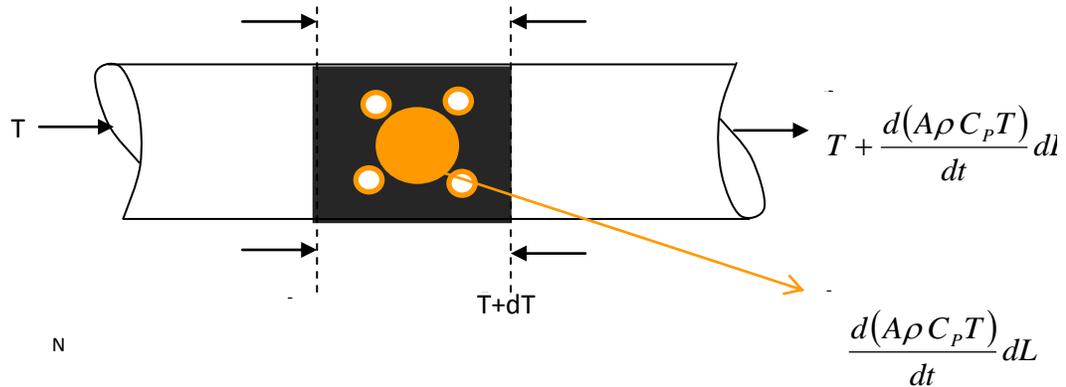


Fig. 7 : Differential volume of reformer reactor for energy balance

Discretization of feed components mole fractions into a system of ODEs

$$\begin{aligned}
 \frac{dN_{(0)}}{dt} &= - \left(\frac{N_{0,z+1} - N_{0,z}}{\Delta L} \right) - K_{f1} N_1 P_T + \frac{K_{f1}}{K_{p1}} N_2 N_4^3 P_T^4 \\
 &- K_{f2} N_1 P_T^2 N_4 + \frac{K_{f2}}{K_{p2}} N_3 P_T - K_3 N_1
 \end{aligned} \tag{35}$$

Naphthenes, N_N

$$\begin{aligned}
 N = 1 \quad \frac{dN_{(1)}}{dt} &= - \left(\frac{N_{1,z+1} - N_{1,z}}{\Delta L} \right) - K_{f1} N_1 P_T + \frac{K_{f1}}{K_{p1}} N_2 N_4^3 P_T^4 \\
 &- K_{f2} N_1 P_T^2 N_4 + \frac{K_{f2}}{K_{p2}} N_3 P_T - K_3 N_1
 \end{aligned} \tag{36}$$

Aromatics, N_A

$$N = 2 \quad \frac{dN_{(2)}}{dt} = \left(\frac{N_{2,z+1} - N_{2,z}}{\Delta L} \right) + \frac{K_{f1}}{K_{p1}} N_2 N_4^3 P_T^4 - K_{f1} N_1 P_T \tag{37}$$

Paraffins, N_P

$$N = 3 \quad \frac{dN_{(3)}}{dt} = - \left(\frac{N_{3,z+1} - N_{3,z}}{\Delta L} \right) - \frac{K_{f2}}{K_{p2}} N_3 P_T + K_{f2} N_1 N_4 P_T^2 - K_4 N_3 \tag{38}$$

Hydrogen, N_{H_2}

$$N = 4 \quad \frac{dN_{(4)}}{dt} = - \left(\frac{N_{4,z+1} - N_{4,z}}{\Delta L} \right) - 3 \left[\left(\frac{K_{f2}}{K_{p2}} N_2 N_4^3 P_T^4 \right) + K_{f1} N_1 P_T \right] - K_{f2} N_1 N_4 P_T^2 + \frac{K_{f2}}{K_{p2}} N_3 P_T - \frac{n_c}{3} K_3 N_1 - \frac{n_c - 3}{3} K_4 N_3 \quad (39)$$

Temperature, T

$$N = 5 \quad \frac{dN_{(5)}}{dt} = \left(-N_T C_p \frac{(T_{5,z+1} - T_{5,z})}{\Delta z} \right) + 3 \left(\frac{K_{f1}}{K_{p1}} N_2 N_4^3 P_T^4 + K_{f1} N_1 P_T \right) \Delta H_1 - \left(K_{f2} N_1 N_4 P_T^2 + \frac{K_{f2}}{K_{p2}} N_3 P_T \right) \Delta H_2 - \frac{n_c}{3} K_3 N_1 \Delta H_3 - \frac{n_c - 3}{3} K_4 N_3 \Delta H_4 \quad (40)$$

The PDEs were rewritten in terms of the index variable i.

For i = 1, ... I + 1, and
t = 1, 2, ... t + 1.

The partial differential equations were converted into a set of simultaneous ordinary differential equations. The ordinary differential equations have L as the independent variable and were subsequently solved by popular conventional means of Runge-Kutta numerical methods. It is however stated that the ODEs obtained using the method of lines (MOL) are very stiff and computational efficiency can be achieved by using an ODE – solver of the Matlab computer language. Above all, the brevity of the exercise (matlab) is quite plausible.

11. ESTIMATION OF KINETIC PARAMETERS

Essentially, the reactor model equations developed contain certain kinetic parameters which are specific for the Nigerian crude oil fractions for simulation of the reactor model equations. These are: rate constant, K_f for the forward reaction, and reverse reaction, K_r , frequency / pre – exponential factor, activation energies E_i for the kinetic lumps reactions, equilibrium constant, K_p , rate constant K_c for the naphthene and paraffin hydro-cracking reactions. These constants associated with the models were determined for resolution of these model equations using standard numerical technique. The determination of these constants are imperative as results obtained using literature values Oboho,(2005) showed conversion and yield results which gave maximum deviations of naphthene 2.45%; paraffins 28.5%; aromatics 15.4%; temperature 1.29% when compared with industrial plant data. These deviations can be attributed to the difference in the feed stock properties between that for which the kinetic parameters were generated and the one being

tested. Frantic effort was made to reach the industrial process plant for lifting the needed data for resolution of dynamic models and validation of the results.

The dependence of the reaction rate constant on temperature is given by the popular Arrhenius equation as:

$$K_{i,j} = A_{oi,j} e^{-E_{i,j}/RT} \quad (41)$$

where,

- $K_{i,j}$ = Rate constant for the four reactions
- $A_{oi,j}$ = Frequency or pre-exponential factor or constant.
- $E_{i,j}$ = Activation energy of the reactions
- R = Universal gas constant
- T = Absolute K

The introduction of this expression into the model equation makes the model equation intractable non-linear equations and difficult to solve for the reaction rate constants $K_{i,j}$ analytically. Therefore, appropriate numerical solution technique is adopted. The purpose for this exercise is to be able to estimate the activation energies, E_i for each of the kinetic lumps while literature values of $A_{oi,j}$ will be taken as posited by Bommaman, *et al.*, (1989).

The frequency or pre-exponential factor or constant is a function of the molarity of the reaction; but independent of the catalyst used. However, the values reported by Bommaman,*et al.*, (1989) were used in this work. The deactivation constant is lumped into the kinetic parameters.

Another parameter involved in the reversible reactions is the equilibrium constant K_p , which can be calculated from

thermodynamic considerations. There are no significant differences between the values reported in literature (Smith, 1959), (Bommannam, Srivastava, and Saraf, 1989), (Radosz and Kramarz, 1978), and (Van der Baan, 1980). Various values of the heats of the various reactions have been reported in literature. However, the values reported by (Smith, 1959) lie somewhere intermediate between other reported values and were used in this work. The values of the various kinetic parameters used in this work are given in Table 1, while the feed stock properties and reforming conditions are given in Table 2.

The activation energies depend on catalyst composition and in the absence of reliable experimental values can best be estimated from plant data. A constrained optimization procedure was used to find the activation energies that minimized the sum of the squares of the differences between calculated and experimental values of dimensionless reactor outlet temperatures and mole fractions of the various lumps from the third reactor simultaneously. Data for the mole fractions at the outlet of the first two reactors were not available and could not be used in the parameter estimation.

The dimensionless reactor temperature was defined as the ratio of the outlet temperature to the inlet temperature.

12. OBJECTIVE FUNCTION

The objective function is therefore defined as follows:

$$S = \sum_{i=1}^m \left[\sum_{i=1}^3 (N_{i\text{ calculated}} - N_{i\text{ plant}})^2 + \sum_{j=1}^3 (T_{out\text{ calculated}} - T_{out\text{ plant}})^2 \right] \quad (42)$$

where,

m = number of data sets used

$i = 1, 2$ and 3 for naphthene, paraffin and aromatic hydrocarbon respectively

$j = 1, 2$ and 3 for reactor 1, 2 and 3 respectively.

For assumed value of E_i , Equations 1 to 5 (i.e. the five model equations developed for the research) were integrated numerically using matlab ode – 15s solver for stiff ordinary differential equations to obtain calculated values of the yields of naphthene, paraffin and aromatic hydrocarbons at the third reactor outlet and the dimensionless reactor temperatures.

An improved estimate of the activation energies were obtained as described in (Senifeld and Lapidus, 1974) according to the model,

$$E_i^{(z+1)} = E_i^{(z)} - \gamma' \left. \frac{\partial S}{\partial E_i} \right|_{E=E^{(z)}} \quad (43)$$

The partial differentials $\frac{\partial S}{\partial E_i}$ were evaluated numerically

by varying the activation energy over a narrow interval (2 KJ Kmol⁻¹) about the current values and evaluating the correspondence changes in S . A suitable upper and lower bound were specified for the activation energies to avoid convergence to some spurious or false values.

The iteration process was discontinued or terminated when the difference between successive values of the sum of the squares of the deviations ($S^{(z+1)} - S^{(z)}$) became less than 10^{-9} . This was achieved after 96 iterations (Oboho, 2005). The values of the activation energies obtained were as follows: $E_1 = 174500$; $E_2 = 356460$; $E_3 = 394380$. The obtained values lie within the range quoted in (Smith, 1959) and (Bommannam, Srivastava and Saraf, 1989) research works. The values were used to predict the outlet temperature and product composition from the third reactor for conditions different from those used for parameter estimation.

13. SOLUTION TO DYNAMIC MODEL

A system of intractable partial differential equations (36), (37), (38), (39), and (40) were resolved numerically applying the method of lines (MOL). Essentially, this method converts the partial differential equations into a set of ordinary differential equations using a finite difference approximation of the spatial derivatives and integrates the set of equations with respect to time. First order forward difference was used to approximate the spatial derivatives. The reactor length was divided into 6 sections giving rise to 30 coupled ordinary differential equations. These were integrated applying Mat-Lab Ode 15s Solver (Simulink) for stiff ordinary differential equations.

14. RESULTS AND DISCUSSION

The analysis of the dynamic response of the open-loop system to changes in reformer reactor inlet temperature, feed-rate, and pressure are presented.

Temperature (Inlet)

The dynamic response of the yield of products and outlet temperature to step changes in the inlet temperature is presented in Figures

15. DISCUSSION

15.1 Effect of Temperature on The Dynamics of the Process

For a dynamic model simulation temperature of 812K, and pressure of 993 KPa, the mole fractions of naphthene increased from 0.1261 to a steady state value of 0.1372 after 40 seconds. Whereas, that of aromatics decreased from 0.5261 to 0.5149 after about 40 seconds, thus, stability is attained after 40 seconds in reactor 3.

Similarly, the temperature of the reactor 3 decreased from 768 to a steady state value of 771 after about 30 seconds to attain stability of the process.

Step increase in temperature from 812k to 833k for pressure of 993 kpa

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 3

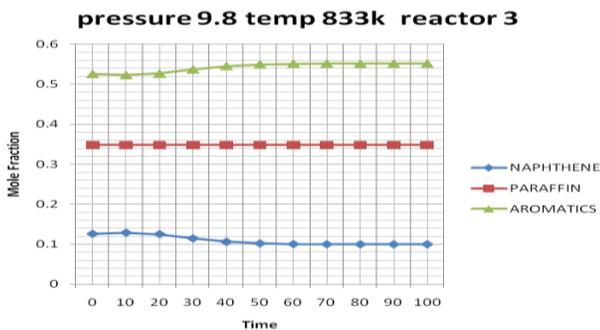


Fig. 9 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 2

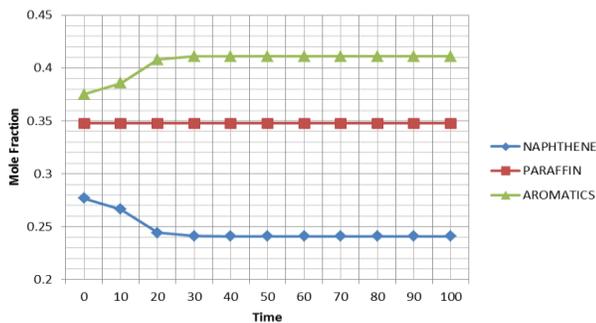


Fig. 10 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 1

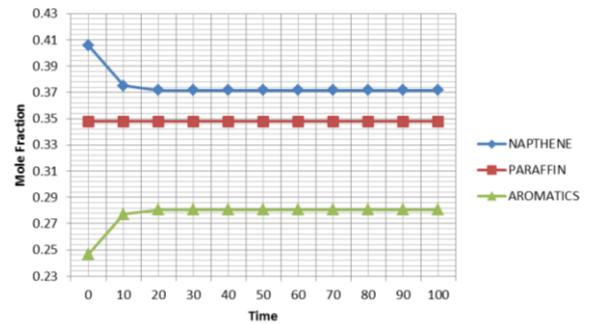


Fig. 11 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

16. EFFECT OF PRESSURE ON THE DYNAMICS OF THE PROCESS

Steady state simulation of reactor 3 using the parameters pressure 9.8 x 10^{1.35} KPa, indicates the mole fractions of naphthenes and aromatic with temperature 812K increased from 0.1261 to a steady state value of 0.1372, while that of aromatic decreased from 0.5261 to 0.5149 to attain stability of the process. The paraffins maintained a constant value of 0.3478.

The change of the mole fractions of the components with time is quite small as to be considered in industrial practice. Therefore, this is the required pressure range or plant design pressure. Consequently, this pressure value assists in the suppression of coke formation and does not encourage or facilitate equipment wear.

Table 4.19: Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 3

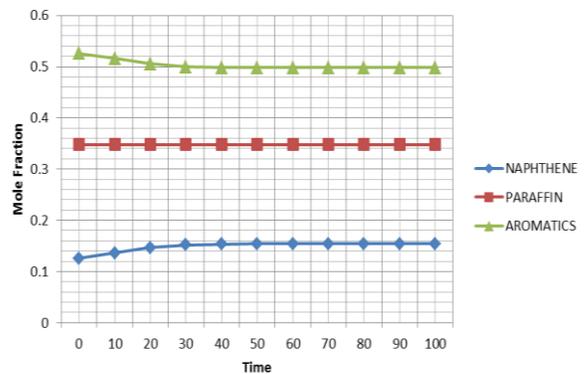


Fig. 12 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 2

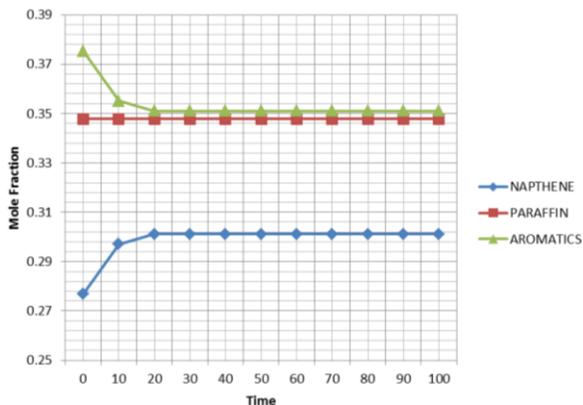


Fig. 13 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 3

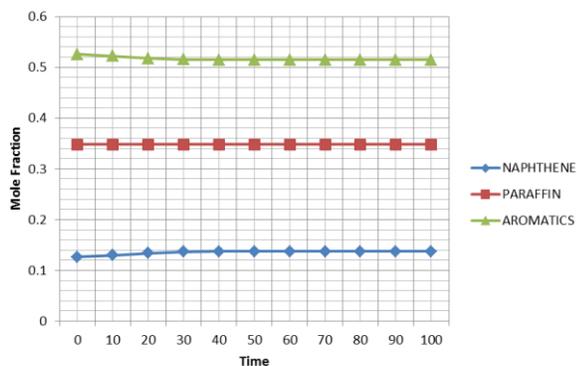


Fig. 14 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

Dynamic Model Simulation Results: Dependence of Mole Fractions of Naphthene, Paraffin and Aromatics with Time – Reactor 1

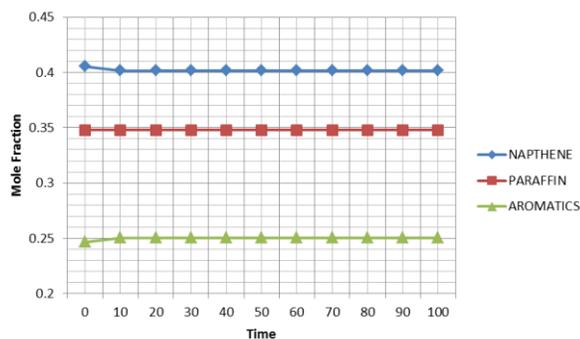


Fig. 15 : Dependence of mole fractions of naphthene, paraffin and aromatic on time

17. CONCLUSION

A rigorous mathematical model of industrial reformer reactors operated as a continuous catalyst reforming mode applying a detailed kinetic scheme was developed. The kinetic scheme was developed considering four idealize reactions by employing the lumping scheme method Wordu and Oboho (2003). The method of lumping the constituent hydrocarbons fractions into three major class of naphthene, paraffin and aromatic is for better management of the process, since the chemistry of the reforming reactions are quite complex from the point of view of its research.

The research employed naphtha (generic) naphthene, paraffin and aromatic as feed, while hydrogen which is the combined feed functions in two folds which serves to terminate the reactions of the double bonds formed in reactor 3, thereby suppresses the formation of coke through the reactions of the precursors of Sulphur, Nitrogen etc. during the process. Secondly, hydrogen facilitates transport of the liquid feed naphtha into the reaction zone, and maintains the required partial pressure of the reactors.

The intractable nonlinear model equations for the dynamic process were partial differential equations. These are difficult to solve analytically. Therefore, they were converted to ordinary differential equations utilizing the MOL i.e method of lines. This implies that the time were discretised using the finite difference method. This method aided the resolution of the equations through numerical method on application of Matlab Simulink ODEs-23 devices.

The dissertation utilizes the fundamental principles of energy and component mass balance approach to formulate the required dynamic model equations. The model equations were sectionalized into five (5) and make up to thirty (30) simultaneous ordinary differential equations. They were resolved numerically to obtain the expected results.

A simulation procedure which considers the steady state values as initial conditions were adopted to attain stability in each of the three reformer reactors 1, 2 and 3.

This is the basis of the present dissertation.

It is important to state that the study focused on continuous catalyst reforming unit, which operates as a mobile bed reactor, there is mobile flow of catalyst through the stacked reactors 1, 2 and 3.

This method of reforming is considered better than the semi-regenerative catalytic reforming unit which need to

be shut down when the activity of the catalyst is brought to a minimum.

The results of the dynamic models simulations were calculated values that were benched marked with the plant data of the Nigerian Port Harcourt Refineries process plant. The following deviations for dynamic and steady state models were obtained.

The dynamic model steady state simulations results for reactors, 3, 2 and 1 gave average value of naphthene 4%, paraffin 0.09%, aromatics – 6.8% and Temperature 1.83%. Whereas, that of steady state model results are given as ---Naphthene 3.7%, Paraffin 0.6%, Aromatics 1.2%, and temperature 1.03%.

From the results, it can be observed that the comparison of the dynamic model and steady states model are in agreement. The chemical species underwent a progressive conversion from initial conditions to final value of certain temperature specific for each of the reactors maintaining reversible endothermic reactions.

From the dynamic simulations of the reactors, it was observed that dynamics were from 10 to 80 seconds, for the reactions processes to stabilize. This can be attributed to the high activity of the mobile catalyst in the reactors.

Finally the overall conclusion of the dissertation is that the dynamic studies of the Nigerian reactors would be expected to result in significant economic benefits when the process is operated with pressure range of 7 to 14 * 101.35 KPa to achieve economic balance.

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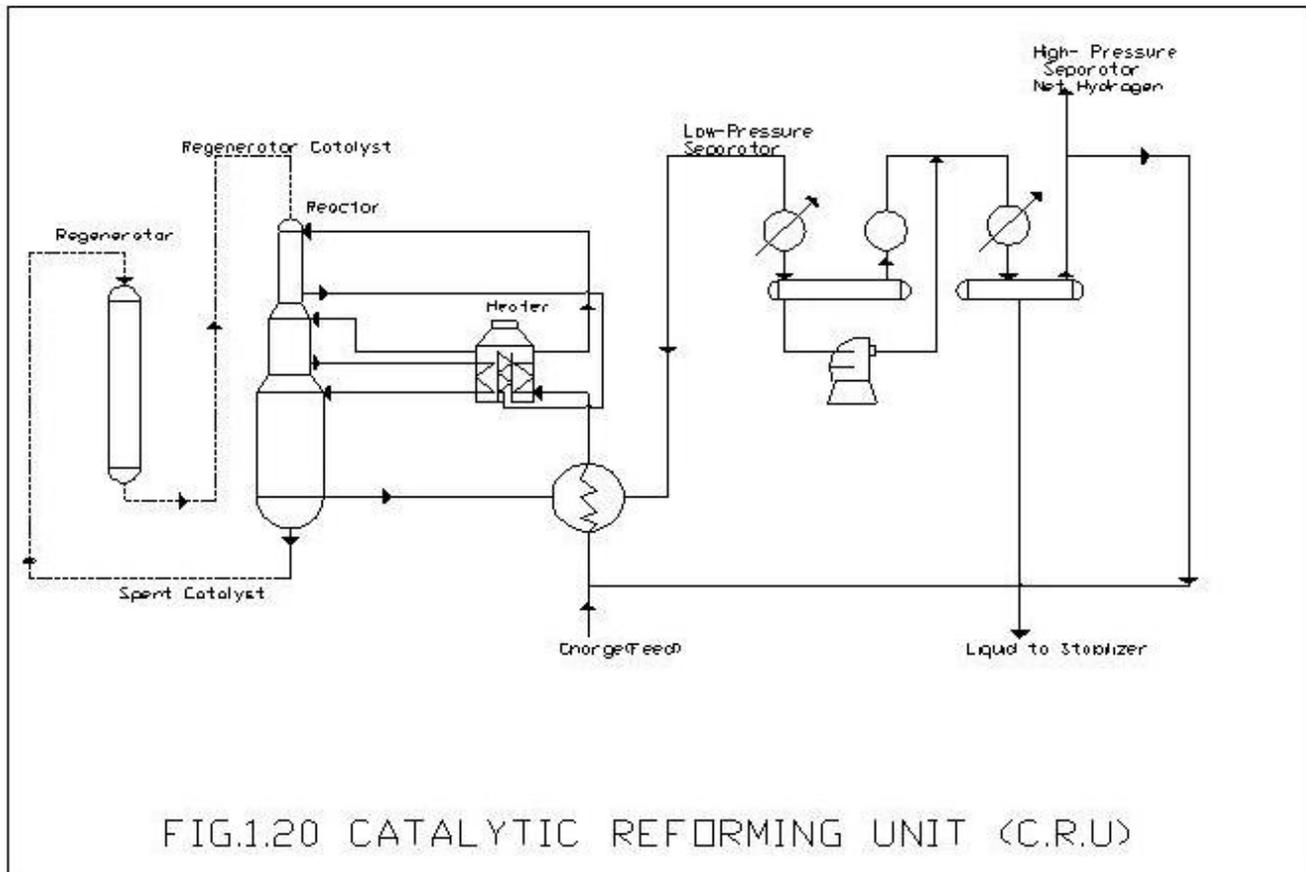
NOMENCLATURE

G'_F	=	Feed rate moles / second
$-r_i$	=	Moles of Feed components per moles of fresh feed per second $\frac{\text{moles } N_N}{\text{moles of fresh feed .sec}}$
N_{H_2}	=	Moles of Hydrogen in the reaction moles / second
\bar{U}_F	=	Superficial velocity of the Feed moles / second
$v_{o,F}$	=	Volumetric flowrate of Feed moles / second
A	=	Aromatics
A_c	=	Cross sectional areas in meters
C_A	=	Concentration of Aromatic in the Feed moles / second
C^B	=	Concentration of reacting species moles/dm ³
C_C	=	Concentration of products moles/dm ³
C_D	=	Concentration of products moles/dm ³
C_F	=	Total concentration of the Feed component moles / second
C_N	=	Concentration of Naphthene in the Feed moles / second
C_P	=	Concentration of Paraffin in the Feed moles / second
F_A	=	molar flow rate of pure A for fractional conversion α_A .
F_{Ao}	=	molar flow rate of completely unconverted Feed into the reactor moles/ second
G	=	Cracked gases
H	=	Hydrogen gas
K_p	=	Equilibrium constant for the reaction.
K_c	=	Rate constant hydrocracking reaction
K_f	=	rate constant for forward reaction
K_r	=	rate constant for reverse reaction
N	=	Naphthenes
N_A	=	Moles of Aromatic component in the Feed moles / second
N_N	=	Moles of Napthene component in the Feed moles / second
N_p	=	Moles of Paraffin component in the Feed moles / second
N_{total}	=	Total number of moles
P	=	Paraffins
Pa	=	Pressure in Pa or atmospheres
P_A	=	Partial pressure of aromatics Pa
P_N	=	Partial Pressure of Naphthenes Pa

P_p	=	Partial Pressure of Parafins Pa.
P_{total}	=	Reactor total Pressure or system total pressure
r_2	=	rate of backward reaction moles/seconds
R_A	=	rate of chemical reaction moles/second
r_1	=	rate of forward reaction moles/seconds
t	=	Element of time in second
T	=	Temperature °C or K
V_o	=	Volumetric flow rate m^3/sec
y	=	Space-time in the reactor S^{-1}
y_i or N_i	=	Mole fraction of component i, $N_A, N_p, N_N, N_{H_2}, N_{gases}$
z	=	Elemental distance in differential volume element meters
α_A	=	Fractional conversion at inlet of the elemental volume
$\alpha_A+d\alpha_A$	=	Fractional conversion at outlet of the elemental volume
α_{Af}	=	Final friction at the outlet of reactor
α_{Ao}	=	Fractional conversion at time = 0

APPENDIX

Continuous Catalyst Reforming Reactor Units



Continuous Catalyst Reformer Reactor Unit