

Development of Models for Fluid Catalytic Cracking Fluidized Bed Reactor Using Four-Lump Kinetic Scheme

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Abstract: Models' describing the steady state behavior of the fluidized bed reactor for catalytic cracking of gas oil has been presented. The four-lump kinetic scheme was used to describe the cracking reactions occurring in the reactor while the two-phase hypothesis comprising of the bubble and the emulsion phases of Kunii and Levenspiel was used to describe the fluidized bed models. The model equations consisted of sets of non linear first order differential equations and sets of quadratic equations. The differential equations were integrated numerically using the fourth order Runge-Kutta algorithm while the quadratic equations were solved using formulae method. Results predicted by the model were validated using data obtained from a operating plant, deviations of -21.99%, 9.85%, 5.27%, and 4.12% were obtained for the conversion of gas oil, yields of gasoline, gases and coke respectively. The results shows that plug flow- plug flow combination of the fluidized bed gave a higher conversion of gasoil than the plug flow-CSTR model. Sensitivity analyses showed that superficial velocity, bubble diameter, catalyst bed height, reactor temperature, catalyst-to-gasoil ratio and the diameter of reactor are important process variables that affect the yield of the products.

Keywords: Gasoil cracking, Four-lump kinetics, Reactor model

I. Introduction

Fluidization is the operation by which solid particles are transformed into a fluid like state through suspension in a gas or liquid. Cracking is the breaking down of higher hydrocarbon molecules into products of lower molecular weight. This process can either be thermal or catalytic. Catalytic cracking involves the breaking down of higher molecular hydrocarbon in the presence of a catalyst and high temperature. It can be carried out in a continuous, slowly descending layer of spherical catalysts or in a fluidized bed of a powdered or micro-spherical catalyst [1]. Fluid catalytic cracking (FCC) is one of the most important processes in any modern refinery; employed for the conversion of straight-run atmospheric gas-oil, vacuum residues, and other related heavy stocks, into a broad spectrum of products in the presence of a catalyst. The products obtained from an FCC unit include fuel gas, liquid petroleum gas, high-octane gasoline, light fuel oil, diesel oil, heavy fuel oil, etc. Fluid catalytic cracking unit consists of a reaction section and a fractionating section that operate together as an integrated processing unit. The reaction section has two reactors: (1) the riser-reactor where almost all the endothermic cracking reactions and coke deposition on the catalysts occur, and (2) The regenerator – reactor, where air is used to burn-off the accumulated coke on the catalyst. The catalyst regenerated process also provides the heat required for the endothermic cracking reactions in the riser- reactor. In the FCC unit, the catalyst and the feed enter the riser-reactor as a dense bed. The catalyst is pneumatically carried upwards by the dispersing steam and thereby vaporizing the gas-oil feed. It is during this period of conveying the catalysts that catalytic cracking of gas oil (feed) takes place through efficient and effective catalyst and gas oil intimate contact. The catalysts later becomes deactivated due to coke deposition on it and the spent-catalyst slide valve in the riser-reactor and enters the top of the regenerator. The major function of the regenerator is to oxidize the coke on the spent catalyst with oxygen to form carbon monoxide, carbon dioxide and water, thereby ultimately reactivating the catalysts. Within the entire refinery process, FCC process offers the greatest potential for increasing productivity, even a small improvement giving higher gasoline yields can result in a substantial economic gain due to the risen need of this desired products, gasoline. Thus, the economic incentive for a better understanding and modeling of the FCC process is immense [2].

The Fluid Catalytic Cracking Riser Reactor has been modeled as a single transport (plug-flow) reactor by several authors [1],[3],[4],[5],[6]. The FCC Riser reactor has also been modeled as two-phase fluidized bed reactor with the assumption that the catalyst in the emulsion phase are in equilibrium with the gases in the bubble phase, and the gases in the both phases are in tubular flow[7],[8],[9]. These authors modeled the both phases as tubular (plug-flow) reactors connected in parallel.

In the present study, the catalytic cracking fluidized bed is modeled as two-phase (bubble and emulsion); however, the gases in the emulsion phase is assumed to be at minimum fluidization velocity and totally mixed [10], while the gases in the bubble phase are above the minimum fluidization velocity and in plug-

flow mode, noting that the sum of the respective velocities in both phases equals to constant superficial velocity. The assumption of the minimum fluidization velocity in the emulsion is justified by the fact that the catalysts spend longer time (low velocity) in the emulsion phase with inherent high catalyst density than in the bubble phase. The models presented adopted the four-lump kinetic scheme proposed by Lee et al. [11] due to its advantage of predicting the amount of coke deposited on the catalyst used for heat integration in the system to simulate the riser reactor of a functional FCC unit in Nigerian Refinery.

II. Model Equations

2.1 Kinetic Model

The Figure 1 represents the reaction scheme for the four lump kinetic models for catalytic cracking of gas oil.

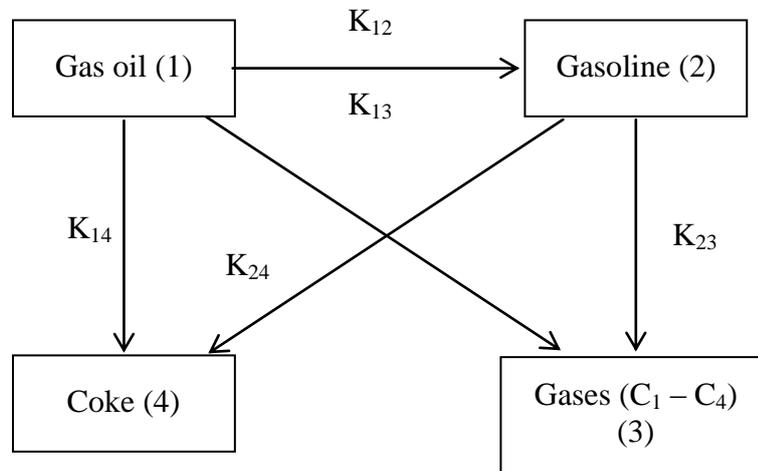


Figure 1: The four lump kinetic model

The rate of the reactions denoted by $(-r_{ij})$ is the mass of reactant converted per unit mass of reactant per volume of bubbles per unit time. It is expressed mathematically as:

$$(-r_{ij}) = k_{ij} y_i \phi \quad (1)$$

Where: r_{ij} is the rate of reaction of reactant (i) and product (j), k_{ij} is the rate constant, $y_i =$ mass fraction of species i, $\phi =$ deactivation constant, $I =$ reactant, $j =$ product.

The rate equation for gas oil, gasoline LPG and coke are written in terms of their mass fractions as follows:

$$(-r_1) = (k_{12} + k_{13} + k_{14}) y_1^2 \phi \quad (2)$$

$$(-r_2) = -[k_{12} y_1^2 \phi + (k_{23} + k_{24}) y_2] \phi \quad (3)$$

$$(-r_3) = -(k_{13} y_1^2 \phi + k_{23} y_2) \phi \quad (4)$$

$$(-r_4) = -(k_{14} y_1^2 + k_{24} y_2) \phi \quad (5)$$

where, k_{12} , k_{13} and k_{14} are the kinetic rate constant for the production of gasoline, gases and coke from gas oil. k_{23} and k_{24} are the kinetic rate constant for the production of gases and coke for gasoline respectively. ϕ is the deactivation constant, y_1 , y_2 , y_3 and y_4 are the mass fraction of gas oil, gasoline, gases and coke respectively.

2.3 The Reactor Model

Figure 2 shows a hypothetical representation of a two-phase fluidized bed reactor. The fluidized bed is modeled as a two-phased model with the bubble phase being modeled as a plug flow reactor and the emulsion phase as continuous stirred tank reactor (CSTR). The basis of modeling the emulsion phase as a CSTR dwells on the fact that there is violent motion of solids which leads to mixing in the gas phase [12].

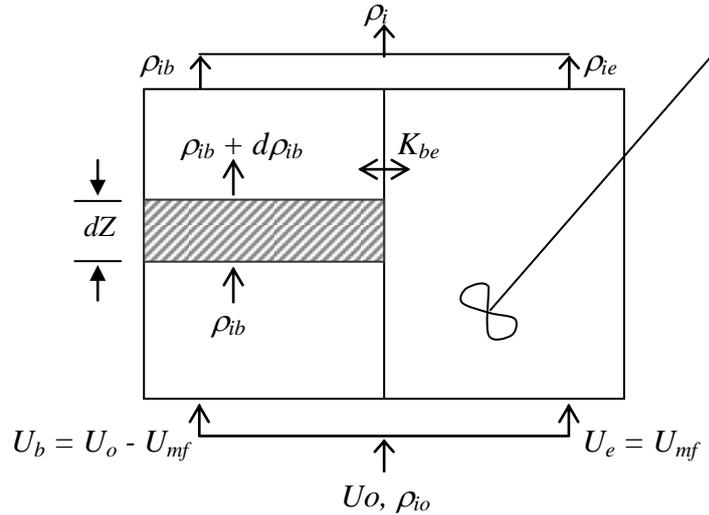


Figure 2: Two phase fluidize Bed

where, U_o is the superficial velocity, $\rho_i G$ is the density of gas oil, U_e is the velocity of the emulsion phase, U_{mf} is the minimum superficial velocity, U_b is the velocity of the bubble phase, ρ_{ib} and ρ_{ie} are the densities of the bubble and emulsion phases respectively, ρ is the total outlet density.

In the derivation of the mathematical models for catalytic cracking of gas oil to products, the following assumptions were made:

1. Feed entering the bed is at incipient velocity U_o , and is partitioned between the emulsion phase where the velocity is that of minimum fluidization, U_{mf} , and the bubble phase, where the velocity is $U_o - U_{mf}$.
2. Isothermal condition throughout the reactor due to the vigorous agitation of the bed.
3. The bubble phase has high gas velocity and contains no solid particle hence no reaction takes place therefore it is modeled as plug flow reactor.
4. The emulsion phase is modeled as a CSTR since there is total mixing due to the definite flow pattern of solids.
5. Reaction occurs in the emulsion phase since it contains solid catalyst particles.
6. Interchange of mass occurs between bubble and emulsion phases.
7. Solid particles are perfectly mixed and of uniform sizes.
8. Steady state conditions are assumed.

2.4 The Continuity Equation

2.4.1 The Bubble phase

The law of conservation of mass for a reacting component is applied on a differential element of the bubble phase of the fluidized bed to give:

$$\frac{-U_b dy_{ib}}{dl} = (-r_i) \epsilon + k_{be} (y_{ib} - y_{ie}) \quad (6)$$

Equation (6) represents the model equation for the bubble phase.

2.4.2 The Emulsion Phase

The law of conservation of mass for a reacting component is applied in the entire system of the emulsion phase of the fluidized bed to give:

$$y_{ie} = \frac{U_e y_{ieo} - (-r_i) \epsilon L + k_{be} y_{ib} L}{(U_e + K_{be} L)} \quad (7)$$

Equation (7) is the model equation for the emulsion phase.

Recalling the model equation of bubble phase, equation (6) and that of emulsion phase, equation (7) and representing them in dimensionless form by defining a dimensionless bed Z , we have:

$$Z = \frac{L}{L_F} ; \quad L = LFZ \quad (8)$$

where Z is the dimensionless bed height, L_F is the catalyst bed height. Differentiating equation (8) gives:

$$dL = LF dZ \quad (9)$$

Substituting equation (8) and (9) into equation (6) and (7) gives:

$$\frac{dy_{ib}}{dZ} = -(-ri) \frac{\varepsilon L_F}{U_b} - \frac{k_{be} L_F}{U_b} (y_{ib} - y_{ie}) \quad (10)$$

$$y_{ie} = \frac{U_b y_{ieo} - (-ri) \varepsilon L_F Z + k_{be} y_{ie} L_F Z}{U_e + k_{be} L_F Z} \quad (11)$$

Substituting the kinetic rate equation (2, 3, 4, 5) into both model equations (10) and (11), for the respective reactant and products, give:

$$\frac{dy_{1b}}{dZ} = -[k_{12} + k_{13} + k_{14}] y_{ib}^2 \frac{\phi \varepsilon L_F}{U_b} - \frac{K_{be} L_F}{U_b} (y_{ib} - y_{ie}) \quad (12)$$

$$y_{1e} = \frac{[k_{12} + k_{13} + k_{14}] y_{ib}^2 \phi \varepsilon L_F Z + K_{be} y_{ib} L_F Z}{U_e + k_{be} L_F Z} \quad (13)$$

$$\frac{dy_{2b}}{dZ} = [k_{12} y_{ib}^2 - (k_{23} + k_{24}) y_{2b}] \frac{\phi \varepsilon L_F}{U_b} - \frac{K_{be} L_F}{U_b} (y_{ib} - y_{ie}) \quad (14)$$

$$y_{2e} = \frac{U_e y_{2eo} - [k_{12} y_{ib}^2 + (k_{23} + k_{24}) y_{2e}] \phi \varepsilon L_F Z + K_{be} y_{2b} L_F Z}{U_e + k_{be} L_F Z} \quad (15)$$

$$\frac{dy_{3b}}{dZ} = [k_{13} y_{ib}^3 + k_{23} y_{2b}] \frac{\phi \varepsilon L_F}{U_b} - \frac{k_{be} L_F}{U_b} (y_{3b} - y_{3e}) \quad (16)$$

$$y_{3e} = \frac{U_e y_{3eo} + [k_{13} y_{ib}^3 + k_{23} y_{2e}] \phi \varepsilon L_F Z + k_{be} y_{3b} L_F Z}{U_e + k_{be} L_F Z} \quad (17)$$

$$\frac{dy_{4b}}{dZ} = [k_{14} y_{ib}^4 + k_{24} y_{4b}] \frac{\phi \varepsilon L_F}{U_b} - \frac{k_{be} L_F}{U_b} (y_{4b} - y_{4e}) \quad (18)$$

$$y_{4e} = \frac{U_e y_{4eo} + [k_{14} y_{ib}^4 + k_{24} y_{2e}] \phi \varepsilon L_F Z + k_{be} y_{4b} L_F Z}{U_e + k_{be} L_F Z} \quad (19)$$

2.5 Model Equations Parameters Evaluation

2.5.1 Mean Activity of the Catalyst, ϕ

The mean activity for mixed flow of catalyst in a fluidized bed reactor is given by Kunni and Levenspiel [1] as:

$$\phi = \frac{1}{1 + k_d \tau_{mean}} \quad (20)$$

But the dependence of the rate constant, k_d on reaction temperature follows the Arrhenius law.

$$k_d = k_{do} \exp\left(\frac{-E}{RT}\right) \quad (21)$$

The mean residence time, τ_{mean} of the catalyst in the reactor is given by:

$$\tau_{mean} = \frac{V_{reactor}}{G_o \text{ catalyst}} = \frac{\text{Volume of reactor}}{\text{Volumetric flow rate of catalyst}} \quad (22)$$

$$\text{But, } V_{reactor} = \frac{ARL_F Z}{F_{GR} CTO} \quad (23)$$

$$G_o \text{ catalyst} = \rho_s \quad (24)$$

where, ρ_s is the density of catalyst, FGR is the mass flow rate of gas oil, CTO is the catalyst to gas oil ratio.

\therefore The mean residence time becomes:

$$\tau_{mean} = \frac{\rho_s A_R L_F Z}{F_{GR} CTO} \quad (25)$$

Hence, substituting equations (21) and (25) into (20) yields:

$$\phi = \frac{CTO F_{GR}}{CTO F_{GR} + k_{do} \exp\left(\frac{-E}{RT}\right) \rho_s A_R L_F Z} \quad (26)$$

2.5.2 Specification of Fluidized Bed Parameters

Using the Davidson's theoretical expression for bubble-cloud circulation and the Higbie theory for the cloud-emulsion diffusion, the interchange coefficient between bubble-cloud and cloud-emulsion phases is given by Kunii and Levenspiel [1]:

$$K_{bc} = 4.5 \frac{U_{mf}}{d_b} + 5.85 \left(\frac{D^{1/2} g^{1/4}}{db^{5/4}} \right) \quad (27)$$

$$K_{ce} = 6.78 \left(\frac{\epsilon_{mf} D U_{be}}{d_b^3} \right)^{1/2} \quad (28)$$

The interchange between bubble- emulsion phases is given by rule of addition of two parallel resistances:

$$\frac{1}{K_{be}} = \frac{1}{K_{bc}} + \frac{1}{K_{ce}} \quad (29)$$

Bubble velocity U_b is related to the superficial gas velocity U_o , the velocity at incipient fluidization, U_{mf} and bubble d_b , by the Davidson model as:

$$U_b = U_o - U_{mf} + 0.711 (g d_b)^{1/2} \quad (30)$$

The rise velocity of the emulsion gas is given by:

$$U_e = \frac{U_{mf}}{\epsilon_{mf}} \quad (31)$$

2.5.3 Exit Mass Fraction

The exit mass fractions of the respective components are given by Dagde and Puyate [9]:

$$y_1 = \beta y_{1b} + (1 - \beta) y_{1e} \quad (32)$$

$$y_2 = \beta y_{2b} + (1 - \beta) y_{2e} \quad (33)$$

$$y_3 = \beta y_{3b} + (1 - \beta) y_{3e} \quad (34)$$

$$y_4 = \beta y_{4b} + (1 - \beta) y_{4e} \quad (35)$$

$$\text{where } \beta = 1 - \frac{U_{mf}}{U_o} \quad (36)$$

2.6 Materials And Method

2.6.1 Operating Parameters

The parameters were expected from the plant data from the FCC unit of the New Port Harcourt Refinery, Nigeria. Table 1 shows the physical properties of the feed and products while Table 2 shows the properties of the catalyst and also heats of reaction. The feedstock composition is shown in Table 3; the air and hydrocarbon physical properties were obtained from the API Data Book [13]. Table 4 shows the reactor dimensions.

Table 1: Properties of feed and products of FCC plant [14]

Components	API Gravity	Composition weight %	Flow rate
Gas oil feed	21.2	100	67.8
Fuel gas	–	5.4	3.66
C3 LPG	–	6.3	4.27
C4 LPG	–	10.7	4.27
Gasoline	60.0	45.9	31.12
Light Cycle oil	14.0	17.8	12.07
Bottoms	0.5	8.8	5.97
Coke	–	5.1	3.46

Table 2: Physical properties and heat of reactions of reacting species [3]

Parameters	Values
Hydrocarbons	
Vapour density, kgm ⁻³	9.52
Liquid density at 288K, kgm ⁻³	924.8
Specific heat of gas, Kj.kg ⁻¹ k ⁻¹	3.3
Specific heat liquid, Kj.kg ⁻¹ k ⁻¹	2.67
Heat of vapourization, Kj.kg ⁻¹ k ⁻¹	156
Vapourization temperature, K.	698
Catalyst	
Catalyst Bulk density, kgm ⁻³	975
Particle size, m	75 x 10 ⁻⁶
Mass flowrate of catalysts from Reactor to regenerator, kg/s	1729750

Table 3: Feedstock composition (Mass spectroscopic method) % mass [14]

Hydrocarbon	% Mass
Paraffins	35.4
Naphthenes	16.1
Aromatics	48.5

Table 4: FCC Unit dimensions [14]

Parameters	Value (cm)
Reactor length	22.9
Reactor diameter	2.9
Cyclone height	14.24
Cyclone diameter	1.5
Disengage height	24.49

The preheated values of the activation energy and the pre-exponential factor, E and ko of the four-lump kinetic scheme are shown in Table 5:

Table 5: Predicted values of activation energy and frequency of pre-exponential factor [11]

Reaction Path	Activation Energy	Pre-exponential factor
Gas oil to gasoline	66994	221.611
Gasoline to gases	83283	1263.611
Gas oil to coke	62121	10.4583
Gasoline to gases	54191	0.90417
Gasoline to coke	140008	2210.2778

3.6.2 Solution Technique

The model equations developed gave a set of four ordinary differential equations for the bubble phase and a set of four quadratic equations for the emulsion phase. The ordinary differential equations for the bubble phase were solved numerically by using fourth order Runge-Kutta method while the emulsion phase quadratic equations were evaluated using the quadratic formula adapted to the visual basic program. Since the gas oil is cracked into the various products, the mass fraction of gas oil at the inlet (L = 0) of the reactor is unity while the mass fraction of the products at the inlet are equal to zero. These initial boundary conditions are stated mathematically as:

$$Z = 0: y_{1b0} = y_{1e0} = 1 \text{ and } y_{2b0} = y_{2e0} = y_{3b0} = y_{3e0} = y_{4b0} = y_{4e0} = 0$$

where y_{1b0} and y_{1e0} are the inlet mass fractions of gas oil in the bubble end emulsion phases respectively; $y_{ib0} = y_{ie0}$ are the inlet mass fractions of the products in the bubble and emulsion phases respectively, with $i = 2,3,4$ as gasoline, gases and coke respectively.

III. Results And Discussion

Table 6 shows the comparison between plant yields and predictions from the model (Equations 12 – 19) for the (CSTR/Plug flow), indicating that the predicted data agree reasonably well with plant data and the plug flow-plug flow model [8]. The prediction of coke yield which is the major advantage of the four-lump kinetic scheme adopted in this model matches the plant data very closely. These results show deviations ranging from 4.118% to 21.992% for CSTR/plug flow model and 3.8% to 10% for plug flow/plug flow model adopted from Oboho et al. [8].

Table 6: Comparison of Model Predictions with Industrial FCC riser reactor yields

Parameters	Model Prediction		Plant Data	% Deviation.	
	Plug/ CSTR	Plug/ Plug		Plug/ CSTR	Plug/ Plug
Gasoline	0.4138	0.4131	0.4590	9.847	10.00
H/C Gases	0.2122	0.2620	0.2240	5.268	-3.800
Coke	0.0489	0.0483	0.0510	4.118	5.294
Gas Oil	0.3245	0.2863	0.2660	-21.99	-7.090

The fluidized bed (CSTR/plug flow) model predicted lower conversion of gas oil, high yield of gasoline and coke and low yield of gases as compared to the plug flow/plug flow model. The lower conversion of gas oil is in agreement with the inherent behaviour of fluidized bed reactors as compared to plug flow riser reactors.

Cheremisinoff and Cheremisinoff [15] substantiated this deviation as inherent in fluidized bed system where back mixing exist in the emulsion phase, the flow is somewhere between plug and total mixed flow. Also some portion of the gas oil (feed) may escape without contact with the catalyst due to channeling and by-passing effects. Hence, the amount of gas oil (feed) available for reaction with the catalysts in the emulsion phase is reduced, thus conversion of gas oil is apparently low. While the yield of gasoline and coke are high that of gases is low as compared to the plug flow/ plug flow fluidized bed model.

Figure 3 shows the variation of mass fraction of gas oil, gasoline, gases and coke along dimensionless bed height. It depicts a gas oil conversion of 67.55%, gasoline yield of 41.38%, hydrocarbon gases yield of 21.22% and coke yield of 4.89%.

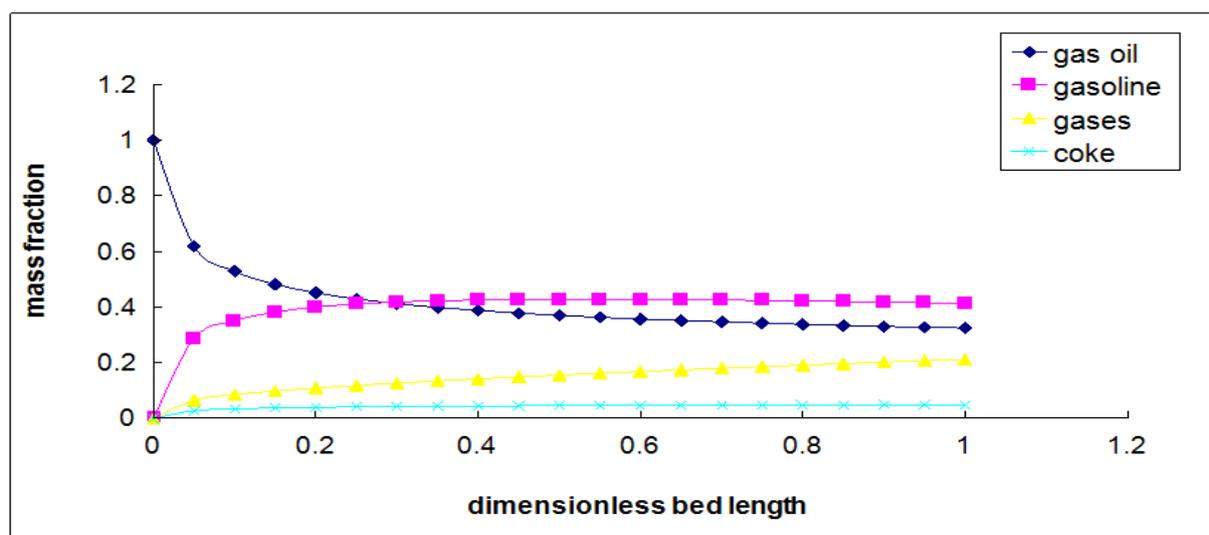


Figure 3: Variation of mass fraction of gas oil, gasoline, gases and coke along dimensionless bed height.

It is observed that the mass fraction of gas oil decreases along the bed height, while that of the cracked products increased along the bed height. Maximum gasoline yield of 42.85% was detected at a bed height of 7.89 meters, on getting to a bed height of 15 meters due to secondary cracking of gasoline. The yields of hydrocarbon gases and coke continuously increased along bed height.

3.1 Reactor Simulation

Plant performance can be optimized by choosing the optimal set of operating conditions obtained from a simulation model. Hence a sensitivity analysis is performed to determine the effects of certain process variables on the performance of the models developed.

3.1.1 Reaction Temperature

From Figure 4, increase in reactor temperature led to an increase in conversion (decrease in mass fraction of gas oil).

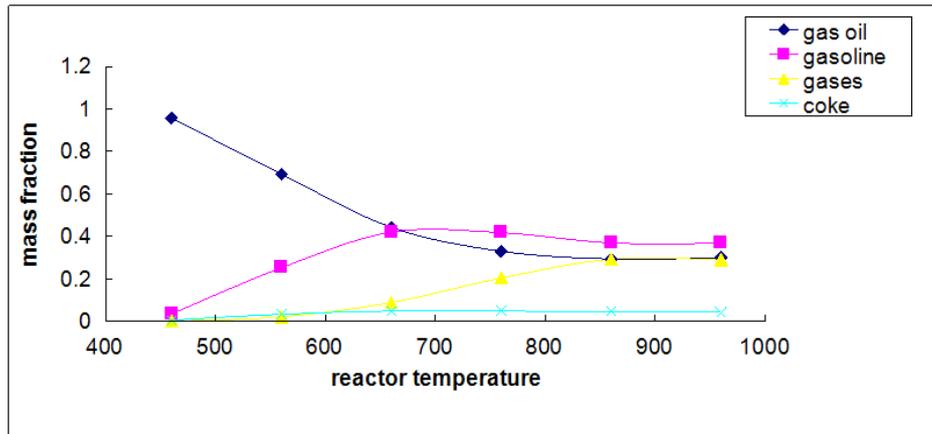


Figure 4: Variation of mass fraction of cracked components with reactor temperature.

At a reactor temperature of 860 K, the conversion increased to 70.61%, and then dropped to 69.97% at a reactor temperature of 960 K. The yield of gasoline increased with increase in reactor temperature to a maximum of 42.01% at 660 K. Further increase in reactor temperature resulted in a decrease in yield of gasoline which is due to secondary cracking of gasoline resulting to increased yield of light gases and coke.

3.1.2 Catalyst to Oil Ratio

Figure 5 shows the variation of mass fraction of reactant and products with catalyst to oil ratio. Increase in catalyst to oil ratio signifies an increase in the catalyst inlet flow rate. This provides more catalyst for the cracking reaction, increasing the availability of many active sites for the reaction.

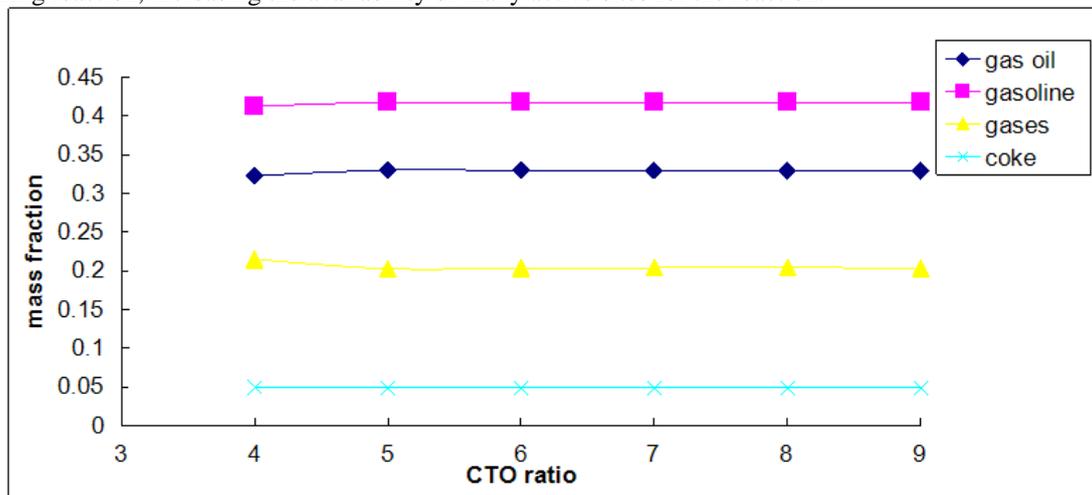


Figure 5: Variation of mass fraction of reactant and products with catalyst to oil ratio.

3.1.3 Superficial Velocity

Figure 4.4 shows the effect of the superficial velocity of gas oil on the conversion of gas oil, yield of gasoline, hydrocarbon gases and coke. Below a superficial velocity of 0.09 m/s, the program did not run successfully. Thus 0.09 m/s became the minimum superficial velocity value and turned out to give an optimum yield. It is seen that optimum product yield was attainable at a superficial velocity value of 0.09 m/s, with a gas oil conversion of 78.81%, yield of 40.28%, and 32.79% and 5.74% for gasoline, hydrocarbon gases and coke respectively. Above 0.09 m/s, the conversion of gas oil decreased and also the yield of the products reduced.

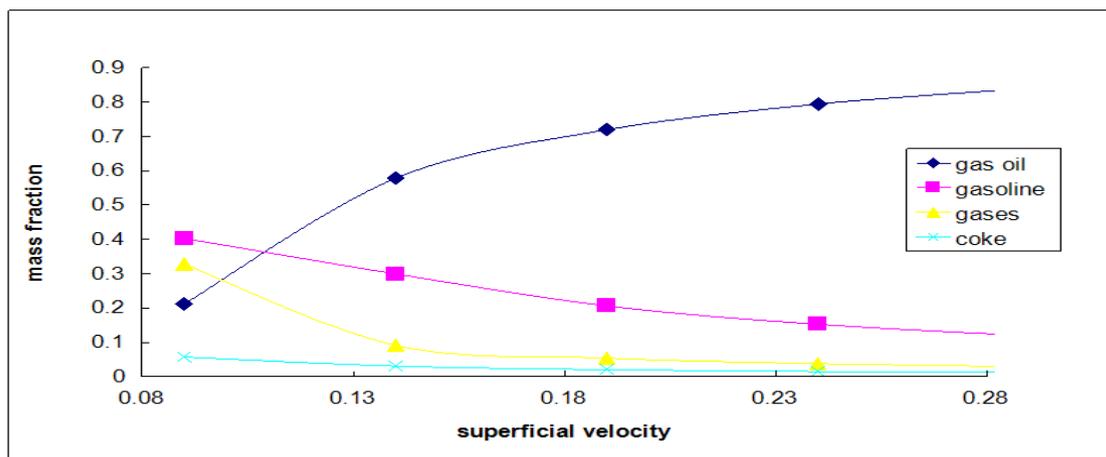


Figure 6: Variation of mass fraction of reactant and products with superficial velocity.

This was attributed to the fact that at higher superficial velocities, the residence time of the catalyst was low, resulting to low conversion of gasoil which was caused by channeling and by-passing effect inherent in fluidized beds at high superficial velocity [10].

3.1.4 Bubble Diameter

The plot on Figure 7 shows that an increase in bubble diameter increases the conversion of gas oil. The yield of gasoline increased to a maximum of 41.99% at a bubble diameter of 0.08 m. A decrease in gasoline yield was observed which is due to the slugging effect and the fact that an increase in bubble size causes the bubble to move upward in a piston-like manner, then disintegrates and rains down thereby creating a local space velocity different from the overall space velocity [15].

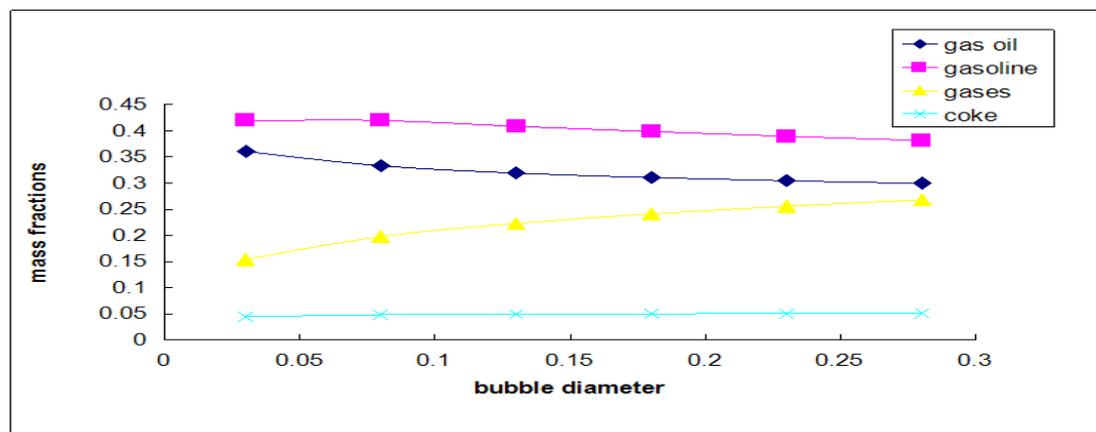


Figure 7: Variation of components mass fraction with bubble diameter.

IV. Conclusion

The fluidized bed model for the catalytic cracking of gas oil using the four-lump kinetic scheme has been successfully developed. The results predicted by the model were validated using data obtained from an operating plant, deviations of -21.99%, 9.85%, 5.27%, and 4.12% were obtained for the conversion of gas oil, yields of gasoline, gases and coke respectively. The high deviation in gasoil conversion can be attributed to by-passing in the bubble phase and channeling in the emulsion phase of the fluidized bed.

However, this work, in which the emulsion phase of the fluidized bed was modeled as a continuous stirred tank reactor, was performed to provide a means of comparison with that in which the emulsion phase was modeled as a plug flow reactor.

The results shows that plug flow- plug flow combination of the fluidized bed gave a higher conversion of gasoil, 73.99% [9] than that of this model; the plug flow-CSTR model, 65.55%. This agrees with the postulate that a plug flow reactor gives a higher conversion than a CSTR. However, the yield of gasoline was higher in the plug flow-CSTR combination.

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