

**KINETIC MODELING AND SIMULATION
OF FLUID CATALYTIC CRACKING
UNITS**

By

PRABHA KIRAN DASILA

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(Prabha Kiran Dasila)

DECLARATION

I, Prabha Kiran Dasila hereby declare that the work presented in this thesis titled **“KINETIC MODELING AND SIMULATION OF FLUID CATALYTIC CRACKING UNITS”** is original and my own work carried out at **University of Petroleum and Energy Studies** and **Indian Oil Corporation, Research & Development Center**, and has not been submitted elsewhere for a degree.

Prabha Kiran Dasila,

PhD Research Scholar

Department of Chemical Engineering

College of Engineering Studies

University of Petroleum and Energy Studies,

Dehradun-248007

&

Indian Oil Corporation,

R & D Center

Faridabad - 121007

THESIS COMPLETION CERTIFICATE

This is to certify that the thesis on “**KINETIC MODELING AND SIMULATION OF FLUID CATALYTIC CRACKING UNITS**” by Ms. Prabha Kiran Dasila in partial completion of the requirement for the award of the degree of Doctor of Philosophy in engineering is an original work carried out by her under our joint supervision and guidance along with that from Dr. Indranil Roy Choudhury of Indian Oil Corporation, R & D Center, Faridabad.

It is certified that the work has not been submitted anywhere else for the award of any other diploma or degree of this or any other university.

Internal Guides

Dr. D.N. Saraf

Advisor

University of Petroleum and Energy Studies

Dehradun -248007, India

Dr. S.J Chopra

Chancellor

University of Petroleum and Energy Studies

Dehradun -248007, India

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External Guide

Dr. Indranil Roy Choudhury
Research Manager
Modelling and Simulation Group
Indian Oil Corporation - R&D centre
Faridabad -121007
India

CONTENTS

Acknowledgment	i
Declaration	iii
Certificates	iv
Contents	vi
Executive Summary	viii
List of Figures	xiv
List of Tables	xvii
Nomenclature	xix
1 INTRODUCTION	1
1.1 Fluid Catalytic Cracking Unit	1
1.1.1 FCC Feed	2
1.1.2 Reactor/Regenerator Section	3
1.2 FCC Modeling	5
1.3 Scope of Present Work and Thesis Organization	6
2 LITERATURE REVIEW	9
2.1 FCC Feed Characterization	11
2.2 Catalyst and Cracking Mechanism	15
2.3 Riser Reactor Kinetic Model	19
2.4 Catalyst Deactivation and Coke Formation	26
2.4.1 Modeling of the Catalyst Deactivation	28
2.4.2 Coke Formation	31
2.4.3 Coke Formation Mechanisms	32
2.5 Hydrodynamics of Riser Reactor	33
2.6 Regenerator Modeling	35
3 FIVE LUMP KINETIC MODEL	40
3.1 Riser Model	40
3.1.1 Riser Model Equations	43
3.1.2 Catalyst Deactivation	44
3.2 Regenerator Model	44
3.2.1 Dense Phase Model Equations	47
3.2.2 Dilute Phase Model Equations	49
3.3 Simulation Procedure for Continuous Reactor –Regenerator	50
3.4 Model Validation and Parametric Sensitivity Analysis for Five Lump Kinetic Model	51
3.4.1 Effect of Feed Preheat Temperature on FCC Performance	52
3.4.2 Effect of Feed Flow Rate on FCC Performance	55
3.4.3 Effect of Air Flow Rate (Fair) on FCC Performance	57
3.5 Concluding Remarks	60
4 EXPERIMENTAL WORK	62
4.1 FCC Feed Properties	63
4.1.1 Specific Gravity	63
4.1.2 Simulated Distillation Temperature	65
4.1.3 Carbon Residue (CR)	65
4.1.4 Elemental Analysis (Total Sulfur and Total Nitrogen)	66
4.2 FCC Feed Characterization by Hydrocarbon Type	67
4.3 Plant Data from Refinery	70
5 ESTIMATION OF FEED COMPOSITION THROUGH ANN MODELING	72
5.1 Artificial Neural Network (ANN) Model Development	73
5.2 Model Performance Evaluation Criteria	77

5.3	Model Results	77
6	DEVELOPMENT OF A NEW TEN LUMP KINETIC MODEL	84
6.1	Ten Lump Kinetic Scheme	84
6.1.1	Ten Lump Model Equations	86
6.2	Determination of Reaction Parameters	88
6.2.1	Refinery Test Run Data	89
6.2.2	Data Generation from ASPEN FCC Simulator for Model Development	89
6.2.3	Kinetic Parameters Estimation	89
6.2.4	Solution of Model Equations	91
6.2.5	Validation of 10-Lump Kinetic Model	93
6.3	Sensitivity of Rate Constants in Ten Lump Kinetic Model	95
7	VALIDATION OF FCC SIMULATOR USING 10- LUMP KINETIC MODEL AND ITS COMPARISON WITH THAT USING A 5-LUMP MODEL	99
7.1	Validation with Plant Data	99
7.2	Parametric Sensitivity Study for Ten Lump Model with respect to Operating Conditions	103
7.2.1	Effect of Feed Flow Rate on FCC Performance	104
7.2.2	Effect of Feed Preheat Temperature on FCC Performance	106
7.3	Kinetic Parameters re-estimation for Five Lump Model	108
7.3.1	Comparison of Ten Lump FCC Kinetic Model with Five Lump Kinetic Model and Plant Data	110
8	CONCLUSIONS AND RECOMMENDATIONS	113
8.1	Conclusions	113
8.2	Recommendations for Future work	115
	REFERENCES	117
	APPENDICES	130

EXECUTIVE SUMMARY

Fluid Catalytic Cracking (FCC) is an important secondary process, converting low- priced heavy feedstocks like heavy oil from either the refinery crude unit or vacuum unit and heavy fractions from other conversion units (cooker gas oil and hydrocracker fractionator bottoms etc) into lighter, more valuable hydrocarbons such as liquefied petroleum gas (LPG) and gasoline and thus increases the profitability in the entire refinery. Coke is formed as a byproduct during the process along with dry gas, both of which are undesirable. The conversion and yield pattern strongly depend on the feedstock quality, operating conditions of the riser reactor-regenerator sections and the type of catalyst. The FCC process is very complex due to complicated hydrodynamics, heat transfer and mass transfer effects and complex cracking kinetics. These complex interactions coupled with economic importance of the unit have prompted many researchers to put their efforts on the modelling of FCC processes. Transport phenomena based mathematical models are the most popular because of their analytical description of the process in detail. Modeling is an iterative process and, therefore, leads to deeper understanding of the physics involved in the FCC process. Parametric sensitivity study helps in designing better control of the process unit. Process optimization, which can be subsequently carried out, can lead to improved productivity by maximizing throughput and choosing optimal operating conditions. Optimizing online can help maximize long-term profits. Additionally, running a model simultaneously in parallel with the plant operation can help in monitoring the plant and its health.

FCC feed being a mixture of hundreds of hydrocarbons, it is not possible to account for each component undergoing reactions individually. It is therefore, necessary to represent the reaction kinetic process in terms of a small number of

kinetic lumps which take part in the cracking reactions. In the often used 5-lump model, the feed is represented by a single lump of average carbon number and molecular weight. However the limitation of such models is that the kinetics is valid only for the particular vacuum gas oil (VGO) with which the model parameters were estimated and is generally not applicable to other feeds especially if the composition is significantly different. Hence, there is a need to develop a more realistic kinetic model based on detailed feed description which can be general and equally applicable to a wide range of gas oils.

The main objectives of the present work were as follows:

- Development of an Artificial Neural Network (ANN) model, which relates the simple feed properties such as specific gravity, CCR, total sulfur, nitrogen and ASTM distillation temperatures to the detailed composition of feed in terms of paraffins, naphthenes and aromatics.
- Development of a new ten lump kinetic model for the riser reactor including estimation of kinetic parameters which when coupled with a regenerator model can simulate the behaviour of the FCC unit.
- Combining the ANN model with the ten lump kinetic model along with a solution procedure into a simulation package for the prediction of FCC product yields from simple feed properties. This model should be feed composition invariant and be applicable to a variety of heavy gas oils.
- Comparison of present development with conventional five lump model results.

The proposed 10- lump kinetic model uses 6 lumps to describe the feed gas oil, namely; heavy paraffins, heavy naphthenes, heavy aromatics, light paraffins, light naphthenes and light aromatics. However, in day to day refinery operations, it is not possible to analyze every VGO stream in terms of these lumps for use in the FCC

model. It was, therefore, considered necessary to develop an artificial neural network (ANN) - based model which relates the easily measurable properties of VGO such as specific gravity, ASTM distillation temperatures, Conradson carbon residue (CCR), total sulfur and total nitrogen to the six kinetic lumps, which characterize the feed in terms of hydrocarbon types. However, laboratory distillation of various FCC feed samples showed that lighter fractions (221- 343 °C) were always less than one percent by weight. Therefore, feed is assumed to consist, only, of heavy fractions (343+ °C) of paraffins, naphthenes and aromatics. The detailed compositions of several feed samples were measured in the laboratory by using high-resolution mass-spectrometric method in terms of heavy paraffins, heavy naphthenes and heavy aromatics only. These feed samples were also analyzed in terms of routinely measured properties such as specific gravity, ASTM distillation temperatures, CCR, total sulfur and total nitrogen by using the different ASTM test methods. 60% of all the laboratory data sets were used to train several different neural nets, 20% for testing and remaining 20% were used for the model validation. Several feed forward back propagation networks with different number of neurons in hidden layers were studied using Levenberg Marquardt (LM) training algorithm. Two different ANN models (Model -1 and Model -2) were finally chosen in the present study. Model -1 predicts three output parameters: weight percentages of paraffin, naphthene and aromatic content in the FCC feed, from a single ANN architecture having three neurons in the output layer. Model -2 predicts the paraffin, naphthene and aromatic content individually from three different ANN architectures each with a single output neuron followed by normalization. In ANN modeling there is always the big question about what should constitute the input parameters and there is no straight forward way to answer. One, therefore, tends to cautiously choose all possible inputs that are likely to influence the

output. But this comes at a cost. Besides increasing computation load, particularly during training, it calls for larger data sets. Since all experimental data are prone to measurement errors, learning rates must be kept low resulting in further slowing down the training besides increasing model uncertainty and decreasing accuracy to convergence. It is therefore, desirable to use an optimal set of input parameters where the contribution of each input is more significant than the noise it adds. Initially all the 13 measured properties were chosen as input, namely: specific gravity, ASTM distillation temperatures - IBP, 5% ,10 % ,30 % ,50% ,70 % , 90 % , 95 % and FBP, Conradson carbon residue (CCR), total sulfur and total nitrogen. Subsequently sensitivity of each variable was examined. Based on the sensitivity study and intuitive reasoning, five of the 13 variables were dropped. IBP and FBP can be seldom determined with any amount of certainty while CCR, Sulfur and nitrogen content in VGO are unlikely to influence its PNA composition. Remaining 8 – variables were used as inputs. The results were presented for both 13 as well as 8 inputs for Model 1 and model 2. Among all these different investigated models, the ANN model with 8 inputs, namely specific gravity and distillation temperatures except IBP, FBP to predict paraffin, naphthene and aromatic contents individually shows best agreement with the experimental results within permissible error limits.

A conventional 5- lump kinetic model with 9 reactions was coded in C programming language and validated with the data from literature. The model was then extended to include 10- kinetic lumps and the code again validated. This was achieved by numerically integrating the model equations over the entire length of the riser reactor. A regenerator model, available in the literature, was somewhat modified before coding and was coupled with both the 5- lump and 10- lump models.

A new ten lump kinetic scheme was adopted in the present study and a total of 25 cracking reaction paths were identified after dropping eight reactions because of their very low rates. The kinetic parameters were determined for these 25- reactions, which are generally invariant to feed gas oil composition. The high volume of experimental data required for kinetic parameters estimation was not practical to generate from any operating plant. It was, therefore, decided to use a combination of experimental data obtained from a refinery and those generated from ASPEN FCC simulator. These data were regressed using an evolutionary optimization technique, genetic algorithm, to evaluate the rate constants. An objective function was constructed from the sum of squares of errors between the measured and model calculated values which was minimized subject to the model equations as constraints. Since genetic algorithm (GA) is a global optimization technique, it was assumed that the converged set represented the true kinetic parameters. The detailed composition required as input to the 10- lump kinetic model was obtained from the validated ANN model described above requiring only routinely field laboratory measured feed properties as input.

A parametric sensitivity analysis of estimated kinetic constants was done by varying all the 25- frequency factors and 25- activation energies in steps of $\pm 10\%$, $\pm 20\%$ and $\pm 40\%$ from their mean position, one at a time, to see how sensitive are the gas- oil conversion and yields of gasoline, LPG, dry gas and coke to these parametric variations. While a gradual variation of the output is expected, unusually high sensitivity may reflect adversely on the validity of the kinetic parameters. Alternatively, such analysis also helps in designing appropriate control systems. The variation of gas oil conversion and product yields with respect to frequency factor (k_0 , i) and activation energies (E_i) were plotted only for the most sensitive reactions. All

changes were found to be gradual as expected. The 25- heat of reactions (ΔH_i) were also varied in steps of $\pm 40\%$, and $\pm 60\%$ from their mean position, one at a time, to study their sensitivity on gas- oil conversion and product yields, which showed very small change in gas- oil conversion and product yields.

The data that were regressed to obtained kinetic parameters for the 10 - lump model were reused to calculate kinetic parameters for the 5- lump model to facilitate comparison between the two models. Several sets of test run data and one set of normal operating data were obtained from an operating FCC plant in a refinery for validation of the developed simulator. The different data sets were collected with different feed compositions but with the same catalyst. While one wishes to validate the model at several locations in the riser reactor but in absence of plant measured values, it was only possible to compare the product yields and the reactor outlet temperature which are the only available measured experimental values. While the 5- lump model predicted values deviated significantly, the 10- lump model predictions were found to be in good agreement with plant data for all the cases investigated. This established the validity of the work done in this study including the ANN model, the simulator based on 10- lump kinetics and the kinetic parameters determination. This also demonstrated that the present simulator is independent of the feed heavy gas oil composition. Finally a parametric sensitivity study was undertaken in respect of operating conditions. The feed preheat temperature and feed flow rate to the riser reactor and input air rate to the regenerator were the three independent input parameters which were found to influence the FCC operation most.

LIST OF FIGURES

Figure No.	Title	Page No.
Fig.1.1	Schematic of riser reactor - regenerator	4
Fig.2.1	Scheme of different FCC units	10
Fig.2.2	Three- lump model (Weekman and Nace, 1969)	21
Fig.2.3	Four- lump model (Lee et al., 1989)	21
Fig.2.4	Five lump kinetic Scheme (Bollas et al., 2007a)	22
Fig.2.5	Six lump kinetic scheme (Takatsuka et al., 1987)	22
Fig.2.6	Seven lump scheme (Maya- Yescas et al., 2005)	23
Fig.2.7	Ten lump kinetic scheme (Jacob et al., 1976)	25
Fig.2.8	Twelve lump kinetic scheme (Cerqueira et al., 1997)	26
Fig.2.9	Coke formation	33
Fig.2.10	Schematic depiction of TDH	36
Fig.3.1	Five lump kinetic scheme	41
Fig.3.2	Effect of feed preheat temperature on gas oil conversion and product yields at fixed F_{feed} (32.14 kg/sec) and fixed regenerator temperature (937.5K)	53
Fig.3.3	Effect of feed preheat temperature on riser outlet temperature (ROT) at fixed F_{feed} (32.14 kg/sec) and fixed regenerator temperature (937.5K)	53
Fig.3.4	Effect of feed preheat temperature on regenerator temperature (Trgn) at fixed feed flow rate (32.14 kg/sec) and fix ROT (769 K)	54
Fig.3.5	Effect of feed preheat temperature on gas oil conversion and product yields at fixed feed flow rate (32.14 kg/sec) and fix ROT (769 K)	55
Fig.3.6	Effect of feed flow rate on the conversion and product yields at fixed feed preheat temperature (625K) and fixed regenerator temperature (937.5K)	55
Fig.3.7	Effect of feed flow rate on the reactor outlet temperature (ROT) at fixed feed preheat temperature (625K) and fixed regenerator temperature (937.5K)	56
Fig.3.8	Effect of feed flow rate on the conversion and product yields at	57

	fixed reactor outlet temperature (768.8K) and fixed feed preheat temperature (625K)	
Fig.3.9	Effect of feed flow rate on the regenerator temperature (T_{rgn}) at fixed feed preheat temperature (625K) and fixed reactor outlet temperature (768.8K)	57
Fig.3.10	Effect of air flow rate on the regenerator temperature (T_{rgn}) and reactor outlet temperature (ROT)	58
Fig.3.11	Effect of air flow rate on the conversion and product yields.	58
Fig.3.12	Effect of regenerated catalyst temperature (T_{rgn}) on the conversion and product yields.	59
Fig.3.13	Effect of regenerated catalyst temperature (T_{rgn}) on reactor outlet	59
Fig.5.1	A typical neuron with linear synaptic operation	75
Fig.5.2	Neural network architecture with two hidden layers and an output layer	76
Fig.5.3	Parity plot between ANN predicted compositions and experimental values for models using 13 input variables.	82
Fig.5.4	Parity plot between ANN predicted compositions and experimental values for models using 8 input variables	83
Fig.6.1	The ten lump kinetic scheme	85
Fig.6.2	Sensitivity as a function of frequency factors (a) conversion (b) gasoline yield (c) LPG yield (d) coke yield	96
Fig.6.3	Sensitivity as a function of activation energies (a) conversion (b) gasoline yield (c) LPG yield (d) coke yield	97
Fig.6.4	Sensitivity as a function of heat of reactions (a) conversion (b) gasoline yield (c) LPG yield (d) coke yield	98
Fig.7.1	Effect of feed flow rate on the conversion and product yields at fixed feed preheat temperature (621K) and fixed regenerator temperature (935K)	104
Fig.7.2	Effect of feed flow rate on the reactor outlet temperature (ROT) at fixed feed preheat temperature (621K) and fixed regenerator temperature (935K)	104
Fig.7.3	Effect of feed flow rate on the conversion and product yields at fixed reactor outlet temperature (767.4 K) and fixed feed preheat	105

	temperature (621 K)	
Fig.7.4	Effect of feed flow rate on the regenerator temperature ($T_{r_{gn}}$) at fixed feed preheat temperature (621 K) and fixed reactor outlet temperature (767.4K)	106
Fig.7.5	Effect of feed preheat temperature on gas oil conversion and product yields at fixed F_{feed} (50.16 kg/sec) and fixed regenerator temperature (935K)	106
Fig.7.6	Effect of feed preheat temperature on riser outlet temperature (ROT) at fixed F_{feed} (50.16 kg/sec) and fixed regenerator temperature (935K)	107
Fig.7.7	Effect of feed preheat temperature on gas oil conversion and product yields at fixed F_{feed} (50.16 kg/sec) and fix ROT (767.4 K)	108
Fig.7.8	Effect of feed preheat temperature on regenerator temperature ($T_{r_{gn}}$) at fixed feed flow rate (50.16 kg/sec) and fix ROT (767.4 K)	108

LIST OF TABLES

Table No.	Title	Page No.
Table 3.1	Input Data Used in the Simulation	41
Table 3.2	Thermodynamic and Other Parameters Used in the Simulation of FCC Unit	42
Table 3.3	Design Data Used for the Simulation of FCC Unit	42
Table 3.4	Kinetic and Thermodynamic Parameters Used for Reactor Modeling	43
Table 3.5	Comparison of Plant Measured and Models Prediction Data	52
Table 3.6	Comparison of FCC Performance at Three Different Feed Preheat Temperatures with Increased Cat/Oil Ratio and Air Flow Rate	60
Table 4.1	Properties of Different Heavy Gas Oil Samples	63
Table 4.2	Mass Spectrometry Analysis for Different Samples	70
Table 4.3	Feed Properties for Different Blends	71
Table 5.1	Range of Input Datasets Used for ANN Training	78
Table 5.2	Range of Input Datasets Used for Model Validation	79
Table 5.3	Summary of ANN Model Architectures for 13 Input Variables	80
Table 5.4	Summary of ANN Model Architectures for 8 Input Variables	80
Table 5.5	A Comparison of Model Predictions with Experimental Observations for Two Different ANN Models with 13 Input Variables (Validation Set): (a) Model 1 (b) Model 2	81
Table 5.6	A Comparison of Model Predictions with Experimental Observations for Two Different ANN Models with 8 Input Variables (Validation Set)	82
Table 6.1	Feed Composition and Properties Measured in the Lab	91
Table 6.2	Plant Operating / Design Data Used in Simulation	92
Table 6.3	Thermodynamic and Other Parameters Used in Simulation	92
Table 6.4	Comparison of Tuned ASPEN Model Outputs with Plant Test Run Data	93
Table 6.5	Calculated Frequency Factors, Apparent Activation Energies and Heat of Reactions	94
Table 6.6	Comparison of Model Calculated Values with Plant Data	95
Table 7.1	Data Used in Simulation of Ten Lump Model	100

Table 7.2	Comparison of Model Calculated Values with Plant Data (Case I)	100
Table 7.3	Comparison of Model Calculated Values with Plant Data (Case II)	101
Table 7.4	Comparison of Model Calculated Values with Plant Data (Case III)	102
Table 7.5	Comparison of Model Calculated Values with Plant Data (Case IV)	103
Table 7.6	Calculated Kinetic Parameters for 5-Lump Model	110
Table 7.7	Comparison of Ten Lump and Five Lump Model Calculated Values with Plant Data (Case I)	110
Table 7.8	Comparison of Ten Lump and Five Lump Model Calculated Values with Plant Data (Case II)	111
Table 7.9	Comparison of Ten Lump and Five Lump Model Calculated Values with Plant Data (Case III)	111
Table7.10	Five Lump Model Calculated Values with Literature Kinetic Parameters (Case I)	112
Table7.11	Five Lump Model Calculated Values with Literature Kinetic Parameters (Case II)	112

NOMENCLATURE

A_{rgn}	Regenerator cross- section area, m^2
A_{ris}	Riser cross- sectional area, m^2
C_h	Weight fraction of hydrogen in coke, $(kg H_2)/(kg \text{ coke})$
C_c	Coke on catalyst, $kg \text{ coke}/kg \text{ catalyst}$
C_i	Concentration of i th component, $kmol/m^3$
C_{pc}	Catalyst heat capacity, kJ/kgK
C_{pco}	Mean heat capacity of CO, kJ/kgK
C_{pco2}	Mean heat capacity of CO_2 , kJ/kgK
C_{pfl}	Liquid feed heat capacity, kJ/kgK
C_{pfv}	Vapor feed heat capacity, kJ/kgK
C_{ph2o}	Mean heat capacity of water, kJ/kgK
C_{pN2}	Mean heat capacity of N_2 , kJ/kgK
C_{pO2}	Mean heat capacity of O_2 , kJ/kgK
C_{rgc}	Coke on regenerator catalyst, $(kg \text{ coke})/kg \text{ cat}$
C_{sc}	Coke on spent catalyst, $(kg \text{ coke})/kg \text{ cat}$
E_{β}	Activation energy for CO/ CO_2 at the catalyst surface
E_j	Activation energy of i th cracking reaction in the riser
E_{13c}	Activation energy for homogeneous CO combustion
E_{13h}	Activation energy for heterogeneous CO combustion
f_c	Molar flow rate of carbon in the regenerator, $kmol/sec$
f_{co}	CO molar flow rate in the regenerator, $kmol/sec$
f_{co2}	CO_2 molar flow rate in the regenerator, $kmol/sec$
f_{h2o}	H_2O molar flow rate in the regenerator, $kmol/sec$
f_{N2}	N_2 molar flow rate in the regenerator, $kmol/sec$
f_{O2}	O_2 molar flow rate in the regenerator, $kmol/sec$
f_{tot}	Total gas molar flow rate in the regenerator, $kmol/sec$
F_{air}	Air flow rate to the regenerator, $kmol/sec$
F_{ent}	Entrained catalyst flow rate kg/sec
F_j	Molar flow rate of j th lump, $kmol/sec$
F_{rgc}	Catalyst Circulation Rate (CCR), kg/sec
F_{sc}	Spent catalyst flow rate, kg/sec

F_{feed}	Oil feed flow rate, kg/sec
h	Dimensionless riser height
H_{ris}	Riser height, m
ΔH_{evp}	Heat of vaporization of oil feed, kj/kg
H_{co}	Heat of Formation of oil feed, kj/kmol
H_{co2}	Heat of formation of CO_2 , kj/kmol
H_{h2o}	Heat of formation of H_2O , kj/kmol
ΔH_i	Heat of cracking of ith lump, kj/kmol
i	total no. of reactions in the reactor
j	Total no. of kinetic lumps
$k_{0,i}$	Frequency factor for ith reaction in the riser
k_{co}	Frequency factor for coke combustion, 1/ (atm) (s)
k_{13c}	Frequency factor in heterogeneous CO combustion expression, kmol CO/ (m^3) (atm^2) (s)
k_{13h}	Frequency factor in homogeneous CO combustion expression, kmol CO/ (m^3) (atm^2) (s)
MW_j	Molecular weight of jth lump, kg/kmol
MW_c	Molecular weight of coke, kg/kmol
MW_g	Average molecular weight of gas oil feed, kg/kmol
MW_H	Molecular weight of hydrogen
P_{ris}	Riser pressure, atm
P_{rgn}	Regenerator pressure, atm
P_{O2}	Average mean oxygen partial pressure, atm
Q_{air}	Heat flow rate with air, kj/sec
Q_C	Heat released by the carbon combustion, kj/sec
Q_{ent}	Heat input to the dense bed from entrained catalyst returning from cyclone, kj/sec
T_{feed}	Gas oil feed temperature, K
T_{rgn}	Regenerator dense bed temperature/Regenerated catalyst temperature, K
T_{sc}	Temperature of spent catalyst, K
ΔT_{st}	Stripper temperature drop (~ 10 °C)
W	Catalyst inventory in the regenerator, kg
X_{pt}	Relative catalytic CO combustion rate
X_j	Mole fraction of jth component

Z	Axial height from the entrance of the riser or regenerator, m
Z_{bed}	Regenerator dilute bed height
Z_{dil}	Regenerator dilute phase height, m
Z_{rgn}	Regenerator height, m
$Q_{\text{loss, rgn}}$	Heat losses from the regenerator, kj/sec
$Q_{\text{loss, ris}}$	Heat losses from the riser base, kj/sec
r_i	Rate of the i th reaction (kmol/kg.cat.s)
R	Universal gas constant
ROT	Riser outlet temperature (K)
T	Riser temperature at any axial height, K
T_{air}	Temperature of the air to the regenerator
T_{base}	Base temperature for heat balance calculations, K (866.6 K)
Q_{rgc}	Heat flow with regenerated catalyst, kj/sec
Q_{sc}	Heat flow rate with spent catalyst, kj/sec
Q_{sg}	Heat flow rate with gases from the regenerator dense bed, kj/sec
Q_{H}	Heat released by the hydrogen combustion, kj/sec

Greek Letters

α_{ij}	Stoichiometric coefficient of j th species in i th reaction
β_c	CO/CO ₂ ratio at the surface in the regenerator
β_{c0}	Frequency factor in β_c expression
ε	Riser or regenerator void fraction
ρ_c	Catalyst density, kg/m ³
ρ_{den}	Catalyst density in the regenerator dense bed, kg/m ³
ρ_{den}	Catalyst density in the dilute phase of the regenerator, kg/m ³
ρ_g	Molar gas density in the regenerator, kmol/m ³
ρ_v	Oil vapor density, kg/m ³
ϕ	Catalyst activity
Θ	Catalyst residence time, sec