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Original Research Article

A Four-Lump Kinetic Model for Atmospheric Residue Conversion in the Fluid Catalytic Cracking Unit: Effect of the Inlet Gas Oil Temperature and Catalyst-Oil Weight Ratio on the Catalytic Reaction Behavior

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Abstract: The effects of the inlet gas oil temperature and catalyst-to-oil weight ratio on the catalytic cracking behaviors and the distribution of the product and the yield of the riser outlet main products as unconverted gas oil, gasoline, and coke were investigated on a real industrial FCC Riser reactor with a model LRC-99 catalyst and atmospheric residue derived from Niger's crude oil of Agadem bloc was used as the feedstock. A four-lumped kinetic mathematical model is developed to predict the feedstock conversion and the product distribution by using the deactivation function coke-on- catalyst approach. The model was validated with a good agreement against real industrial fluidized bed riser reactor plant data from Niger and industrial riser data from literature sources. The results show that the inlet gas oil temperature and catalyst-to-oil weight ratio obviously affect the cracking reactions behavior and govern the reaction rates.

Keywords: Fluid Catalytic Cracking, Atmospheric residue, Riser-reactor, Hydrodynamic, Lump-kinetic, Modeling, Prediction, Temperature, CTO.

1. INTRODUCTION

The riser reactor is the most important piece of the Residue Fluidized Bed Catalyst Cracking (FCC) unit where almost the endothermic cracking reactions and coke deposition on the catalyst occur [1]. It ensures the conversion of heavy or long-chain hydrocarbons to small ones through the catalyst(zeolite) acid sites [2]. The inlet gas oil temperature and catalyst-to-oil weight ratio are some of the key parameters to control, optimize and predict the conversion and the product distribution along the riser reactor [3-6]. Many kinetic riser models were developed to apply to the large game of feedstocks including the heavy charge as an atmospheric residue.

In this work, the model has been validated by comparison with real industrial FCC riser data in Niger [7] and industrial FCC plant data (Ali & Rohani,1997) available from several studies published in the open literature [2, 8, 9]. A parametric study of the influence of the inlet gas oil temperature and catalyst-to-oil weight ratio was conducted on Zinder Oil Refining company's FCC Plant Data.

2. Feedstock and LRC-99 Catalyst

The atmospheric residue derived from Niger's crude oil of the Agadem bloc was used as feedstock, and its main properties are listed in Table 1. Catalyst LRC-99, a commercial catalytic cracking zeolite used by Zinder Oil Refining Company Limited as the main catalyst for the FCC process, and its properties are detailed in Table 2.

3. MATERIALS AND METHODS

The coke on the catalyst regenerated is measured according to Q/SYLS Standard using combustion and IR Test Methods [10, 11]:

Sample Analysis: Take 0.200g sample from the crucible, weigh it accurately to 0.001g, add 1.5g flux, and then start the instrument, put the sample into the burning furnace for analysis. The instrument will automatically report the result after the analysis is over.

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Computation Formula: The content of carbon in the catalyst C_{coke} (%) should be calculated according to Formula (1):

$$C_{Coke}(\%) = \frac{F+G}{W} * 100$$

Where:

C_{coke} - the mass percentage content of carbon, % (mass fraction);

F - Correction index drawn from the external standard;

G - The sample concentration determined by the IR detector, % (mass fraction);

W - The sample mass, g.

The results are presented in table 3, and for our purpose, 0.05 is used as the average value of carbon content on the catalyst. C_{coke} =0.05

Table 1: Feedstock (case II) properties [7]

Item	value
Density (20°C)	0.9215 g/cm^3
API	22.054
Aniline point	52.9 °C
Residual carbon	5.5%
Ni	22.9 ppm
V	0.3 ppm
Fe	3.2 ppm
Sulfur content	0.24 m%
Nitrogen content	0.17 m%
Molecular weight	462
Distillation TBP	°C
50%	343.0
90%	378.0
95%	385.0

Table 2: Properties of Catalyst LRC-99

Item	Value
Micro-activity index	58-59 v%
Carbon content on Catalyst<=0.3	0.04-0.05%
Compositions	Al_2O_3; FeO_3; Na_2O
Particule sizes distribution	
0.01-18.5 µm	2.05 v%
18.5-39.5 µm	21.14 v%
39.5-83.5 µm	48.24 v%
83.5-111 µm	14.94 v%
111>= µm	13.63 v%

Table 3: Coke on regenerated catalyst

Carbon content <=0.3	
Q/SYLS 1515	**C_{coke} (%)**
Sample 1	0.04
Sample 2	0.06
Sample 3	0.05
Sample 4	0.05

4. Mathematical Modeling

4.1. Reactor Model

In order to develop a mathematical model for this system the following assumptions are introduced:

i. One-dimensional transported plug flow reactor prevails in the riser without radial and axial dispersion;
ii. The riser wall is adiabatic;
iii. Dispersion and adsorption inside the catalyst particles are negligible;
iv. In each section of the riser, the catalyst and gas have the same temperature;
v. The riser dynamic is fast enough to justify a quasi-steady-state model;
vi. Instantaneous vaporization occurred at the entrance of the riser.

4.2. Hydrodynamic and Model of the Riser Reactor

4.3. FCC Reaction Kinetics

The four-lumped reaction schemes as follows [12]:

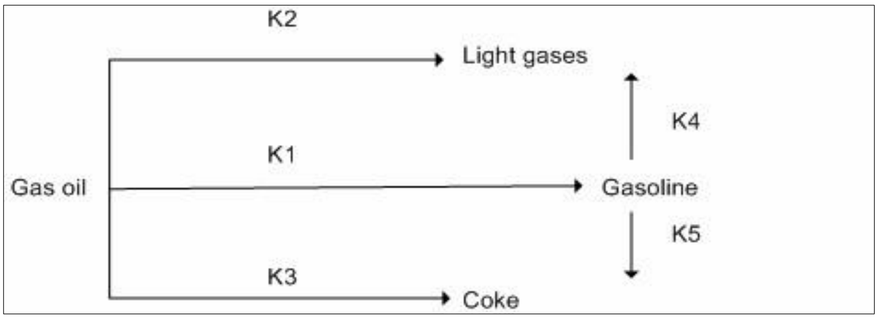

Figure 1: Schematic of four-lumped reactions

The constants of the reaction velocity k_i depend on the temperature in the riser and the reaction activation energy E_a obtained based on Arrhenius' law [13]. In order to determine these constants, the following relations are used [14]:

$$k_i = k^0{}_i * e^{(\frac{E_a}{R})(\frac{1}{T_R}-\frac{1}{T_g})}$$

4.4. The Material Balance

In order to calculate the concentration profile for each lump with coke on catalyst surface approach, a differential material balance can be applied along the riser-reactor, the following next equation thus being obtained [15-17] :

For gas oil (feedstock)

$$\frac{dy_1}{dz} = -\frac{\phi_s A_R L_R \, \rho_g \varepsilon_g}{F_g} * [k_1 + k_2 + k_3]{y_1}^2 * CTO$$

For gasoline

$$\frac{dy_2}{dz} = -\frac{\phi_s A_R L_R \, \rho_g \varepsilon_g}{F_g} * [(k_5 + k_4)y_2 - k_1 {y_1}^2] * CTO$$

For light gases

$$\frac{dy_3}{dz} = \frac{\phi_s A_R L_R \, \rho_g \varepsilon_g}{F_g} * [k_4 y_2 + k_2 {y_1}^2] * CTO$$

For coke

$$\frac{dy_4}{dz} = \frac{\phi_s A_R L_R \, \rho_g \varepsilon_g}{F_g} * [k_5 y_2 + k_3 {y_1}^2] * CTO$$

The hydrocarbon gases void fraction in the riser is assumed constant along the riser highest and based on the literature the 0.2 value respectively are chosen [5, 16].

The catalyst deactivation function depending the weight percent of coke on the catalyst surface are much more advisable because coke is the main cause of catalyst deactivation [18-21]. As determined early the average of coke on catalyst surface, in this work is used the catalyst deactivation function proposed by Pitault *et al.*, 1995 [9, 22].

$$\phi_s = \frac{X+1}{X+\exp(Y*C_{cok})}$$

The values for deactivation constants X and Y reported by the authors are 4.29 and 10.4, respectively; the same are used in this work. C_{coke} is the concentration of coke on catalyst surface (wt%). It takes into account the chemical deactivation as site recovery and diffusional (pore clogging) [23, 24].

6. Details of Calculations

6.1 Necessary Data

In this part is presented the industrial plant data of Zinder Oil Refining company's FCC unit [7] is using to steady the effects of different key's parameters of cracking reaction, while the industrial plant data (Ali and Rohani,1997) from literature is using for a model validation, show in Table 4. The specifications kinetic parameters for cracking reactions from the literature [9] used in this study can be found in Table 4. The atmospheric residue thermodynamic properties as feedstock of this work are also presented in Table 6.

Table 4: Operation parameters and riser reactors properties of the industrial plant data of Zinder Oil Refining company's FCC unit [7] and Ali and Rohani industrial FCC plant data[9]

	Ali & Rohani, 1997	**Data of FCC riser in Niger**
Catalyst feed rate kg/s	144	
Gas oil feed rate(kg/s)/CTO	20/7.2	26.1036/7
Inlet temperature of gas oil feed (°K)	496	490
Inlet temperature of catalyst feed (°K)	960	
Riser diameter (m)	0.8	1.020
Riser height (m)	33	38
Catalyst diameter (μm)	70	75
Inlet riser pressure (atm)	2.9	
Catalyst density (Kg/m^3)	1800	

Table 5: Kinetic parameters reported from the literature [9] for FCC cracking reactions

Reaction	K_0 (s^{-1})	E_a (Kg/Kmol)
Gas oil to gasoline	1457.5	57359
Gas oil to light gases	127.59	52754
Gas oil to coke	1.98	31830
Gasoline to light gases	256.81	65733
Gasoline to coke	0.022	66570

Table 6: Thermodynamic properties of the atmospheric residue

Gas oil vaporization temperature	641.8°K
Heat capacity	2.1kj/kg

6.2 Solution Algorithm

A variable step Runge Kutta Method (Matlab tool ODE 45) is used to numerically solve the system of four ordinary differential equations [25, 26]. Hence a MATLAB code has been developed for this work.

With the bondery conditons :

$$At\ Z = 0 \begin{cases} y1 = 1 \\ y2 = y3 = y4 = 0 \\ \text{And TR} = 623.15°K \end{cases}$$

The relative error of calculated from different plant data were estimated as follows:

$$\%\ \text{Relative error} = \frac{\text{Plant data} - \text{model data}}{\text{Plant data}} * 100$$

7. RESULTS AND DISCUSSION

The estimated kinetic parameters effects on the cracking reaction behavior and the distribution of the atmospheric residue derived from Niger's crude oil of Agadem bloc as feedstock (case II) are presented in this section. Moreover, the results of the parameter estimation for Ali and Rohani industrial FCC plant data (case I) and the four lumped kinetic model from literature [9] to demonstrate the accuracy and the validation of the model used in this present work are also presented in Table 8.

The estimated results of the atmospheric residue derived from Niger's crude oil in Table 8, gives very close estimates as compared against the plant data values while, the relative errors between Ali and Rohani industrial plant data and the estimate values by using the same kinetic model appeared to large compared with the preview once. Keeping the feedstock rate and the coke to oil weight ration constant as the plants data. It's noted that the relative error of the gasoline and the coke are 10.87%, 0.09% respectively for case II, while the relative errors of the gasoline and the coke are 17.78% and 19.47% respectively for case I. It's also important to note that different between these industrial plant data and the present developed model values appeared to be very large, it may not be very significant. The reason being that some operation conditions and kinetic parameters no needed for this work are neglected and for this model is used the coke on catalyst approach with specific catalyst deactivation data different. However, the good agreement between the case II plant data and the model prediction data is because the catalyst deactivation take in consideration both of the necessary data about the feedstock and riser operation conditions during its determination as feed metals content, catalyst properties and the riser operating conditions. The inlet gas oil temperature relative errors are very small for both of the two cases with the average 2.87% to 3.72% as presented in Table 8.

Figure 2 shows the optimal profile of the estimated parameters of industrial plant data for case II, the plan data's overall unconverted gas oil is about 30.2 % whereas the gasoline, and light gas yields are about 45.4%, 14.54% respectively, while the model predicted 26.50% for unconverted gas oil, 50.33% for gasoline, and 13.75% for light gas at the riser exit.

It is noted that at the riser height of 19m (50% of the total riser heigh) more than 41.01% of gasoline already formed, this is because the most important conversion of the feedstock take place at the bottom of the riser (vaporization section) but not in all type of riser [3].

According to the different between the plan data and model predict values (Table 8) an acceptable agreement is observed, and this show, that kinetic parameters modification (Table 7) calculate by Arrhenius' law has been achieved successfully.

Table 7: Modified Kinetic Parameters

Reaction	K_0
Gas oil to gasoline	0.0256
Gas oil to light gases	0.0054
Gas oil to coke	0.0045
Gasoline to light gases	9.1038e-04
Gasoline to coke	6.6471e-08

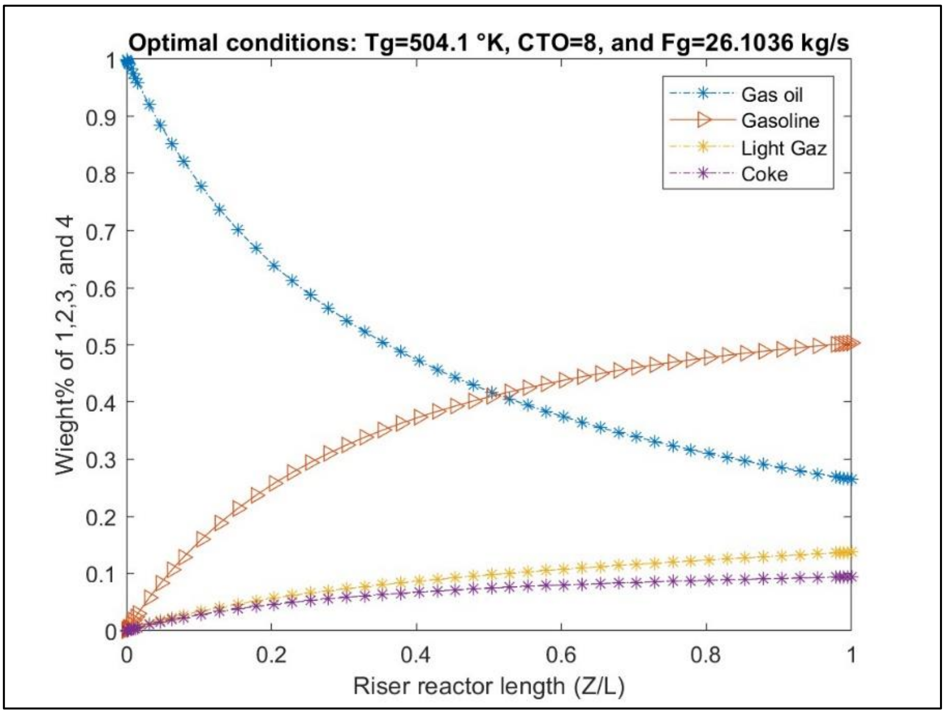

Figure 2: Profile of feedstock (case II) cracking in the riser-reactor.

Table 8: Comparison of Coke and Gasoline Yields and Riser inlet Temperature Predicted by the Model with Plant Data of Zinder Oil Refining Company Limited in Niger [7] and Plant Data Supplied by Ali and Rohani [2]

	Case I Ali & Rohani, 1997			**Case II FCC riser In Niger**		
	Plant	Model	% Relative error	Plant	model	% Relative error
Gas oil feed rate(Kg/s)	25.7			26.1036		
CTO (Kg/Kg)	6.33			8		
Gasoline yield (wt%)	46.9	55.24	17.78	45.4	50.33	10.87
Coke (wt%)	5.34	6.38	19.47	9.4	9.4089	0.09
Feed temperature (°K)	496	514.5	3.72	490	504.1	2.87

7.1 Effect of the inlet gas oil temperature and the catalyst to oil weigh ratio on catalyst cracking behavior and product distribution in the riser

Keeping the feed flow rate and the weight of catalyst to oil weigh ratio constant as the same in case II, and varying the inlet gas oil temperature 495°k to 520°K, this research investigated the effect of the temperature on product distribution and the yield of gas oil, gasoline, light gas and coke as showed in figure 3 (a, b).

As the temperature increases in the range 495°K to 504.1°K , both the conversion and yield of gasoline, dry gas, and coke phases increase [6, 27], while the gas oil yield decrease and on further increasing the inlet gas oil temperature to 515°K a decline in gasoline yield can be observed at the riser exit because of the secondary reaction [6, 28]. And at 520°k, we can noted that the gas oil is almost converted 6.3% while the gasoline yield considerable decrease to 46.95%.

The effect of the catalyst to oil weight ratio was investigated in the range 5 to 9 by keeping the temperature and feedstock flow rate constant as the same in case II.

As the catalyst to oil weight ratio increases, the gas oil yield progressively decreases and both the gasoline, light gas, and coke yields increase [27]. Due to more catalyst flow rate , the number of active sites increases, cause more cracking the long chain hydrocarbons to small ones, and increases conversion of feedstock to light products and coke [29, 30]. Example: when we maintained the intel gas oil temperature and the feedstock flow rate constant, the conversion of the gas oil increase 13.8% to 9.18% for catalyst to oil weight ration 5 and 9 respectively, while the yield of the gasoline passes 74.859 % to 77.16% for catalyst to oil ration 5 and 9 respectively as show in Figure 4(a, b). The important things in this case is that the more the catalyst to oil weight ration decline the more the coke yield decrease and gasoline yield increase.

In addition to exploring the inlet gas oil temperature and the catalyst to oil weigh ratio (CTO) on the cracking reaction behavior, the Figure 5 show in 3D the product distribution and critical riser exit fractions as gasoline(a), gas oil(b), light gas(c) and coke(d) [29].

With the growth the CTO and temperature to high values 9 and 520°K respectively, the gasoline, unconverted gas oil and coke rates decrease as show in figure 5 (a, b, d), while the light gas increase as show in figure 5 (c).

With the low values CTO and temperature 6 and 495°K respectively, there is more unconverted gas oil rate than the most important fraction as gasoline. Thant mean in this condition there are an important no cracking molecule and this is because the catalysts over the feedstock is not enough to provide the necessary acid sites for good molecules cracking.

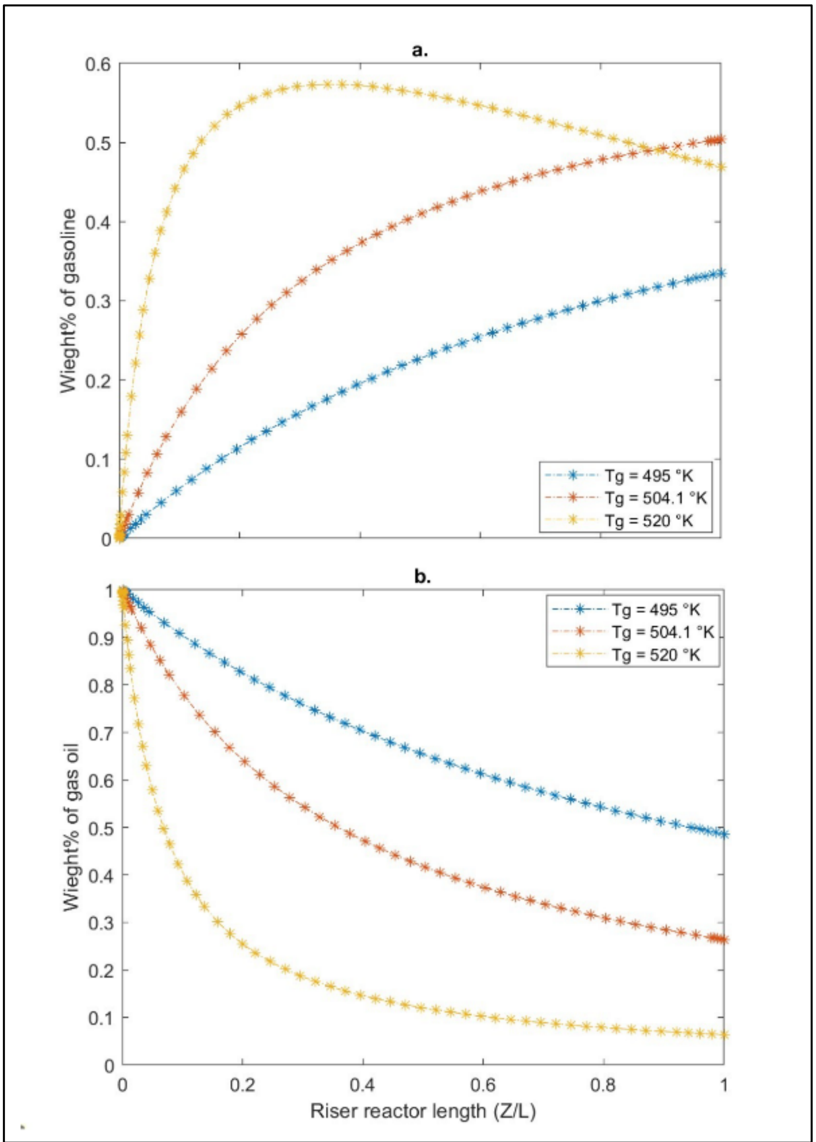

Figure 3: Effect of changing inlet gas oil temperature on the weight% gas oil and the gasoline yield in the riser. Operating conditions: CTO ratio = 8, and F_g= 26.1036 Kg/s.

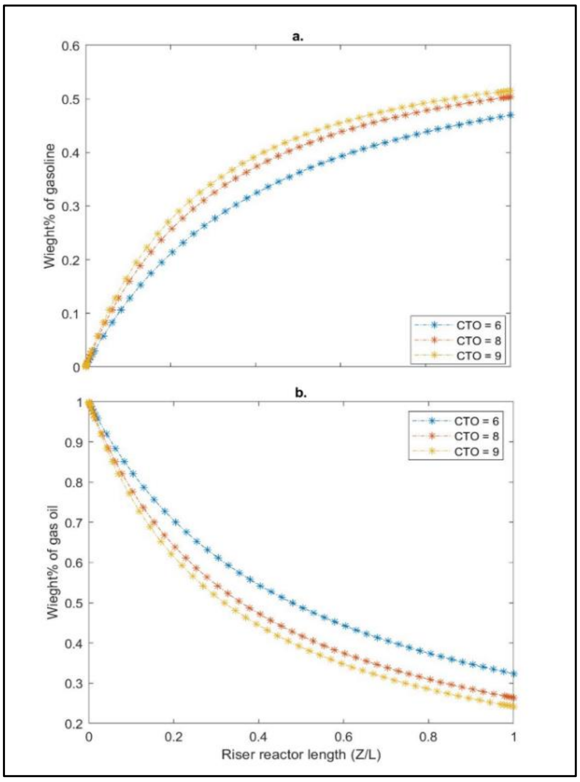

Figure 4: Effect of changing catalyst-to-oil ratio on the weight% gas oil and the gasoline yield in the riser. Operating conditions: inlet gas oil temperature= 504.24°k and F_g = 26.1036 Kg/s.

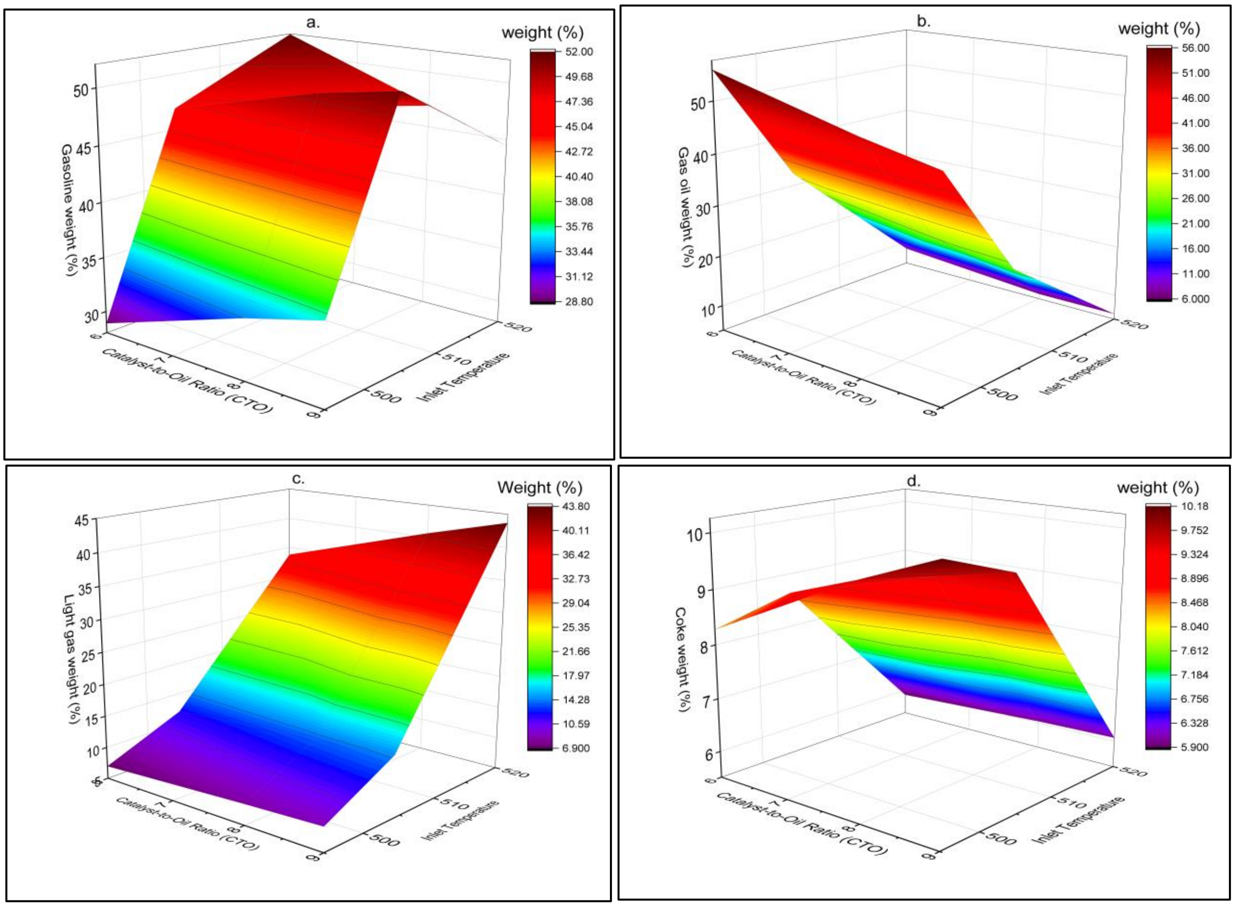

Figure 5: Effect of the two key parameters (CTO and T_g) on the cracking reaction behavior and the riser exit critical fractions yields as gasoline (a), gas oil (b), light gas (c), and coke (d)

CONCLUSION

The feedstock inlet temperature and the catalyst-to-oil ratio are the most important parameters in the conversion and distribution of cracking feedstock to light products. This present work shows that both of the two key parameters (T_g and CTO) have obvious effects on the cracking reaction behavior of the atmospheric residue derived from Niger's crude oil of Agadem bloc over a commercial LRC-99 catalyst.

The optimal feed temperature is 504.1°K, and the optimal catalyst to oil weight ratio is about 8 for a feed flow equal 26.1036 Kg/s. Under the optimal operation conditions, the four-lump kinetic model used with Pitault *et al*.,(1995) catalyst deactivation function can well predict the product distribution in the industrial riser reactor.

Nomenclature

A	Cross-sectional area, m^2
CTO	catalyst to oil weight ratio, Kg/Kg
F_g	hydrocarbon gases mass flow rate in the riser, Kg/s
K_i	rate constants between species i and j
L_R	riser length, m
R	Gas constant
T_g	inlet gas oil temperature °K
TR	riser temperature °K
Y_i	weight percent of hydrocarbons in the riser (i = 1,2,3, and 4)
ε_g	hydrocarbon gases void fraction in the riser
ρ_g	density of gas phase in the riser, kg m^{-3}
Φ	catalyst deactivation
FCC	Fluid Catalyst Cracking

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