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Vacuum Distillate Catalytic Cracking Technology Modelling: Feedstock and Products Composition and Properties Investigation

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Abstract

This paper investigates and explains how the feedstock composition affects the catalytic cracking products. For this purpose, at the first stage, the group composition of the catalytic cracking vacuum distillate (297-540ºC) and the products of the process (heavy fractions fr. 195-340ºC, fr. 340$^+$ºC), the feedstock structural-group composition, as well as the coke structure of coked and regenerated catalysts by TG-DSC. Using a mathematical model, it was found that with an increase in the ratio of saturated hydrocarbons to the sum of aromatics and resins from 1.44 to 3.17 units, the yield of wet gas increases by 1.64, the yield of the gasoline fraction increases by 2.62% wt., the yields of fractions 195–340ºC and 340$^+$ºC decreases by 1.16 and 1.93% wt respectively. It was shown that feedstock with a high content of saturated hydrocarbons (feedstock No. 4) provides the highest yield of gasoline fraction (45.65 wt%) and wet gas (36.98%).

Keywords: Vacuum distillate; Structural-group analyses; Catalytic cracking; Group composition; Gasoline; Coke; Catalyst.

1. Introduction

The mathematical modeling method is widely used in oil refining for predicting and optimizing the industrial unit's operation [1–4], testing catalysts [5-6], planning the products yield and quality [7–9], as well as for conducting complex experiments on the process conditions influence upon various feedstock processing.

In terms of the catalytic cracking technology, the yield and composition of the products vary significantly depending on the processed feedstock type and the catalysts used. In contrast to the light feedstock processing, the middle distillates [10–14] and heavy feedstock (vacuum distillate, residual feedstock) refining modeling [15–22] is significantly complicated due to the identifying complexity of high molecular weight hydrocarbons involved in the reactions, as well as the feedstock group and structural-group composition. These data are necessary to create a reliable mathematical model considering changes in the feedstock hydrocarbon composition, at the stages of thermodynamic analysis, hydrocarbon transformation scheme formation and solving the inverse kinetic problem, etc. The main stages of the catalytic cracking process mathematical model development are listed below:

1) study of the composition and properties of the catalytic cracking process feedstock and products, monitoring the operation of an industrial unit;
2) the riser-reactor thermodynamic analysis required at the hydrocarbon conversion schemes formation stage for assessing the hydrocarbon groups reactivity and the possibility of the reaction under the process conditions ($\Delta_r G^o 810$), as well as for solving the process heat balance equation ($\Delta_r H^o 810$).

3) the processes in the riser-reactor hydrodynamic regularities study, which is necessary for the selection of approximate (model) flow movement structure and ensuring real flow physical essence reflection in the reactor.
4) study of the cracking catalysts deactivation process, including deactivation by coke and heavy metals;
5) kinetic regularities study and the differential equations system development for the material and thermal balances of the processes in the riser-reactor and the regenerator.
6) the catalytic cracking mathematical model adequacy analysis.

Therefore, in order to predict the catalytic cracking unit performance when the feedstock hydrocarbon composition changes, the first and most important step is to study the heavy feedstock fractions and the process products composition.

The aim of the work is to study the effect of Kazakhstan, and West Siberian vacuum distillates mixture hydrocarbon composition on the catalytic cracking products yield using the process mathematical model.

2. Samples and research methods

The object of the research is the vacuum distillate catalytic cracking unit. The schematic representation of the unit is presented in Figure 1. The feedstock of the catalytic cracking process is a vacuum distillate, which is characterized by a wide fractional composition of 290–540°C with a density of 0.880–0.920 g/cm^3.
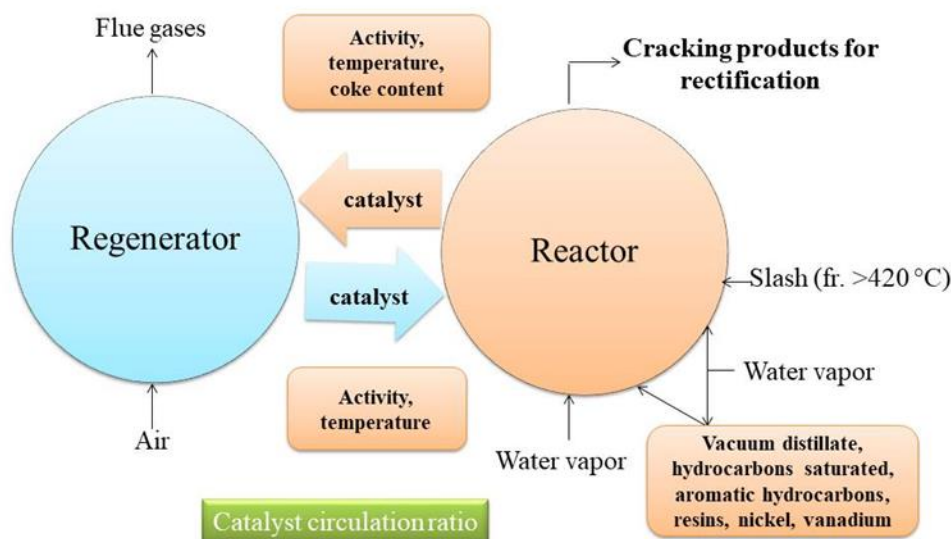

Fig. 1. The reactor and regenerator parameters relationship

A synthetic microspherical zeolite-containing catalyst with an average bulk density of 880.0 kg/m^3 and a pore volume of 0.450 cm^3 is used as a catalyst for the catalytic cracking process of the unit.

The main products of the process after the rectification stage are wet gas, gasoline fraction, fr. 195-340°C, fr. 340$^+$°C. With an increase in the aromatic hydrocarbons and resins concentration in the process feedstock, the coke yield, a by-product, significantly increases; as a result, the catalyst activity decreases, affecting the yield of target products.

To establish and take into account the processed feedstock composition influence on the yield and composition of the catalytic cracking products, information on the group composition of the heavy oil fractions processed in the catalytic cracking process is required. For that purpose

experimental studies of catalytic cracking vacuum distillate samples (297-540°C) and process products samples (heavy fractions fr. 195-340ºC, fr. 340⁺ºC) were carried out, as well as for coked and regenerated catalyst samples to assess the amorphicity degree of coke formed on the catalyst during cracking.

3. Research methods

The following methods were used to conduct experimental studies to establish the composition and physicochemical parameters of the catalytic cracking process feedstock and products, as well as the properties of the catalyst:

1) Liquid adsorption chromatographic (LC) analysis method using activated silica gel (grain size 0.2–0.5 mm) to separate heavy fractions of the feedstock and process products into paraffin-naphthenic, aromatic hydrocarbons and alcohol-benzene resins (the accuracy of the method is ± 5% - with the content of the determined component (50–70) wt%, ± 10% with the content of the determined component (15–50) wt% and ± 20 wt% - with the content of the determined component less than 15%).

2) GOST 18995.2-73 «Liquid chemical products. Method for determination of refractive index» using an Atago NAR-3T refractometer to measure the refractive index of hydrocarbon fractions by the optical method (measuring range of refractive index 1.3–1.7 nD, measurement accuracy ± 0.0001 nD, temperature 5–50°C, the accuracy of temperature measurements ± 0.2°C);

3) Cryoscopic method for determining the molecular weight (MM) of catalytic cracking process feedstock and products using the KRION-1 laboratory equipment (accuracy of temperature measurements is 0.001°C, accuracy index ± 9%);

4) Determination of the petroleum products kinematic viscosity is carried out in accordance with GOST 33-2000. The basis of the method is the measurement of the flow time of a certain volume of liquid under the action of gravity through a calibrated glass capillary viscometer. The permissible relative error of measuring viscosity is ± 0.35%; the volume of the studied samples is at least 3.0 mL.

5) Methods of structural group analysis: the Hazelwood method for determining the structural group composition of the catalytic cracking process feedstock and products aromatic hydrocarbons and the N-d-M method for determining the structural group composition of the catalytic cracking process feedstock and products saturated hydrocarbons (reproducibility of the method in determining the distribution of carbon is 1.5%, the number of rings - 0.1 units);

6) NETZSCH STA 449 F3 is a combined TGA/DSC analyzer, which allows you to simultaneously record changes in the mass of the sample and processes accompanied by the release or absorption of heat. The samples are heated from 50 to 1200 °C at a speed of 10 deg/min in an atmosphere of air, in corundum crucibles. The reproducibility of the device is ± 2%; the sensitivity, according to the DSC signal is 1 μW [23].

7) The mathematical modeling method for studying the feedstock composition influence on the catalytic cracking products yield [24].

4. Results of the study and discussion

4.1. Physical-chemical studies of the vacuum distillate catalytic cracking process feedstock and products composition and properties

Table 1 presents the content determination of the resins, saturated and aromatic hydrocarbons, density, molecular weight, and refractive index of catalytic cracking feedstock and heavy products of the process (results of liquid adsorption chromatography). In addition, the refractive indices of the selected fractions of saturated and aromatic hydrocarbons were measured to study their structural group composition. Moreover, in fractions of oil with a boiling point above 200°C, cyclic hydrocarbons have a mixed (hybrid) character. Typically, cyclic hydrocarbons contain paraffin side chains, and partly both naphthenic and aromatic rings.

Table 1. The results of the catalytic cracking process feedstock and heavy products samples physical-chemical features measurements

Hydrocarbon group/Feature	Value	
	№1	№2
Feedstock		
Saturated hydrocarbons, [% wt]	59.79	58.95
Aromatic hydrocarbons, [% wt]	35.46	36.61
Resins, [% wt]	4.75	4.44
Density, [kg/m^3]	0.8892	0.888
Saturated hydrocarbons refractive index	1.4552	1.4548
Aromatic hydrocarbons refractive index	1.5308	1.5214
Molecular weight, [g/mol]	362.88	312.85
Crude fraction refractive index at 50°C	1.4845	1.4830
Fraction 195-340°C		
Saturated hydrocarbons, [% wt]	25.91	27.09
Aromatic hydrocarbons, [% wt]	73.75	72.38
Resins, [% wt]	0.34	0.53
Density, [kg/m^3]	0.9652	0.9652
Molecular weight, [g/mol]	154.78	150.64
Refractive index at 50°C	1.5554	1.5564
Fraction >340°C		
Saturated hydrocarbons, [% wt]	6.99	8.59
Aromatic hydrocarbons, [% wt]	85.52	84.32
Resins, [% wt]	7.49	7.09
Density, [kg/m^3]	1.0592	1.0575
Molecular weight, [g/mol]	186.03	178.34
Refractive index at 50°C	1.7034	1.7018

Experimental studies have shown that with an increase in the boiling point of the fraction, the group composition of the feedstock and molecular weight (312.85–362.88 g/mol) change even with a slight change in density (0.892–0.894 g/cm^3). The unit's feedstock refractive index at 50°C varies in the range of 1.48300–1.4845, and the ratio of saturated hydrocarbons to aromatic hydrocarbons in the fractions of the samples have a close value of 1,686 for feedstock 1 and 1,61 for feedstock 2. With an increase in the density and molecular weight of the process feedstock, the content of resinous components increases (4.75 and 4.44 wt% for feedstock No. 1 and No. 2, respectively).

The 195-340°C fraction and the 340$^+$°C fraction of catalytic cracking are characterized by a high aromatic hydrocarbons content (72.38–73.75 wt% and 84.32–85.52, respectively). The densities of heavy products are close, while the molecular weights of the fractions 340$^+$°C differ significantly (186.03 and 178.34 for feedstock No. 1 and No. 2, respectively), which is associated with a high resinous and aromatic components content.

The use of structural-group analysis for catalytic cracking feedstock made it possible to determine the static distribution of structural elements in saturated and aromatic concentrates regardless of how these elements are combined in the molecules. The methods of structural group analysis made it possible to get an idea of the "average molecule" of a studied fraction and to determine the distribution of carbon (carbon atoms content (%) in aromatic, naphthenic, and paraffin chains) and the content of the ring in oil fractions. By determining the content of the ring, it is meant to find the number of aromatic and naphthenic rings in the average molecule or fraction average. So, this number expresses the fraction cyclicity degree, in this case, in saturated and aromatic concentrates. Since samples with small amounts of aromatic and saturated concentrates were studied, the calculated formulas of VP. Isaev, VV. Ilyinskaya, R. Hersha and M. Fenske, were used to determine the density and molecular weight, respectively.

Table 2 presents the studied catalytic cracking feedstock saturated hydrocarbons structural group composition.

Table 2. The structural-group composition of the catalytic cracking feedstock saturated hydrocarbons

Feature	Value	
	№1	№2
Carbon content, [%]		
– in paraffinic structures	73.2	72.4
– in naphthenic structures	26.8	27.6
– in aromatic structures	0.0	0.0
Average number, ea.		
– aromatic rings	0.0	0.0
– naphthenic rings	1.6	1.6
Total number of rings	1.6	1.6
The carbon proportion ratio in paraffin and naphthenic structures of the fraction	2.7	2.6

According to the structural group composition of saturated hydrocarbons, it was determined that the carbon proportion ratio in the paraffinic and naphthenic structures of the fraction is close in values (2.6-2.7 units), the average number of naphthenic rings in the saturated hydrocarbons fraction is 1.5 units.

The results of the catalytic cracking process feedstock aromatic concentrates structural-group analysis using the Hazelwood method (Table 3) made it possible to establish the catalytic cracking process feedstock aromatic cyclicity degree and to evaluate the carbon content in aromatic structures.

Table 3. The vacuum distillate aromatic hydrocarbons structural-group composition

Feature	Value	
	№1	№2
Carbon content, [%]		
– in paraffinic structures	34.0	31.3
– in naphthenic structures	21.2	21.9
– in aromatic structures	44.8	46.8
Average number, ea.		
– aromatic rings	1.8	1.5
– naphthenic rings	1.4	1.4
Total number of rings	3.2	3.0
The carbon proportion ratio in paraffin and naphthenic structures of the fraction	1.6	1.43

With an increase in the fraction molecular weight, the carbon proportion in the aromatic structures of the aromatic concentrates fraction is higher than −34.0%. With an increase in the catalytic cracking process feedstock molecular weight from 312.85 to 362.88 g/mol, the carbon content in the aromatic structures of the aromatic concentrates fraction increases from 31.3 to 34.0%. Since the carbon in paraffin structures includes both the carbon of paraffin molecules and the alkyl radicals of cyclic hydrocarbons the presence of naphthenic-aromatic structures and aromatic hydrocarbons with alkyl substituents in aromatic concentrates was established, the ratio of carbon in aromatic and naphthenic fragments is 1.43–1.60, and 0.67–0.76 in aromatic and paraffin fragments. Moreover, the degree of the feedstock cyclicality increases with increasing molecular weight, the total number of rings is 3.2 for feedstock No. 1 and 3.0 for feedstock No. 2, of which 1.4 is rings of naphthenic structure.

Differences in the cracking feedstock composition and features have a significant effect on the degree of coke formation on the catalyst, the temperature mode in the reaction – regeneration cycle, and hence the yield and composition of the products. At high catalytic cracking temperatures, resinous compounds almost completely condense to form coke and deactivate the catalyst.

The results of the coked and regenerated catalysts experimental study made it possible to establish the amount and structure of coke [25] deposited on the catalyst in the riser-reactor (Figures 2, 3).
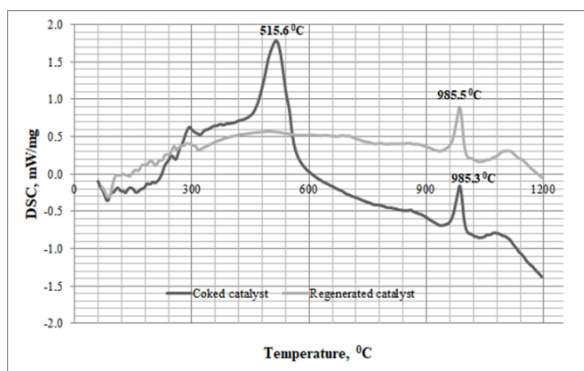

Fig. 2. Comparative thermogram of coked and re-generated catalysts differential scanning calorimetry curves
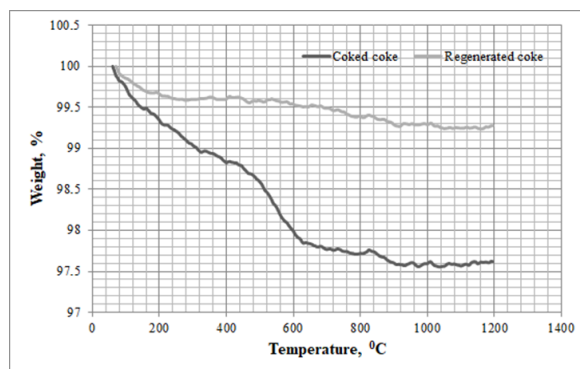
Fig. 3. Comparative thermogram of coked and regenerated catalysts thermogravimetric curves

On the DSC curve, at the initial stage of the analysis, the desorption of vapors and hydrocarbons entrained with the catalyst from the desorption zone occurs, as evidenced by the endothermic peak at a temperature of 84.0°C for the coked catalyst and 86.9°C for a regenerated catalyst. At a temperature of 515.6°C – for the coked catalyst and 433.3°C – for a regenerated catalyst, pronounced exothermic peaks characterizing the removal of carbon deposits are observed, and coke deposits have an amorphous (porous) structure since the temperature range of coke oxidation does not exceed 740°C. The DSC curve shows a pronounced exothermic peak at temperatures of 985.3 - 985.5°C, which corresponds to the destruction of the crystalline structure of the zeolite-containing catalyst, microcrystals are sintered.

A comparative TG-DSC analysis of the coked and regenerated catalysts at the crystal lattice destruction temperature and the sample constant mass showed that on the coked catalyst, sampling weight is reduced by 0.37% as a result of steam moisture desorption and by 2,03 % as a result of coke oxidation and destructive processes. While in the regenerated catalyst, the values of these quantities are 0.18% and 0.53%, respectively, which indicates the almost complete absence of coke on the regenerated catalyst and the high efficiency of the catalyst regeneration stage. The temperature of regeneration does not exceed 730°C due to the destructive processes.

To quantify the feedstock composition effect on the yield of coke and target products, the mathematical model of the catalytic cracking process was used, which is based on the hydrocarbon groups transformation reactions, it makes it possible to predict the yield and composition of the products depending on the feedstock composition. Using the mathematical model of the process, a study was carried out on the influence of the feedstock composition on the catalytic cracking yield and products composition, as well as the coke content on the catalyst.

4.2. Assay of the feedstock composition influence on the yield of target products and coke formation using a mathematical model of the vacuum distillate catalytic cracking process

In general terms the mathematical model is represented by a system of ordinary differential equations of material and thermal balances with initial conditions $C_i = C0, T0 = Tt.e.$:

$$\begin{cases} \dfrac{dC_i}{d\tau} = \sum_{j=1}^{18} \pm W_j \\[3mm] \rho_{cm} c_{cm} \dfrac{dT}{d\tau} = \sum_{j=1}^{18} \pm (\Delta_r H_T^{\circ})_j \cdot W_j \end{cases} \tag{1}$$

where C_i – the concentrations of the i-th group of hydrocarbons, mol/m^3; τ – the contact time; s; Tt.e. – the thermal equilibrium temperature of the feedstock and the catalyst, K; T – the temperature of the stream, K; W – the total reaction rate, taking into account both the direct and reverse channels of the transformation, mol/(s · m3); $\Delta rH°T$ – the thermal effect of the reaction, kJ/mol; ρcm, ccm – density, kg/m^3, and heat capacity of the flow, kJ/kg, K.

The use of a mathematical model of the catalytic cracking process allows a comprehensive assessment of the feedstock composition influence on the yield and composition of the products. The feedstock composition for model calculations was selected by the results of liquid adsorption chromatography of the catalytic cracking process feedstock and is presented in Table 4. The main process conditions were maintained at the short-term level (Table 5). The calculation results are presented in Figure 4.

Table 4. The cracking feedstock composition for model calculations

Hydrocarbon group/Feature	Content, [% wt.]				
	№1	№2	№3	№4	№5
Saturated hydrocarbons	59.79	58.95	63,89	66,83	73,3
Aromatic hydrocarbons	35.46	36.61	33,47	30,24	23,1
Resins	4.75	4.44	2,64	2,93	3,6
The ratio of saturated hydrocarbons to the sum of aromatics and resins, $C_{SH}/(C_{AH}+C_R)$	1.44	1.49	1.77	2.01	2.75

Table 5. Main process conditions of the riser-reactor R-201

Process conditions	Value
Feedstock consumption. [m^3/h]	202.76
Feedstock temperature. [ºC]	298.56
Sludge flow into the reactor. [m^3/h]	10.63
The water vapor flow rate to the reactor gripper. [kg/h]	6163.7
Water consumption for feedstock spraying. [kg/h]	2399.65
Regeneration outlet temperature. [ºC]	661.14
Cracking temperature. [ºC]	530.02
Reactor pressure. [Pa]	141215.8
Catalyst circulation rate. [$t_{cat}t/t_{feed}$]	7.23

Using a mathematical model. it was found that when the ratio of saturated hydrocarbons to the sum of aromatics and resins increases. the yield of wet gas increases by 1.64 (from 35.64 to 36.98% wt.). the gasoline fraction yield increases by 2.62% wt. (from 43.03 to 45.65% wt.). the yield of fr. 195–340ºC and fraction 340$^+$ºC decreases by 1.16 % wt. (from 8.12 to 6.96% wt.) and by 1.93% wt. (from 8.07 to 6.14% wt.). respectively.

When processing feedstock with a high content of resinous components (feedstock No. 1 and No. 2). the coke yield is significantly higher (5.14 and 4.82%) relative to other feedstock types (3.14–4.27 wt%). Since resinous components condense at high rates to form coke structures. an increase in the resin content significantly affects the degree of coke formation on the catalyst. and therefore. the yield of the target products.

The minimum coke content was found during the processing of feedstock No. 3 with a low content of resinous components (2.64% wt). the gasoline fraction yield for such feedstock was high (44.5 wt%). despite the low ratio of saturated hydrocarbons to aromatic hydrocarbons (1.77 units).
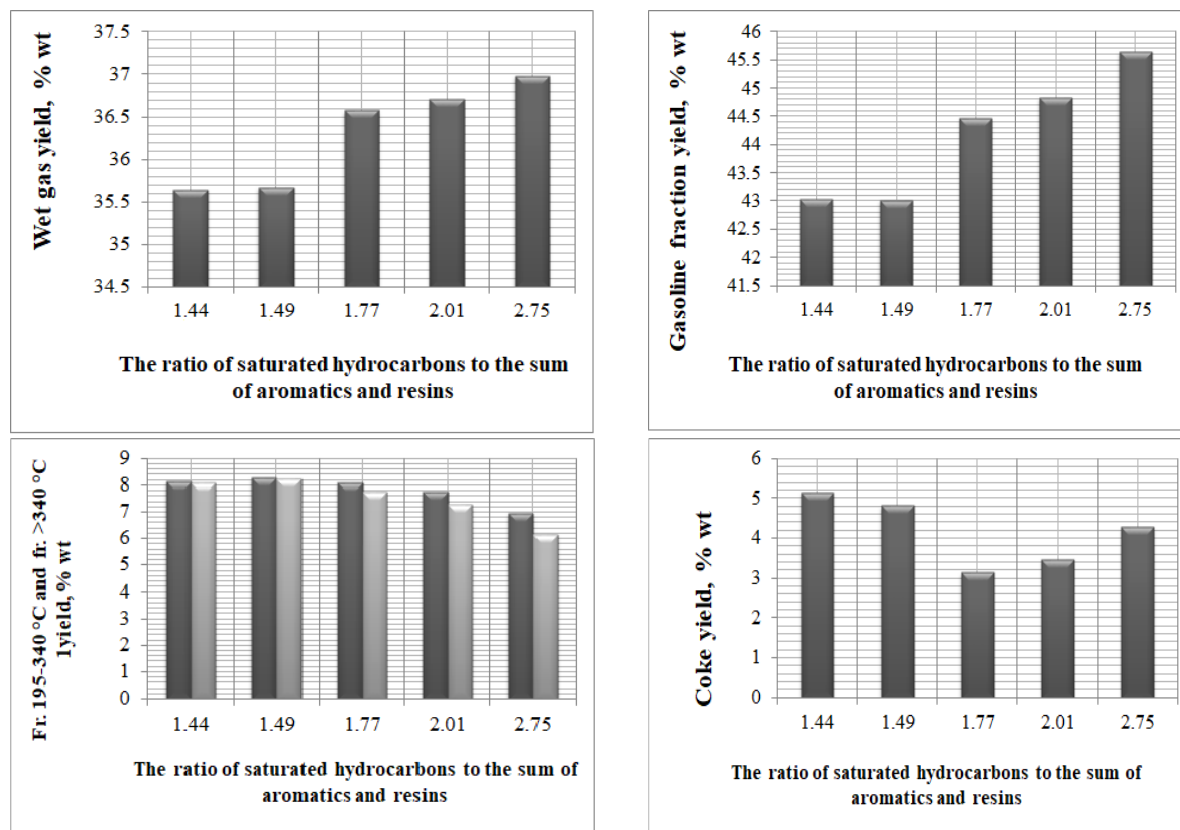
Fig. 4. The cracking products yield dependence of feedstock compositions

Thus. it was found that the high content of saturated hydrocarbons in the feedstock provides a high gasoline fraction yield during cracking and isomerization of alkanes. hydrogen transfer. cracking. and dealkylation of high molecular weight cycloalkanes. At the same time. the high content of aromatic hydrocarbons and resinous compounds in the process feedstock. as well as high rates of hydrogen transfer reaction. increase the coke content on the surface and in the pores of the catalyst. which leads to a decrease in its activity and the feedstock conversion degree.

5. Conclusion

The use of mathematical models built on the basis of thermodynamic and kinetic regularities of processes reactions involving high molecular weight hydrocarbons provides a prediction of the feedstock composition influence on the process products yield. The studies made it possible to establish the group and structural-group composition of the catalytic cracking process feedstock and heavy products. It was determined that the hydrocarbon composition of the processed feedstock. in particular. the high content of resinous compounds. significantly affects the rate of target and side reactions of the process. Upon the resinous components content change in the feedstock in the range of 2.64–4.75 wt%. the yield and coke content on the catalyst changes in the range of 3.14–5.14 wt% and 0.43–0.71 wt%. respectively. ceteris paribus conditions. It was shown that feedstock with a high content of saturated hydrocarbons (feedstock No. 4) provides the highest yield of gasoline fraction (45.65 wt%) and wet gas (36.98%).

In the future. the results of this paper will be used to develop a methodology for calculating the group composition of the catalytic cracking feedstock. depending on the feedstock quality. which is carried out in factory laboratories on a real-time basis. To adapt the calculation

method. it is necessary to replenish the process feedstock features database. which will subsequently make it possible to predict the feedstock composition on the basis of data on the fractional composition. density. molecular weight. refractive index. etc.

Carrying out predictive calculations using the model it will allow to adjust the reactor operating modes. to increase the yield of the target product while maintaining high octane rating and reducing excess coke formation depending on the feedstock composition and the process conditions of the regeneration process in terms of the regenerated catalyst temperature and its activity.

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