Catalysts of the catalytic cracking process and development of a fourteen-component kinetic model

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Abstract. The catalytic cracking process is specific and depends on several technological factors such as temperature, pressure, type of catalyst and reactor, and contact time. This article presents a review of foreign industrial zeolite-containing catalysts RDM SWM, and Russian microspherical catalyst "Oktifayn", granular cracking catalysts "adamant". On the basis of experimental and production data it is possible to conduct mathematical modeling of catalytic cracking process, and to select constants of rates of chemical transformations for each of types of catalysts.

1. Introduction

Today, all over the world there is a need to obtain high-quality motor gasoline. One of the ways to produce gasoline is the process of catalytic cracking, the purpose of which is to obtain a component of automotive gasoline with a high octane number. In addition, catalytic cracking produces dry gas, propane-propylene and butane-butylene fractions, light and heavy gasoil.

Over a long period of development, catalytic cracking is significantly improved both in terms of method of contact of the feedstock and the catalyst (in the stationary layer, to the moving layer of the ball of the catalyst, in the fluidized-bed microspherical catalyst in the apparatus with the elevator-reactor) and in respect of the catalysts (tablets based on natural clays, ball synthetic aluminosilicates, microspheric aluminosilicates, including zeolite). These improvements led to changes in the process technology to obtain the maximum possible amount of gasoline [1].

The choice of the process catalyst plays an important role, since the output and quality of the target products and the speed of the process depend on the selected catalyst.

Currently, the leading position is occupied by zeolite-containing catalysts, which include:
- matrix, consisting of amorphous aluminosilicate;
- active component, which is used as zeolites of type X, Y and ZSM-5;
- a number of additives that increase the activity of the catalyst [2].

This article provides an overview of a number of catalysts: RD-DMS-PM, "Oktifayn", "Adamant Super", "Adamant Extra" and "Lux".

2. Foreign catalysts

Company BASF produces an equilibrium zeolite-containing catalyst RD-DMS-PM [3] with the following parameters: bulk density of 0.97 g/cm\(^3\), specific surface area of 91 m\(^2\)/g, average pore
diameter of 10.1 nm, fractional composition (mcm) of 20-149, consisting mainly of oxides, wt. %. As additives, it is possible to use zeolite CVM (JSC «AZKiOS» Angarsk, Russia) and additives to increase the yield of olefins MOA («BASF» company). On the basis of the RD-DMS-PM catalyst and zeolite additives, catalytic mixtures can be prepared by mechanical mixing, in which the CVM content varies from 10 to 40 wt.% , and MOA - from 4 to 16 wt.%.

Using zeolite additives MOA and CVM in RD-DMS-PM significantly increases the yield of gaseous products. The content of PPF and BBF in the reaction mixture increases, while the content of gasoline decreases. On a catalyst with CVM zeolite the maximum gas yield (37 %) was obtained when the catalytic system contained 8 % CVM. The gas consists of fractions: ethane-ethylene (EEF), propane-propylene (PPF) and butane-butylene (BBF), which contain light olefins C₂—C₄.

On catalysts with a large number of additives, there is a sharp decrease in the yield of the gasoline fraction — from 42 to 26 %.

Table 1. Composition of liquid products in vacuum gas oil cracking

| Catalyst                        | Aromatic hydrocarbon | Alkanes+olefins+cyclic hydrocarbons |
|---------------------------------|----------------------|-------------------------------------|
| RD-DMS-PM                       | 20                   | 80                                  |
| RD-DMS-PM+20% MOA               | 40                   | 60                                  |
| RD-DMS-PM+8% CVM                | 70                   | 30                                  |

Analysis of the composition of liquid cracking products shows that the introduction of additives significantly reduces the proportion of isoparaffins, n-parafins, olefins and naphthenes, and increases the proportion of aromatic hydrocarbons (GTX - gasoline, toluene, xylene).

The production technology of Engelhard catalysts [4] has a number of features. Kaolin clay is used as the catalyst matrix. The surface of the matrix is catalytically active due to the fact that zeolite crystallizes on it, and thanks to the developed system of macropores, it cracks the residual raw material well. However, due to the imperfect technology of catalyst synthesis, the structure of large pores is such that the matrix activity is not selective and contributes to excessive coke- and gas formation.

Grace Davison [4] has improved high-performance matrices with the brand names SAM and TMA. Their specificity is that they are prepared using a special technology based on active aluminum oxide. This allowed us to develop a new generation of cracking catalysts of the Crystal, Futura and Brilliant series, which are distinguished by increased activity and high selectivity for coke and gas. The characteristics of some catalysts are shown in table 2.

Table 2. Characteristics of some cracking catalysts

| Name of indicators | Manufacturer and brand of catalyst | Kristal-242 | Futura-140 | Brilliant-242 | Advans-937 | Kobra | NaphthaMax |
|--------------------|-----------------------------------|-------------|------------|---------------|------------|------|------------|
| Content, % wt.     | Grace                             | 49          | 42         | 49            | 43.5       | 44.4 | 44         |
| Al₂O₃              | Akzo-Nobel                         | 49          | 42         | 49            | 43.5       | 44.4 | 44         |
| Na₂O               | Enhelhard                          | 2.8         | 2.7        | 2.8           | 2.32       | 1.97 | 2.4        |
| P₂O₃              |                                    | 280         | 290        | 300           | 99         | 134  | 162        |
| Specific surface area, m²/g |                      | 700         | 700        | 710           | 870        | 940  | 860        |
| Bulk density, kg/m³ |                                    | 79          | 79         | 79            | 69         | 72   | 72         |
| Microactivity      |                                    | 79          | 79         | 79            | 69         | 72   | 72         |
3. Russian catalysts

The authors of the work [5] proposed the technology for manufacturing the microspheric catalyst "Octifine". The proposed catalyst contains zeolite Y in a mixed ion-exchange form and a matrix consisting of aluminum oxide, kaolin and silicon dioxide. The microspherical cracking catalyst consists of an active component - zeolite Y, characterized by a lattice module and presented in a different cation - decated form, in particular HY, ReHY and ReY and a matrix that serves as a carrier in which the active component is evenly distributed. The use of this catalyst will allow obtaining a high yield of the gasoline fraction, which is achieved due to the unique structure of the catalyst. The catalyst consists of 0.27% Na₂O and 2.3% rare earth element oxides. The specific area of the catalyst

The article [6] reports on the development of a granular cracking catalyst "Adamant" represented by two main brands: "Adamant Super" and "Adamant Extra".

"Adamant Super" is designed for processing vacuum gas oils with maximum output of gasoline and light products. REHY zeolite, which has a specially selected chemical composition and micro-, mesoporous structure, is the basis of the catalyst. Zeolite is resistant to high temperatures and catalytic poisons, has a low selectivity for coke and dry gas and a high selectivity for gasoline. The catalyst has a large pore volume and a porous structure that is favorable for cracking heavy hydrocarbon molecules.

"Adamant Extra" is designed for processing vacuum gas oils with a maximum yield of light olefins, gasoline and light gas oil. Also, the catalyst allows you to get a high octane number of gasoline. RE-USY zeolite, which has specific acidic properties, is the basis of the catalyst. The low density of acid centers in zeolite minimizes hydrogen transfer reactions and contributes to an increase in the content of olefins and aromatic hydrocarbons in fat gas and gasoline, naphthenes in light catalytic gas oil, as well as an increase in the octane and cetane numbers of gasoline and light gas oil, respectively.

Table 3. Test results in the cracking process of the "Adamant Super" and "Adamant Extra" catalysts

| Indicator                          | Actual value |
|------------------------------------|--------------|
|                                    | "Adamant Super" | "Adamant Extra" |
| Conversion, wt. %                  | 50.3         | 42.3           |
| Output for raw materials, wt. %    |              |                |
| gas (H₂, C₁-C₄)                    | 15.8         | 12.2           |
| gasoline (C₃-150 C)                 | 31.3         | 27.2           |
| light catalytic gas oil (150 – 360 C) | 42.6         | 45.4           |
| heavy catalytic gas oil (>360 C)     | 7.1          | 12.3           |
| coke                               | 3.2          | 2.9            |
| Total output of light oil products  | 73.9         | 72.6           |

Note. Test conditions: catalyst capacity 50 cm³; specific feed rate 1.8 h⁻¹; temperature 460 C.

Table 3 shows that the total output of light oil products is higher, and the output of heavy catalytic gas oil is lower.

In the article [7], the authors present a new series of effective vacuum gas oil cracking catalysts. The development is based on the "Lux" catalyst with an additional active component - ZSM-5 zeolite in a different ratio with the ultra-stable zeolite of the NRZEU. Bicelolite cracking catalysts increase the octane number of cracking gasoline by partially cracking normal paraffins and increasing the selection of propane-propylene and butane-butylene fractions with a slight decrease in the selection of the gasoline fraction. The amount of reduction in selection depends on the content of ZSM-5 type zeolite in the bicelolite catalyst.

As can be seen from table 4, when the zeolite content of ZSM-5 in the catalyst composition increases to 8% by weight, the selection of cracking gasoline is significantly reduced with a proportional increase in the selection of propane-propylene (PPF) and butane-butylene (BBF) fractions. At the same time, the increase in raw material conversion is associated with a decrease in the selection of light and heavy gas oils. The yields of dry gas and coke with an increase in the zeolite content of ZSM-5 in the catalyst composition remain virtually unchanged. The obtained data show that an increase in the selection of
PPF and BBF without a significant decrease in the yield of the gasoline fraction can be achieved by involving 2 to 4% of ZSM-5 zeolite in the composition of the "Lux" series catalyst.

Table 4. The extraction of gasoline, PPF and BBF (wt. % ) with different contents of zeolite type ZSM-5 in the composition bitolithic catalysts

| Selections | The content of zeolite ZSM-5 wt.% |
|------------|----------------------------------|
|            | 0  | 2  | 4  | 6  | 8  |
| PPF        | 9.0| 9.4| 9.5| 10.2|11.2|
| BBF        | 15.4| 15.9| 15.9| 16.4|17.6|
| Gasoline   | 45.0| 44.6| 44.4| 42.5|40.9|
| The conversion of raw materials | 79.0| 79.3| 79.6| 80.2|81.3|

In the article [8], the authors considered a new domestic zeolite-containing y-type catalyst based on the technology of aerosol nanocatalysis in a vibro-liquid layer, in which the catalytic system consists of a catalyst with a particle size from 200 to 300 microns and a dispersing material consisting of glass balls with a diameter from 1 to 1.2 mm. When the catalyst particles are dispersed, nanoparticles with a hyperactive surface are formed. The new catalyst made it possible to increase the selectivity of the process for light oil products.

Table 5. Results of experimental studies of catalytic cracking on a zeolite-containing Y-type catalyst

| №  | t, C | f, Hz | Composition of cracking products, % wt. | X, | F, | x, | The speed of cracking |
|----|------|------|----------------------------------------|----|----|----|----------------------|
|    |      |      | coke | gas | gasoline fraction | diesel fraction | kg/(m²p*h) | kg/(kgcat*h) |
| 1  | 450  | 4    | 1.4  | 0   | 7.2  | 28.2  | 36.8  | 96.2  | 35.4  | 541.6 | 180533 |
| 2  | 500  | 4    | 1.7  | 2   | 2.6  | 7.9   | 14.4  | 72.5  | 10.5  | 160.1 | 53360  |
| 3  | 550  | 2    | 1.4  | 0   | 10.7 | 42.4  | 59.6  | 89.1  | 53.1  | 812.1 | 270703 |
| 4  | 450  | 1.3  | 0    | 1.4  | 14.1 | 15.4  | 91.5  | 14.1  | 215.0 | 71653  |
| 5  | 500  | 4.5  | 1.4  | 0   | 2.3  | 9.7   | 13.4  | 72.6  | 9.7   | 148.5 | 49503  |
| 6  | 550  | 1.5  | 4.8  | 0   | 8.4  | 14.7  | 57.2  | 8.4   | 128.9 | 42959  |
| 7  | 450  | 1.3  | 0.3  | 3.7 | 21.4 | 26.6  | 94.0  | 25    | 382.5 | 127500 |
| 8  | 500  | 5    | 1.4  | 1.7 | 6.5  | 17.7  | 27.2  | 88.6  | 24.1  | 369.1 | 123033 |
| 9  | 550  | 1.8  | 5.4  | 6.7 | 13.7 | 27.5  | 73.9  | 20.4  | 311.4 | 103800 |
| 10 | 450  | 1.2  | 0    | 1.2 | 0    | 19.1  | 20.3  | 94.1  | 19.1  | 291.9 | 97290  |
| 11 | 500  | 5.5  | 1.2  | 0   | 4.7  | 37.2  | 43    | 97.2  | 41.3  | 639.5 | 213152 |
| 12 | 550  | 1.5  | 1.1  | 7.1 | 9.4  | 19.1  | 86.3  | 16.5  | 252.2 | 84057  |
| 13 | 450  | 0.9  | 0    | 5.4 | 15.6 | 21.9  | 95.9  | 21    | 320.6 | 106865 |
| 14 | 500  | 6    | 1.2  | 0   | 6.5  | 20.8  | 28.5  | 95.8  | 27.3  | 417.2 | 139056 |
| 15 | 550  | 1.5  | 6.2  | 8.7 | 24.3 | 40.8  | 81.1  | 33    | 505.1 | 168355 |

As can be seen from table 5, the conversion of raw materials does not exceed 59.6% by weight in a single pass, but the selectivity is very high. The highest selectivity of the process is observed at temperatures of 450 and 500 °C. This is due to the fact that the formation of the gas fraction is insignificant under these conditions. Due to the achieved selectivity for light oil products, it is possible to significantly reduce costs at the stage of separation of oil products.
4. Analysis of the possibility of calculating

We have developed a fourteen component kinetic model of the catalytic cracking process (figure 1) [9]. This model will allow you to evaluate the yield and quality of products such as fat gas, gasoline, and catalytic gas oil.

In table 6 shows the designations of the components used in the kinetic model.

\[ \frac{d[y_1]}{dt} = -k_1[y_1] - k_{10}[y_1] - k_{12}[y_1] - k_{23}[y_1] \]
\[ \frac{d[y_2]}{dt} = -k_2[y_2] - k_{9}[y_2] - k_{13}[y_1] - k_{24}[y_2] \]
\[ \frac{d[y_3]}{dt} = -k_3[y_3] - k_{10}[y_3] - k_{14}[y_3] - k_{25}[y_3] - k_{11}[y_3] \]
\[ \frac{d[y_4]}{dt} = -k_4[y_4] - k_{15}[y_4] - k_{26}[y_4] \]

![Figure 1. Scheme of transformations in the fourteen-component kinetic model of the catalytic cracking process.](image)

Table 6. Designations and molecular weights of components of the kinetic model of catalytic cracking

| Designation | Title                                      | Molar mass, kg/kmol |
|-------------|--------------------------------------------|---------------------|
| Ph          | Heavy paraffins                            | 400                 |
| Nh          | Heavy naphthenes                           | 400                 |
| Ah          | Heavy aromatic compounds                   | 400                 |
| Cah         | Heavy compounds with aromatic substituents | 400                 |
| Pm          | Medium paraffins                           | 200                 |
| Nm          | Medium naphthenes                          | 200                 |
| Am          | Medium aromatic compounds                  | 200                 |
| CAm         | Medium compounds with aromatic substituents| 200                 |
| Pl          | Light paraffins                            | 100                 |
| Ni          | Light naphthenes                           | 100                 |
| Al          | Light aromatic compounds                   | 100                 |
| C           | Coke                                       | -                   |
| DG          | Dry gas                                    | 18.4                |
| G           | Wet gas                                    | $\approx 50$        |

On the basis of the transformation scheme shown in figure 1, a system of differential equations characterizing the kinetic model of catalytic cracking is compiled:
\[
\begin{align*}
\frac{d[y_3]}{dt} &= 2k_1[y_1] - k_5[y_3] - k_{16}[y_5] - k_{27}[y_3] \\
\frac{d[y_6]}{dt} &= 2k_2[y_2] - k_{16}[y_6] - k_{17}[y_6] - k_{20}[y_6] \\
\frac{d[y_7]}{dt} &= 2k_3[y_3] - k_7[y_7] - k_{10}[y_7] - k_{29}[y_7] \\
\frac{d[y_8]}{dt} &= 2k_4[y_4] + 2k_{11}[y_3] - k_{19}[y_8] - k_{30}[y_8] \\
\frac{d[y_9]}{dt} &= 2k_5[y_5] + 4k_8[y_1] - k_{20}[y_9] - k_{31}[y_9] - k_{34}[y_9] \\
\frac{d[y_{10}]}{dt} &= 2k_6[y_6] + 4k_9[y_2] - k_{21}[y_{10}] - k_{32}[y_{10}] - k_{35}[y_{10}] \\
\frac{d[y_{11}]}{dt} &= 2k_7[y_7] + 4k_{10}[y_3] - k_{22}[y_{11}] - k_{33}[y_{11}] - k_{36}[y_{11}]
\end{align*}
\]

\[
\begin{align*}
\frac{d[y_{12}]}{dt} &= k_{12}[y_1] + k_{13}[y_2] + k_{14}[y_4] + k_{15}[y_6] + k_{16}[y_8] + k_{17}[y_9] + k_{18}[y_9] + k_{19}[y_9] + k_{20}[y_9] + k_{21}[y_{10}] + k_{22}[y_{11}] \\
\frac{d[y_{13}]}{dt} &= k_{23}[y_1] + k_{24}[y_2] + k_{25}[y_3] + k_{26}[y_4] + k_{27}[y_5] + k_{28}[y_6] + k_{29}[y_7] + k_{30}[y_8] + k_{31}[y_9] + k_{32}[y_{10}] + k_{33}[y_{11}]
\end{align*}
\]

\[
\frac{d[y_{14}]}{dt} = k_{34}[y_9] + k_{35}[y_{10}] + k_{36}[y_{11}]
\]

As can be seen from the explanations in Table 6, the division into components is made taking into account their structural and mass composition, which makes it possible to evaluate the quantitative yields of gasoline, catalytic gas oils, fat gas and losses with dry gas and coke, and the quality indicators of the products obtained. We are interested in the process of catalytic cracking to obtain gasoline as the target product.

To create a database of constants for catalysts, it is also possible to calculate the catalytic cracking process with an increased yield of PPF and BBF using our fourteen-component model. Despite the fact that using zeolite additives MOA and CVM increases the yield of PPF and BBF in the reaction mixture, and gasoline - decreases; the new domestic zeolite-containing catalyst type Y - increases selectivity for light oil products, it makes sense to calculate the kinetics of transformations in this catalyst by a fourteen-component model.

Processes using “Lux” catalysts based on ultra-stable zeolite Y, “Adamant Extra”, and “Oktifayn” are also suitable for our model, since this results in an increase in the octane number of the gasoline fraction.

One of the goals of kinetic research is to choose the form of kinetic equations and find kinetic constants.

We solved direct and inverse problems based on data [7].

To solve the inverse problem, we used the search for minimizing the function of the MatLAB package. The calculation was made for the catalytic cracking process using the “Lux” catalyst.

To find the kinetic constants, the function of the sum of absolute deviation of experimental concentrations from calculated is minimized using the genetic algorithm of the MatLAB package. The results are presented in Table 7.

Based on the minimization data, we can conclude that the fourteen-component model well describes the process of catalytic cracking using various catalysts. Calculations have shown the feasibility of using a genetic algorithm. It can be argued that the choice of the MatLaB package affects the correctness of the results obtained.

In the future, it is possible to refine the universal kinetic model, which allows describing the process of catalytic cracking using any type of catalysts, as well as the selection of reaction rate constants for different types of catalysts.
### Table 7. Calculated kinetic constants of the catalytic cracking process using the "Lux" catalyst

| Components | Constants |
|------------|-----------|
| K1         | 4.8875    |
| K2         | 5.9044    |
| K3         | 0.10655   |
| K4         | 2.8031    |
| K5         | 7.6444    |
| K6         | 1.6104    |
| K7         | 5.4507    |
| K8         | 0.9794    |
| K9         | 0.043696  |
| K10        | 0.31803   |
| K11        | 0.31665   |
| K12        | 8.7216    |
| K13        | 3.9295    |
| K14        | 1.4669    |
| K15        | 2.5238    |
| K16        | 0.20479   |
| K17        | 0.046801  |
| K18        | 2.9029    |
| K19        | 0.18867   |
| K20        | 0.085212  |
| K21        | 0.11097   |
| K22        | 0.0025593 |
| K23        | 6.2816    |
| K24        | 3.4688    |
| K25        | 0.35569   |
| K26        | 1.7373    |
| K27        | 1.0916    |
| K28        | 0.059511  |
| K29        | 0.46295   |
| K30        | 0.0040027 |
| K31        | 0.080006  |
| K32        | 0.15746   |
| K33        | 0.0024528 |
| K34        | 0.011989  |
| K35        | 0.075628  |
| K36        | 0.97685   |

5. Conclusion
To date we can conclude that foreign catalysts are not inferior to the well-known analogues of the brand Kristal-242, Futura-140, Brilliant - 242 by Grace, and in Russian - "Adamant", and even surpass them in a number of parameters.

The principal difference between the KNT Group technology is that the zeolite is first crystallized and subjected to ion exchange treatments, and only then is added to the matrix. Therefore, the content of rare earth elements in the "Adamant" catalyst remains the same both before and after abrasion, and foreign manufacturers use the technology of crystallization of the zeolite component on the surface of pre-formed granules. During operation on industrial installations with a moving layer, the catalyst is abraded and its outer layers are removed. Together with the outer layers, the most active zeolite component is also removed, as a result, such a catalyst is deactivated faster than a catalyst in which the active component is evenly distributed throughout the volume.
Another feature of the "Adamant" series catalysts is the low residual sodium content (2-3 times lower than in imported analogues). The KNT Group technology gives relative freedom in the design of the catalyst composition. While foreign manufacturers pellets consist mainly of kaolin, and the zeolite content in the catalyst is regulated by the duration of crystallization, in the KNT Group technology, the matrix composition is more diverse, and the zeolite content can easily change at the molding stage. "Adamant" catalysts have higher hydrothermal stability. With multiple thermocouple treatments, they better preserve the specific surface area and catalytic activity. During the cracking process, "Adamant" catalysts demonstrate a high total yield of light oil products and a low yield of heavy catalytic gas oil.

The most significant feature of the production technology of cracking catalysts by Engelhard is the method of implanting zeolite into the catalyst particles. Most firms first mixing the zeolite and the matrix, and then subjected to spray-drying the resulting sol with getting the microsphere particles. Engelhard technology first produces matrix microspheres, and then crystallizes the zeolite inside these microspheres.

In connection with the transfer of modern catalytic cracking units to a mode of operation with a short contact time of raw materials with catalysts, all leading companies engaged in their synthesis began to produce industrial samples containing a significant number of large pores in their structure, which has a positive effect on the rate of diffusion of large hydrocarbon molecules to the active centers. The use of wide-pore catalysts is of particular importance, since in a reactor of this type of plant, the contact time of the catalyst with the raw material is calculated in fractions of a second.

Zeolite-containing catalyst RD-DMS-PM allows you to get PPF and BBF, and the catalyst "Adamant Super" - gasoline and light products. The maximum yield of light olefins, gasoline and light gas oil is possible when using the "Adamant Extra" catalyst, and the catalyst allows you to get a high octane number of gasoline. Domestic zeolite-containing catalyst type Y allowed to increase the selectivity of the process for light oil products. The use of a biceolite catalyst led to a decrease in the selection of the gasoline fraction, but the selection of PPF and BBF increased with an increase in their content of olefin hydrocarbons.

Catalysts of the “Lux” series based on ultra-stable zeolite Y have increased activity, thermal stability, improved mechanical properties, provide an increase in the octane number of the gasoline fraction, and also have increased wear resistance.

The task of developing a database of catalytic cracking transformation constants using various new and modified existing catalysts for further use in engineering and technological calculations of catalytic cracking reactors is urgent.

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