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Selective desulphurization and denitrogenation of hydrocarbon mixtures rich in olefins

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ABSTRACT

Selective desulphurization and denitrogenation of hydrocarbon mixtures rich in olefins (FCC-, pyrolysis- and coker naphtha) were investigated over three different catalysts (conventional and new generation $CoMo/Al_2O_3$, Pt(0.1%)Pd(0.3%)/USY). According to the experiments carried out in a wide range of operation parameters it was concluded that the sulphur removal efficiency of the PtPd/USY approximately equals to that of the new generation $CoMo/Al_2O_3$ up to about 200 mg/kg sulphur and 35 mg/kg nitrogen content, but the octane number loss is lower and the nitrogen removing efficiency is significantly better (nearly 100%) as well, than in case of the previous catalyst. In case of feedstock having higher sulphur and nitrogen content, the utilisation of $CoMo/Al_2O_3$ together with PtPd/USY in one reactor enables the production of very low sulphur (<5 mg/kg) and nitrogen (<2 mg/kg) containing products with low octane number loss.

1. Introduction

One of the most significant challenges in catalytic petroleum processing in the last 10–15 years has been to recognise catalysts and develop catalytic systems with high activity and selectivity and good stability to reduce significantly the sulphur content of the petroleum products, particularly in the area of fuels required by the environmental protection motives.

The reason of the sulphur content limitation is not only the formation of sulphur dioxide and sulphur trioxide which cause acid rains and corrosion, but the poisonous effect of those on the catalytic converters. Because of the efficiency loss the harmful matter emission increases significantly [1,2].

The limitation of the sulphur content of the gasolines to 10–15 mg/kg (EU, USA) [3,4] in the developed areas of the world required the reduction of the heteroatom content of the blending streams with relatively high sulphur content (>10 to 200 mg/kg).

Because of the increasing demand for gasolines it is necessary to use the gasoline fractions of high sulphur content produced in different thermal technologies, so their desulphurization is inevitable. These streams are of high olefin content (even of diolefin content), so applying the technological parameters (conventional $CoMo/Al_2O_3$ catalyst, process conditions) of the hydrodesulphurization technologies these olefins are hydrogenated to saturated hydrocarbons having significantly lower octane number, in addition the hydrogen consumption is remarkable. Generally 6–10 unit octane number loss is a considerable disadvantage significantly reducing the value of the product [5,6]. So for desulphurization of these gasoline fractions catalysts having new composition and/or new structure are needed which can selectively remove the sulphur atoms from the hydrocarbons with insignificant hydrogenation of the unsaturated compounds leading to octane number loss. Furthermore regarding the environmental requirements they should be applicable for denitrogenation to reduce the NO_x emission [3,7].

Numerous publications discuss the selective hydrodesulphurization of FCC-, pyrolisis- and coker naphthas, produced in fluid catalytic cracking (FCC), in steam pyrolysis of hydrocarbons for ethylene and propylene production and in thermal coking of heavy hydrocarbon residues of high molecular mass, respectively, but there is a little information about the experiments using zeolite catalysts [8,9] (selective hydrodesulphurization: deep desulphurization without the saturation of olefins and aromatics with minimum octane number loss).

So the main objective of our research work was to find a catalytic system (suitable zeolite catalyst(s), reaction parameters) which is applicable for the selective desulphurization and denitrogenation of gasoline fractions of high olefin content with minimum octane number loss. In this contribution the results obtained over

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Table 1
Main characteristics of the feedstocks.

Properties	Feedstocks		
	FCC HG (>100 °C)[a]	FCC HG + PG (10%)[b]	FCC HG + DCLG (5%)[c]
Density (15.6 °C) (g/cm^3)	0.8098	0.8183	0.7993
Sulphur content (mg/kg)	29/103[d] (152. 204. 306[e])	133	330
Nitrogen content (mg/kg)	14/33	32	37
Research octane number (RON)	92.1/92.4[d]	92.4	91.5
Motor octane number (MON)	79.4/79.5[d]	79.6	79.0
Hydrocarbon composition (%)			
n-Paraffins	3.4[d]	2.9	4.0
Isoparaffins	21.6[d]	21.1	22.1
Olefins	9.8[d]	8.9	11.5
Naphthenes	9.0[d]	9.2	8.8
Aromatics	56.2[d]	57.9	53.6
Distillation properties(°C)			
Initial boiling point (IBP)	98/101[d]	99	28
50 (V/V%)	147/149[d]	149	134
End boiling point (EBP)	210/213[d]	214	209

[a] Heavy FCC gasoline.

[b] Mixture of 90% heavy FCC gasoline and 10% pyrolysis gasoline.

[c] Mixture of 95% heavy FCC gasoline and 5% light coker gasoline.

[d] Two different values of sulphur and nitrogen content apply for heavy FCC gasolines obtained from different batches of FCC unit feed. Feed sulphur of 103 mg/kg was increased by adding benzothiophene.

[e] The sulphur content was raised by adding benzothiophene (BT) is the sulphur molecule occurring in the highest concentration in the FCC gasolines and it is also one of the most refractory ones.

PtPd/USY catalyst are presented and discussed compared with those of a conventional and a new generation CoMo/Al_2O_3 catalyst.

2. Materials and methods

The applied CoMo/Al_2O_3 catalysts were commercial products, while the PtPd/USY catalyst was a member of the proprietary developed ultra stable Y type zeolite family having expediently chosen composition [10], whose properties differed from the bimetal/zeolite catalysts investigated by us previously [11].

The main properties of the applied conventional CoMo/Al_2O_3 hydrogenation-hydrodesulphurization catalyst were: average diameter 1.3 mm; shape: quadrolobe; specific surface area: 220 m^2/g; pore volume 0.4 cm^3/g; molybdenum content: 11.2%; cobalt content: 3.1%. About the patented, confidential properties of the new generation CoMo/Al_2O_3 catalyst developed for the selective desulphurisation of FCC gasolines we have no right to report data.

The main properties of the applied USY zeolite support were: Si/Al ratio: 14.3; total surface area 720 m^2/g, whereof the mesoporous surface area: 89 m^2/g; total acidity 0.13 mmol NH_3/g. Catalysts were prepared by impregnation of the support with $[Pt(NH_3)_4]Cl_2$ and $[Pd(NH_3)_4]Cl_2$ then these were dried, calcined and shaped. The main properties are: shape: cylindrical; average length: 3.2 mm, diameter: 1.2 mm, metal dispersion: 68%; loading density: 0.5 g/cm^3; fracture strength: 6.2 N/m. Before the experiments the catalysts were "in situ" dried and reduced in hydrogen flow at 400 °C for 14 h. According to the results of the preliminary experiments for the used feedstocks the most favourable catalyst was of 0.3% palladium and 0.1% platinum content. For the characterisation of the catalysts the generally applied methods were used [10].

Table 1 contains the main properties of the used olefin rich gasolines. (The diolefin content of these gasoline fractions was selectively hydrogenated to monoolefin previously.) In order to investigate the effect of the higher sulphur concentration on the performance of the catalyst, the feed (FCC gasoline, "FCC HG") of 103 mg S/kg was spiked with benzotiophene (BT; Sigma–Aldrich) of technical grade to 152, 204 and 306 mg/kg.

The sulphur content of the FCC gasolines are low, because the feedstock of the FCC plants is highly desulphurized and/or hydrocracked in the modern refineries to reach favourable product composition and better quality products. For example the pro-

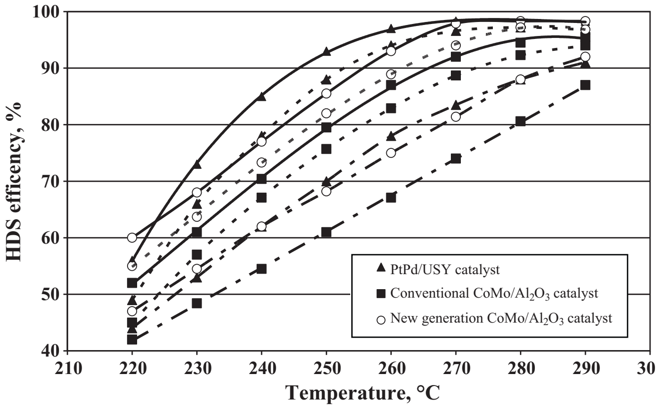

Fig. 1. HDS efficiency [$100(S_{c.feed} - S_{c.prod})/S_{c.feed}$] as a function of the temperature of different catalysts (sulphur in FCC HG feed: 103 mg/kg, *P*: 30 bar, H_2/HC: 250 Nm^3/m^3), where continuous line: LSHV: 1.5 h^{-1}; broken line: LSHV: 2.0 h^{-1}; dots and dashes: LSHV: 3.0 h^{-1}.

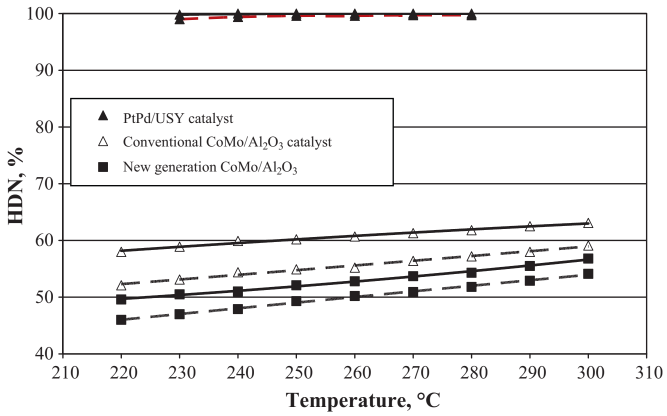

Fig. 2. HDN efficiency of the different catalysts (feed sulphur 103 mg/kg; pressure: 3.0 MPa; H_2/HC: 250 Nm^3/m^3), where continuous line: LSHV: 1.5 h^{-1}; broken line: LSHV: 2.0 h^{-1}.

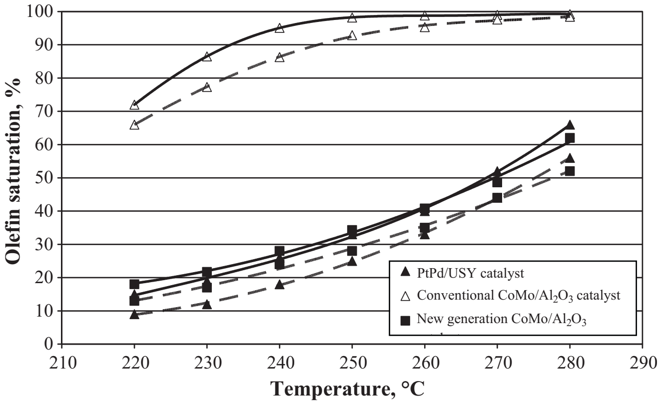

Fig. 3. Olefin conversion vs. reaction temperature on different catalysts (feed sulphur 103 mg/kg; pressure: 3.0 MPa; H_2/HC: 250 Nm^3/m^3), where continuous line: LSHV: 1.5 h^{-1}; broken line: LSHV: 2.0 h^{-1}.

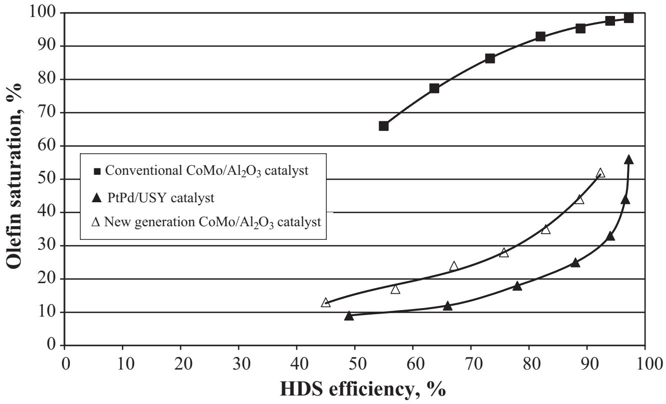

Fig. 4. Degree of olefin hydrogenation vs. HDS on different catalysts (feed sulphur 103 mg/kg; pressure: 3.0 MPa; H_2/HC: 250 Nm^3/m^3).

duction of gasoline blending components containing at the most 10 mg/kg sulphur is easier and more economical from the lower sulphur containing FCC gasoline fractions (20–200 mg/kg instead of 400–600 mg/kg).

The hydrocarbon composition of the feeds and products were determined with gas chromatograph, the composition of the vapour phase products (hydrocrack products) leaving the reactor was determined with gas chromatograph according to ASTM D 5134-98. Research and motor octane numbers of the feeds and products were calculated from the compositions by commercial software developed for this purpose. Other standard quality properties (density, distillation properties, vapour pressure, etc.) of the feeds and products were determined according to the methods of the EN 228:2009 unleaded petrol standard. The sulphur content of the feeds and products was measured with MULTI EA 3100 apparatus (Producer: ANALYTIC Jena, Vendor: GreenLab, Hungary) according to EN ISO 20846 standard, the nitrogen content was determined according to ASTM-D 6366-99 standard.

The experiments were carried out in a high pressure reactor system. The tubular reactor has 100 cm^3 effective volume and it is operated without back-mixing. The system includes all the process equipment and units which are applied in the reactor loop of industrial scale gasoline hydrotreating plants. Experiments were carried out under continuous operation conditions on catalysts having stabilized activity [12].

3. Results and discussion

The experiment series were carried out in a wide range of operation parameters (*T*: 180–310 °C; *P*: 20–40 bar; LHSV: 0.75–4.0 h^{-1}; H_2/hydrocarbon: 150–350 Nm^3/m^3) for the selective heteroatom removal from hydrocarbon mixtures of different origin. (The most important results are presented, only.) In case of different sulphur containing heavy FCC gasolines the yield of the liquid products were higher than 99.5% up to 280 °C and $\leq$1.5 h^{-1} liquid hourly space velocity (LHSV) for the applied conventional and new generation $CoMo/Al_2O_3$ catalysts as well as for PtPd/USY catalyst, independently from the other parameters. The generation of the gas products increased at higher temperature ($\leq$290 °C) and lower liquid hourly space velocity (>1.0 h^{-1}), especially on the PtPd/USY catalyst. The reason of this was the relatively higher acidity of this catalyst than that of the two other. The lowest liquid yields were obtained at 310 °C and 0.75 h^{-1} LHSV, which were: 98.8% on the conventional $CoMo/Al_2O_3$, 98.6% on the new generation $CoMo/Al_2O_3$ and 97.4% on the PtPd/USY catalyst.

At 20–25 bar total pressure and 150–200 Nm^3/m^3 H_2/hydrocarbon ratio it was found that under the same conditions after the usual initial activity decrease a slow activity decrease was observed after the first 72 h as well. The reason of this was the insufficient partial pressure of the hydrogen, since there was not enough disposable hydrogen to remove the sulphur from the surface of the noble metals. This was proved by carbon-like deposits observed on the catalysts. In case of sufficient hydrogen the catalyst could be regenerated because the hydrogen was activated by the platinum and palladium which removed the surface sulphides (Pd–S, Pt–S). At the same time up to 280–290 °C and at least 1.5 h^{-1} LHSV, at 30–40 bar pressure and 250–350 Nm^3/m^3 H_2/hydrocarbon ratio, using the same operation parameters for more than 200 h the activity of the catalyst – apart from a small degree of deactivation in the first few hours – was constant within the general margin of error of the technological experiment and the analytical methods. For this and regarding the importance of the economical requirements furthermore to avoid the copiousness hereunder only the results obtained at 30 bar total pressure, 250 Nm^3/m^3 H_2/HC ratio, $\leq$1.5 h^{-1} LSHV and up to 280 °C will be presented.

HDS activities of the different catalysts are compared in Fig. 1 as a function of the reaction temperature at different LSHV (sulphur content of feed 103 mg/kg). The remaining sulphur contents of the products show that all the catalysts are able to convert the majority of sulphur in the feeds to H_2S resulting in a product of which sulphur content is below 10 mg/kg at the favourable operation parameters. Fig. 1 presents also clearly that the same HDS efficiency can be attained over PtPd/USY catalyst at lower temperature as on the other catalysts, using the same other parameters. As a function of the temperature the sulphur content of the products changed the most sharply in case of PtPd/USY catalyst till the saturation period of the curves.

The highest degree of HDS was about 92–98% for all catalysts, representing a product sulphur level of about 5–9 mg/kg in case of 1.5–2.0 h^{-1} liquid space hourly velocity. Regarding the degree of desulphurisation (the residual sulphur content of the product) and the necessary temperature to reach it the results obtained at $\leq$2.0 h^{-1} LSHV will be discussed in detail.

In case of HDN efficiency of the catalyst great differences were observed (Fig. 2). PtPd/USY zeolite exhibited excellent denitrogenation ability since the nitrogen content of all products was by far lower than 1 mg/kg. Both CoMo catalysts showed only moderate HDN activity, not exceeding 64% removal. The results indicate that the HDN could be enhanced in a small degree under more severe process conditions. Therefore, a major advantage of the PtPd/USY catalyst is the complete removal of the nitrogen compounds from

Table 2
Comparison of the performance of the PtPd/USY and the CoMo/Al_2O_3 catalysts in case of different sulphur containing feedstocks (P = 30 bar; H_2/HC = 300 Nm^3/m^3).

Properties	Product properties and their production conditions (product yield ≥ 99%)				
Sulphur/nitrogen content of the feedstock (mg/kg)	29/14	103/33	152/33	204/33	306/33
PtPd/USY					
Yield (%)	>99.8	99.4–99.6	99.2–99.5	99.1–99.3	97.4–98.7
Sulphur content (mg/kg)	8–6	5–7	5–8	6–9	10–12
Nitrogen content (mg/kg)	<1	<1	<1	<1	<1
ΔRON	−1.8 to −1.9	−1.9 to −2.1	−2.0 to −2.4	−2.7 to −2.9	−4.7 to −5.8
Δisoparaffin (%)	0.3	0.5	0.5	1.3	1.2
Temperature (°C)	210–220	250–260	250–260	260–270	290–300
LHSV (h^{-1})	2.0	2.0	1.5–2.0	1.5–2.0	2.0
Conventional CoMo/Al_2O_3					
Yield (%)	>99.8	99.4–99.6	99.3–99.5	98.7–98.9	98.1–98.4
Sulphur content (mg/kg)	6–9	7–12	6–10	7–10	5–9
Nitrogen content (mg/kg)	18–24	15–17	14–17	13–16	12–14
ΔRON	−4.1 to −4.4	−4.4 to −4.8	−4.6 to −5.1	−4.9 to −5.4	−5.1 to −5.8
Δisoparaffin (%)	0.1	0.2	0.4	0.5	0.7
Temperature (°C)	260–280	270–280	270–280	280–300	290–310
LHSV (h^{-1})	2.0	2.0	1.5–2.0	1.5–2.0	2.0
New generation CoMo/Al_2O_3					
Yield (%)	>99.8	99.5	99.2–99.5	98.9–99.1	98.6–99.0
Sulphur content (mg/kg)	6.5–9	5–10	9–10	7–11	10–12
Nitrogen content (mg/kg)	14–17	16–18	14–16	12–15	10–13
ΔRON	−2.3 to −2.9	−2.1 to −2.3	−2.2 to −2.8	−2.8 to −3.0	−3.4 to −4.7
Δisoparaffin (%)	0.2	0.3	0.4	0.4	0.6
Temperature (°C)	265–275	260–270	260–270	270–280	280–290
LHSV (h^{-1})	2.0–3.0	2.0	1.5–2.0	1.5–2.0	2.0

ΔRON: $RON_{product} - RON_{feed}$; Δisoparaffin, %: isoparaffin $content_{product}$ − isoparaffin $content_{feed}$.

the feed. The higher HDN activity might be attributed to the much higher acidity of the USY zeolite support than that of Al_2O_3 and thus to more significant adsorption of basic nitrogen compounds on the catalyst resulting in increased surface concentration near to metallic sites. (It is probable that the relatively low concentration of nitrogen compounds does not cause irreversible blocking.) This is important, because the nitrogen content is quantitatively converted to NO_x in the combustion chamber, which increases engine emissions. The importance of the deep HDN of the FCC gasolines is that during the combustion of the organic compounds containing bonds of carbon to nitrogen approximately 40,000 tons of NOx generates yearly, supposing 30 mg/kg average nitrogen content of the FCC gasoline used around the world. It is about 1% of the NOx generated during the combustion of the gasolines.

The degrees of olefin saturation on different catalysts are plotted in Fig. 3 as functions of the reaction temperature. At low temperature the PtPd/USY zeolite shows the lowest olefin saturation activity, but at higher temperature it was higher than over the new generation CoMo/Al_2O_3 catalyst. The sequence of olefin conversion on the three different catalysts for the feed of 103 mg/kg sulphur content is the following: conventional hydrotreating catalyst ≫ new generation CoMo/Al_2O_3 > PtPd/USY up to about 250–270 °C (depending on the LSHV), above this temperature the new generation CoMo/Al_2O_3 catalyst had the lowest olefin saturating activity.

Regarding simultaneously the results of sulphur removing and olefin saturation as a function of the operation parameters it was concluded that the best results were obtained at about 260 °C and 2.0 h^{-1} LSHV in case of both the new generation CoMo/Al_2O_3 and the PtPd/USY catalyst. The latter one seems to be more suitable because of the significantly higher hydrodesulphurization efficiency and the unequivocally higher nitrogen removal activity.

The olefin conversion was also plotted as a function of HDS conversion (Fig. 4). This also indicates that for this particular feed the PtPd/USY catalyst was competitive, moreover better, in terms of HDS and especially HDN selectivity (see Fig. 2) than the CoMo/alumina being industrially applied for selective HDS of FCC gasoline.

The favourable results of our experiments carried out for the confirmation of the suitability of the PtPd/USY catalyst for selective desulphurization of heavy FCC gasolines and other sulphur containing feedstocks are summarized in Table 2. Data show that the investigated PtPd/USY catalyst is suitable to produce lower than 10 mg/kg sulphur and lower than 1 mg/kg nitrogen containing products from feedstock having up to about 200 mg/kg sulphur content, while the octane number loss is only about 1.8–3.0. In case of selective desulphurization of feedstocks having higher than about 200 mg/kg sulphur content the use of the new generation CoMo/Al_2O_3 catalyst is more favourable. Similar results were obtained by Wang and Inglesia [13] and Dos Santos et al. [14] who demonstrated the inhibition of H_2S on both HDS and HYD reactions during the hydrodesulphurisation of thiophene and 3-methylthiophene, respectively. However its nitrogen removing activity is low. It seems that the sulphur resistance of the PtPd/USY catalyst was not high enough in spite of the electron withdrawing effect of the support toward platinum.

Table 3
The main product properties after the heteroatom removal of the mixture of FCC and pyrolysis gasolines.

Properties	Feedstock	Product		
	Conventional CoMo/Al_2O_3	New generation CoMo/Al_2O_3	PtPd/USY	
Reactor temperature (°C)	–	280–290	225–260	215–260
LSHV (h^{-1})	–	1.0	1.0–2.0	1.0–2.0
Cracking selectivity (%)	–	1.4–1.9	1.0–1.7	1.2–1.8
Sulphur content (mg/kg)	133	8–9	6–9	7–9
Nitrogen content (mg/kg)	32	24–26	21–23	<1
ΔRON	–	−4.8 to −5.3	−1.9 to −2.3	−1.7 to −1.9

The high, selective desulphurization and denitrogenation activity of the PtPd/USY catalyst in case of lower than 200 mg/kg sulphur containing feedstocks is verified by the results of the mixture of heavy FCC and pyrolysis naphtha (see second feedstock of the Table 1) hydrogenation experiments. These were obtained in the last period of the 240-h-long experiments (Table 3).

For investigation of the suitability of nitrogen removing activity of the PtPd/USY catalyst in case of at least about 200 mg/kg sulphur and high olefin containing feedstocks – among others – a mixture of FCC heavy gasoline and coker light naphtha (see third feedstock in Table 1) was used. In these experiments new generation $CoMo/Al_2O_3$ and the PtPd/USY catalysts were investigated in one catalyst bed after each other using mass ratios from 5:1 to 9:1. The results showed when the two catalyst are inserted in two layers: first new generation $CoMo/Al_2O_3$ thereafter downstream PtPd/USY in a mass ratio of 6.5:1 and the favourable operation parameters (T: 265 °C, P: 30 bar; LSHV: 1.5 h^{-1}; H_2/HC: 250 Nm^3/m^3) are established 8 mg/kg sulphur and 1.5 mg/kg nitrogen containing products are produced, whose octane number was lower by 1.6 unit only than that of the feedstock. The reason of this is that H_2S generated on the new generation $CoMo/Al_2O_3$ catalyst decreases the olefin saturation activity of the PtPd/USY (used as the lower catalyst layer) by the partial poisoning of the active sites, but it practically does not decrease the nitrogen removing activity.

4. Conclusions

During the research work the selective desulphurization and denitrogenation of olefin rich hydrocarbon mixtures (FCC-, pyrolysis-, and coker naphthas; total sulphur content up to about 300 mg/kg, nitrogen content up to about 35 mg/kg) over three different catalysts (conventional and new generation $CoMo/Al_2O_3$, and Pt(0.1%)Pd(0.3%)/USY) were studied. It was concluded that products of lower than 10 mg/kg sulphur content could be obtained at the favourable operation parameters, which depended on the heteroatom content of the feedstock and the applied catalyst. The degree of nitrogen removal was nearly 100% over the PtPd/USY catalyst, while on the other two catalysts it was at best 50–60%. The octane number loss after the hydrotreatment in case of up to about 200 mg/kg sulphur containing feedstock was the lowest over the PtPd/USY catalyst (1.8–2.9 unit). The octane number loss determined by the substantially lower octane number of the paraffins was generated by the hydrogenation of olefins, and by the higher octane number of the isoparaffins formed by the isomerisation of normal paraffins. In case of feedstocks containing more than about 200 mg/kg sulphur the sulphur resistance of the PtPd/USY was lower than that of the new generation $CoMo/Al_2O_3$ catalyst, consequently higher temperature should be used, which caused the decrease of the octane number of the product. It was concluded that in case of higher sulphur (330 mg/kg) and nitrogen (37 mg/kg) containing feedstock (mixture of FCC and coker naphthas) the application of the new generation $CoMo/Al_2O_3$ and the PtPd/USY catalyst in one reactor after each other enables the production of very low sulphur (<5 mg/kg) and nitrogen (<2 mg/kg) containing products with low octane number loss (<1.6). The introduced evaluation based on long-term experiments reflects well the competitiveness of the industrial applicability of the PtPd/USY catalyst for selective sulphur and nitrogen removal.

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