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RESEARCH ARTICLE

Hydrodynamic activation of heavy oil residues

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Abstract

Objectives. Recently, there has been a tendency to increase the volume of high-viscosity heavy oils in the total volume of oil produced. The processing of these oils requires new technological approaches. This task is closely related to the need to increase the depth of oil refining. Among the approaches proposed to solve these problems, mechanochemical activation, which is based on the cavitation effect produced by ultrasonic or hydrodynamic methods, has been suggested. This study evaluated the effects of cavitation in increasing the depth of oil refining.

Methods. Straight-run and “secondary” oil products were used as raw materials: vacuum gas oil, catalytic cracking gas oil, and fuel oil. Activation was carried out in a high-pressure disintegrator. The principle of operation was to compress the oil product and then pass it through a diffuser. When the oil was passed through the diffuser, there was a sharp pressure release to atmospheric pressure, which caused cavitation in the hydrodynamic flow. The pressure gradient on the diffuser and the number of processing cycles ranged from 20 to 50 MPa and 1 to 10, respectively. The density, refractive index, and the fractional composition of petroleum products were determined using standard and generally accepted methods.

Results. This paper reports the influence of mechanochemical activation of petroleum products on their physical and chemical characteristics. An increase in the pressure gradient and the number of processing cycles leads to a decrease in the boiling point of the petroleum products and their density and an increase in the yield of fractions that boil off below 400°C. The yield of the fractions with boiling points of 400–480°C and the remainder were reduced. The density and refractive index of fractions with boiling points up to 480°C decreased, and the density of the residue increased. The effects of cavitation (an increase in the yield of fractions with boiling points up to 400°C and a decrease in the density of the petroleum products) increased with increasing pressure gradient and the number of processing cycles.

Conclusions. The changes in the density, boiling point, and the yield of fractions increased with increasing the pressure from 20 to 50 MPa and the number of hydrodynamic cavitation cycles from 1 to 5. Increasing the number of processing cycles to more than five had little additional effect. The effects of cavitation increased with increasing initial density of the oil product. The average molecular weight of these fractions was estimated from the densities and boiling points of individual fractions of the petroleum products. The calculation confirmed the assumption regarding the course of cracking reactions of petroleum products under the influence of cavitation and indicates the course of the compaction processes.

Keywords: cavitation, oil, yield, light fractions, oil fractions

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НАУЧНАЯ СТАТЬЯ

Гидродинамическая активация тяжелых нефтяных остатков

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Аннотация

Цели. В последние годы прослеживается тенденция увеличения в общем объеме добываемой нефти высоковязких, тяжелых нефтей, переработка которых требует новых технологических подходов. Эта задача тесно связана с необходимостью повышения глубины переработки нефти. Среди подходов, предлагаемых для решения отмеченных задач, встречается метод механохимической активации, который основан на использовании эффекта кавитации, создаваемого ультразвуковым или гидродинамическим способами. Цель работы заключалась в исследовании возможности использования эффекта кавитации для повышения глубины переработки нефти.

Методы. В качестве сырья использовались прямогонные и «вторичные» нефтепродукты: вакуумный газойль, газойль каталитического крекинга, мазуты. Активация проводилась в дезинтеграторе высокого давления, принцип действия которого заключался в сжатии нефтепродукта с последующим его пропуском через диффузор. При этом происходил резкий «сброс» давления до атмосферного, и в гидродинамическом потоке возникало явление кавитации. Градиент давлений на диффузоре варьировался от 20 до 50 МПа, а количество циклов обработки от 1 до 10. Определение плотности, коэффициента рефракции и фракционного состава нефтепродуктов осуществлялось с использованием стандартных и общепринятых методов.

Результаты. В работе представлены результаты влияния механохимической активации нефтепродуктов на изменение их физико-химических характеристик. Показано, что повышение градиента давлений и числа циклов обработки приводит к снижению температур начала кипения нефтепродуктов, их плотности и увеличению выхода фракций, выкипающих до 400 °С. Выход фракции с температурами кипения 400–480 °С и остатка при этом снижается. Отмечено снижение плотности и показателя преломления фракций с температурами кипения до 480 °С и повышение плотности остатка. Установлено, что эффект от явления кавитации (увеличение выхода фракций с температурами кипения до 400 °С, снижение плотности нефтепродукта) возрастал при увеличении градиента давлений и количества циклов обработки.

Выводы. Показано, что увеличение давления от 20 до 50 МПа и количества циклов гидродинамической кавитации способствует большему изменению плотности, температуры начала кипения и выхода фракций. Установлено, что повышение числа циклов обработки свыше 5 нецелесообразно. Отмечено, что с увеличением исходной плотности нефтепродукта эффективность воздействия возрастает. По плотностям и температурам кипения отдельных фракций нефтепродуктов оценена средняя молекулярная масса этих фракций. Расчет подтвердил предположение о протекании реакций крекинга нефтепродуктов под воздействием кавитации и свидетельствует о протекании процессов уплотнения.

Ключевые слова: кавитация, нефть, выход, светлые фракции, масляные фракции

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INTRODUCTION

Efficient processing of heavy oil residues is an urgent problem of the petrochemical industry. The depletion of reserves of traditional light and medium oils, which were a source of raw materials for the production of motor fuels and the petrochemical industry, has highlighted the need to increase the depth of oil refining. Heavy high-viscosity oils are being increasingly involved in the fuel and energy balance, but their processing requires new technological approaches. The resources of heavy and bituminous oils significantly exceed the reserves of light oils and are estimated to be 750 billion tons. Canada (386 billion tons) and Venezuela (335 billion tons) have the largest reserves of heavy oils. The reserves of heavy oils in Russia (the fields of the Volga–Ural, Timan–Pechora, and West Siberian oil-and-gas provinces) are estimated at 30–75 billion tons. Heavy, high-viscosity oils are characterized by a high content of polyaromatic hydrocarbons and asphaltene–resinous substances. In these indicators, they are close to heavy oil residues. This potentially allows a uniform approach to their processing [1–3].

The coking process is the most common method for processing heavy oil residues and accounts for up to 40% of residues. The following processes have been used in the order of the processing volumes: visbreaking (~34%), hydrotreating (~18%),

hydrocracking (~4%), and deasphalting (~3.5%) [4]. The same processes are also considered methods of processing heavy oils, and research has been carried out in many scientific centers [4–7].

Simultaneously with the traditional methods of processing heavy oils and oil products, an active search for new technological approaches to their processing is underway [4, 8–10]. Mechanochemical activation is often considered a new unconventional method of treating heavy oil feedstock. This suggests the generation of cavitation in an oil flow. Cavitation is a physical process of nucleation (bubble formation) in a liquid media followed by bubble collapse [11–15]. Researchers have noted local increases in temperature of up to 10000 K caused by collapsing cavitation bubbles [16, 17]. As a result, cracking reactions become possible, which affects the fractional composition of oil and petrochemicals. On the other hand, information on the changes in the physicochemical characteristics of oil is contradictory. Some researchers noted a decrease in the density and viscosity of the resulting oil products, while others suggested an increase in the content of heavy oil fractions. A possible reason for these contradictions is the difference in characteristics and processing conditions [11–15]. This study examined the influence of mechanochemical activation on the physicochemical characteristics of petroleum products.

Table 1. Physicochemical characteristics of the samples

Indicator	Sample			
	CCG	M1	M2	VG
Density, g/cm ³	1.1002	0.9684	0.9478	0.8998
Yield of fractions boiling in the temperature range, wt %				
From the initial boiling point (T_{IBP}) to 350°C	5.2	5.0	13.2	8.4
350–400°C	25.8	9.0	15.8	34.5
400–480°C	69.0	28.0	47.0	40.9
Above 480°C		58.0	24.0	16.2

Note: CCG – catalytic cracking gas oil, VG – vacuum gas oil.

MATERIALS AND METHODS

Straight-run and “secondary” petroleum products were used as raw materials: vacuum gas oil (VG), catalytic cracking gas oil (CCG), and masut (fuel oil, M1, and M2). Table 1 lists their characteristics.

The activation of petroleum product samples was carried out in a high-pressure disintegrator described elsewhere [18]. The principle of action is that an oil sample is compressed to a pressure of 20–50 MPa and passed through a diffuser. When passing through the diffuser, there is a sharp decrease in pressure to atmospheric pressure, and cavitation occurs in hydrodynamic flow. The passage of a minimum amount of a sample (0.5 L) through the diffuser from the high-pressure zone to the atmospheric zone is considered a single cycle. The number of cycles of such treatment of the raw material ranged from 1 to 10. The temperature at which the samples were activated was chosen because gas bubbles in the liquid volume are the nuclei of cavitation, according to previous reports [19, 20]. Accordingly, the cavitation intensity increases with increasing number of nuclei in the volume of oil products. Because an increase in temperature leads to the degassing of samples, activation was carried out at a minimum temperature that ensures the pumpability of the oil product through the apparatus. Thus, for the VG, HCC, and M1 and M2 samples, pumpability was achieved at 30°C, 50°C, and 70°C, respectively.

The sample density was determined using the pycnometric method. The initial boiling point, the yield of fractions of the vacuum distillation of petroleum products, and the refractive index were examined by refractometry.

For fuel oil and VG oil samples, the yield of fractions boiling out over the temperature ranges, T_{IBP} –350°C, 350–400°C, 400–480°C, and > 480°C, was determined. For CCG oil, the yield was determined for fractions boiling up to 350°C and higher. The fractions were combined (350°C and higher) for CCG oil because an increase in temperature above 350°C (calculated for atmospheric pressure) during the analysis led to hydrocarbon decomposition.

RESULTS AND DISCUSSION

For all samples investigated, the boiling-onset temperatures and density decreased after treatment. For example, the boiling point of sample M2 was 280°C. After five treatment cycles at a pressure gradient of 50 MPa, the boiling-onset temperature decreased to 250°C. More severe the activation conditions (higher pressure gradient, more exposure cycles) resulted in a lower density of the product obtained (Fig. 1). Simultaneously, the characteristics of the samples did not change after five processing cycles.

The significant change in density was attributed to a change in the yield of the fractions (Fig. 2). As a result of activation, the yield of fractions boiling up to 400°C increased, whereas the yield of the heavy oil fraction and residue decreased. The increase in the total yield of light and medium oil fractions depended on the treatment pressure gradient. Hence, the total yield of these fractions in the VG sample was 43 wt %. After five treatment cycles at 20 and 50 MPa, the yield was 46.5 and 52.6 wt %, respectively, showing an increase with increasing pressure.

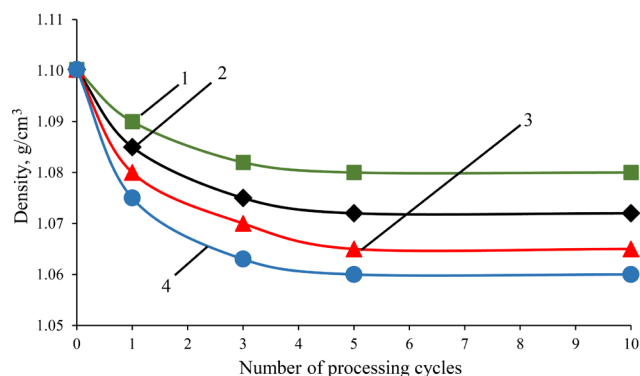


Fig. 1. Influence of the activation conditions on the CCG sample density. Pressure gradient, MPa: (1) 20; (2) 30; (3) 40; (4) 50.

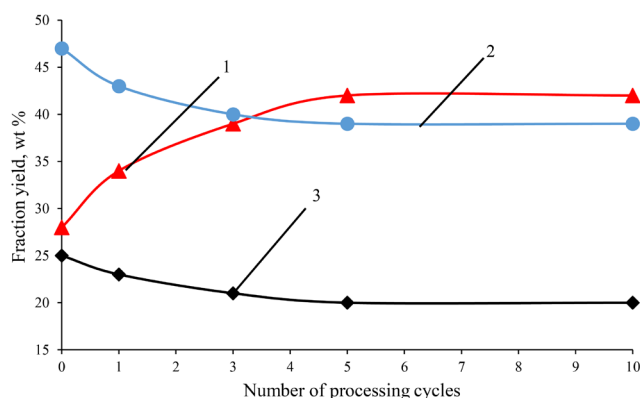


Fig. 2. Influence of the number of activation cycles on the M2 sample fractional composition. Pressure gradient 50 MPa. Fractions: (1) T_{IBP} –400°C; (2) 400–480°C; (3) residue.

After sample activation, both the fractional composition and the physicochemical characteristics of the individual fractions changed (density and refractive index). For all samples studied, an increase in the pressure gradient and the number of processing cycles led to a decrease in the refractive index and the density of fractions boiling up to 480°C. Simultaneously, the density of the fraction with a boiling point above 480°C increased (Figs. 3 and 4).

The changes in the density and refractive index for all oil products under consideration were identical. Increasing the number of activation cycles of samples to more than five had little additional effect on the sample characteristics.

The relationship between the original sample density and the changes that occurred as a result of its processing was noted: the higher its density, the more significant the changes. For example, the yield

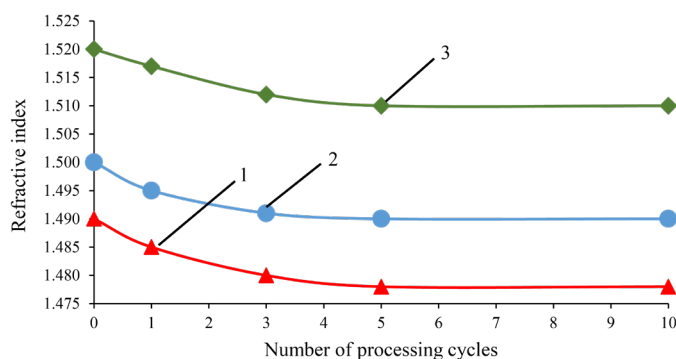


Fig. 3. Influence of the number of activation cycles on the refractive index of the M2 sample fractions. Pressure gradient was 40 MPa. The boiling temperature of the fractions, °C: (1) T_{IBP} –350; (2) 350–400; (3) 400–480.

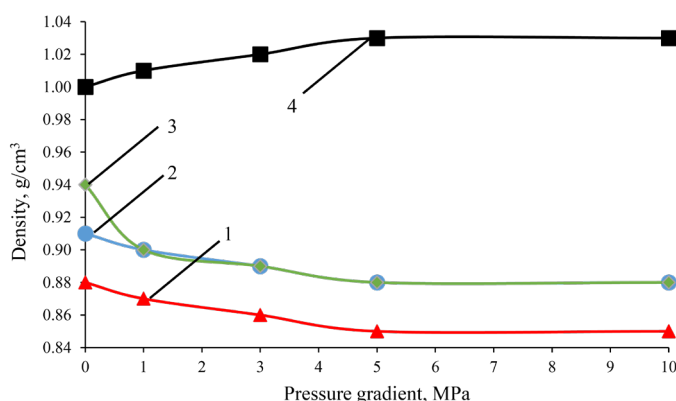


Fig. 4. Influence of the pressure gradient on the M1 sample fractions density; five cycles of activation. The boiling temperature of the fractions, °C: (1) T_{IBP} –350; (2) 350–400; (3) 400–480; (4) residue.

of fractions with a boiling point of up to 400°C for the VG sample (initial density 0.8998 g/cm³) was 42.9 wt %. After five-time treatment at 50 MPa, it increased to 52.6 wt %, i.e., it increased by 9.7%. For the M1 sample (density 0.9684 g/cm³), the yield of this fraction under the same processing conditions increased by 14.8%, and for the GCC sample (density 1.1002 g/cm³), by 24.5 wt %.

The authors of [21] have revealed the formation of unsaturated hydrocarbons when treating alkanes, a decrease in the samples boiling points studied by us, and a change in the physicochemical characteristics of their individual fractions. We can explain this by an increase in temperature as a result of the gas

Table 2. Influence of the activation conditions on the average molecular weight of the fraction boiling in the $T_{\text{IBP}}-350^{\circ}\text{C}$ temperature range

Mechanochemical activation conditions		Average molecular weights of samples			
Number of processing cycles	Processing pressure, MPa	VG	M1	M2	CCG
Original sample		219	239	236	200
1	20	218	238	235	197
	50	213	230	229	192
5	20	215	232	230	194
	50	210	225	224	189

bubbles collapse upon cavitation. We assumed that a local increase in temperature resulted in the formation of alkenes and hydrocarbons with a lower molecular weight as a result of cracking reactions. These hydrocarbons are characterized by lower densities and refractive indexes.

Table 2 shows the influence of the activation conditions on the average molecular weight of the fraction boiling out in the $T_{\text{IBP}}-350^{\circ}\text{C}$ temperature range for the samples studied.

The average molecular weight was estimated using the formula recommended for narrow oil fractions [19]:

$$M = (160 - 5K) - 0.075T_{\text{av}} + 0.000156KT_{\text{av}}^2,$$

where T_{av} is the fraction average boiling point, and K is the characteristic factor calculated based on the fraction density and average boiling point [22].

This formula was chosen because the difference between the calculated and experimental values (determined by the cryoscopic method in a Cryon-1 installation, benzene solvent) did not exceed 10%. For example, for the fraction with boiling points of $400-480^{\circ}\text{C}$ (sample M1, 5 treatment cycles at a pressure of 50 MPa), the experimental and calculated average molecular weights were 350 and 388 units, respectively.

For all the samples studied, the average molecular weight of the fraction boiling at $T_{\text{IBP}}-350^{\circ}\text{C}$ decreased. The molecular weight of fractions with boiling points of $350-400^{\circ}\text{C}$ remained relatively unchanged.

In the case of fractions with high boiling points, the molecular weight increased. For fractions with boiling points above 480°C (residue), the result was consistent with an increase in density.

CONCLUSIONS

The results indicated the effectiveness of a mechanochemical treatment to increase the depth of oil refining (to increase the yield of light and oil fractions). This method has the potential for processing high-viscosity, highly resinous oils. These results also showed that along with cracking reactions, compaction processes also occur, which confirms the data on the occurrence of hydrocarbon cracking reactions under the influence of cavitation reported elsewhere.

Authors' contribution

V.B. Terenteva – conducting the experimental research, determining physicochemical parameters of objects studied and products obtained;

B.V. Peshnev – setting objectives, planning the experimental studies, processing the data obtained, preparation of the data obtained for publication;

A.I. Nikolaev – setting objectives, planning and conducting the experimental research, processing the data obtained, preparation of the data obtained for publication.

The authors declare no conflicts of interest.

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