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RESEARCH ARTICLE

Changes in the hydrocarbon composition of petroleum products under the influence of cavitation

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Abstract

Objectives. While the phenomenon of cavitation is of interest for treatment of hydrocarbon mixtures, in particular crude oil and petroleum products, the literature lacks a systematic approach to conducting such research. This gap stimulates the need for a more in-depth study of the influence of this physical effect on the characteristics and detailed hydrocarbon composition of petroleum feedstock. Thus, the present work set out to explore the influence of the conditions leading to cavitation on the physicochemical properties and hydrocarbon composition of crude oil and petroleum products.

Methods. The objects of the study were two crude oil samples and four straight-run fractions—gasoline, kerosene, diesel, and fuel oil—having different characteristics and hydrocarbon compositions. Cavitation treatment was carried out in a hydrodynamic mode using a Donor-2 apparatus within a range of pressure changes from 20 to 50 MPa. The number of treatment cycles was from 1 to 20. The density was determined by pycnometry using the refractive index, an Abbe refractometer, and the fractional composition or fraction yield, as well as by distillation at atmospheric or reduced pressure for light or dark petroleum products, respectively. The hydrocarbon composition of the gasoline fraction was determined by chromatography and mass spectrometry.

Results. Changes in the densities and fractional compositions of the objects of study following their treatment under various conditions were recorded. Particular attention was paid to the hydrocarbon composition of the gasoline fraction: an increase in the content of normal alkanes was shown to be due to an increase in the number of structures with shorter carbon chains in comparison with the components of raw materials not subjected to cavitation.

Conclusions. The results of the study of the effect of cavitation treatment of crude oil and its individual fractions on their physicochemical characteristics showed that the nature of the changes depends on the treatment conditions and the initial characteristics of the sample. It is suggested that cavitation treatment causes cracking and compaction processes. The possibility of cracking reactions was confirmed by chromatographic determination of the group hydrocarbon composition of the samples.

Keywords

crude oil, petroleum products, cavitation, hydrocarbon composition, classes of hydrocarbons

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НАУЧНАЯ СТАТЬЯ

Изменение углеводородного состава нефтепродуктов под воздействием кавитации

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Аннотация

Цели. Интерес к применению явления кавитации для обработки углеводородных смесей, в частности нефти и нефтепродуктов, и отсутствие в литературе системного подхода к проведению подобных исследований открывает необходимость более глубокого изучения влияния этого физического воздействия на характеристики и детальный углеводородный состав нефтяного сырья. В связи с этим, цель данной работы заключается в изучении влияния условий, при которых возникает кавитация, на изменение физико-химических свойств и углеводородного состава нефти и нефтепродуктов.

Методы. В качестве объектов исследования были взяты два образца нефти и четыре прямогонные фракции — бензиновая, керосиновая, дизельная и мазут, обладающие различными характеристиками и углеводородным составом. Кавитационную обработку проводили в гидродинамическом режиме на аппарате Донор-2 в диапазоне перепадов давления от 20 до 50 МПа. Количество циклов обработки составляло от 1 до 20. Плотность определяли пикнометрическим методом, показатель преломления — на рефрактометре типа Аббе, а фракционный состав или выход фракций — перегонкой при атмосферном или пониженном давлении соответственно для светлых или темных нефтепродуктов. Углеводородный состав бензиновой фракции определяли хромато-масс-спектрометрическим методом.

Результаты. В работе показано изменение плотности и фракционного состава объектов исследования после их обработки при различных условиях. Уделено особое внимание углеводородному составу бензиновой фракции: показано увеличение содержания в ней нормальных алканов за счет увеличения количества структур, обладающих меньшей длиной углеродной цепи, по сравнению с компонентами сырья, не подвергнутого кавитационному воздействию.

Выводы. Представлены результаты исследования влияния кавитационной обработки нефти и ее отдельных фракций на изменение физико-химических характеристик объектов исследования. Установлено, что характер изменений зависит от условий обработки и исходных характеристик образца. Высказано предположение о том, что в результате кавитационного воздействия могут протекать процессы крекинга и уплотнения. Возможность протекания реакций крекинга подтверждена результатами хроматографии и определения группового углеводородного состава образцов.

Ключевые слова

нефть, нефтепродукты, кавитация, углеводородный состав, классы углеводородов

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INTRODUCTION

The alternation of high- and low-pressure regions in a liquid medium leads to cavitation involving the formation and collapse of gas bubbles [1, 2]. The study of this phenomenon confirms its role not only as a destructive factor for ship propellers and hydroelectric turbine blades, but also as an efficient agent in processes taking place agriculture [3], medicine [4], the food industry [5], construction [6], and the oil industry [7–9].

In the oil production and refining industries, cavitation is used as a means of modifying the rheological properties of crude oil and its fractions to reduce the viscosity of petroleum disperse systems and consequent cost of transportation [10], as well as increasing the yield of light fractions [11] and improving the quality of hydrocarbon fuels [12]. The potential for using cavitation to intensify chemical engineering processes has also been noted [13]. However, despite the wide variety of publications on the use of cavitation in oil refining processes, they lack a systematic approach for relating the conditions leading to cavitation phenomena with the observed changes in feedstock characteristics, particularly in terms of their hydrocarbon composition.

The present study set out to explore the influence of the conditions for the occurrence of cavitation on

the characteristics and hydrocarbon compositions of petroleum and petroleum products.

EXPERIMENTAL

The objects of study were samples of heavy (O1) and light (O2) crude oils obtained from the Ilskoe field (Krasnodar krai, Russia); straight-run fractions of crude oil: gasoline (G), kerosene (K), and diesel (D); and fuel oil (F) (all of *Gazpromneft-MNPZ*, Russia). Table 1 presents some of their characteristics.

Cavitation treatment was performed in a Donor-2 apparatus (Russia) [14, 15]. In this apparatus, cavitation occurs in the hydrodynamic flow of a petroleum product through a diffuser under elevated pressure. The pressure in the discharge cylinder was varied from 20 to 50 MPa; the number of treatment cycles, from 1 to 20. The treatment temperature was determined by the viscosity of the samples. Samples G, K, and D were treated at 20°C; Samples O1 and O2, at 50°C; Sample F, at 70°C.

The density of the samples was measured pycnometrically in accordance with GOST R 50.2.075-2010[1].

The fractional composition of light petroleum products was characterized using an ARNP-1 apparatus (*NPP SKIF-PRIBOR*, Ukraine) in accordance with GOST 2177-99[2]. The yields of dark fractions of crude oil and fuel oil were determined by vacuum distillation at a residual pressure of ~2 mm Hg [16].

The hydrocarbon composition of the gasoline fraction was determined using an Agilent GC 7890 chromatograph (*Agilent Technologies*, USA) on an HP-5MS quartz capillary column (30 m × 0.25 mm × 0.25 μm) with a modified methyl silicone (5% phenylmethyl silicone) liquid stationary phase. The chromatograph was equipped with an MSD 5975C mass-selective detector (*Agilent Technologies*, USA) having a quadrupole mass analyzer. Mass spectra were obtained by electron impact ionization with an ionization energy of 70 eV and recorded in the mass range of 29–550 amu at an evaporator temperature of 320°C and a carrier gas (helium) flow rate of 1 mL/min.

RESULTS AND DISCUSSION

The most obvious changes in the characteristics of petroleum products under the influence of cavitation involve variations in the sample density (Fig. 1) and its fractional composition. For example, the initial boiling points of Samples O1, F, and G following five treatment cycles at a pressure of 50 MPa decreased by 30, 25, and 8°C, respectively. It was observed that,

Table 1. Characteristics of objects of study

Property	Object of study					
	O1*	O2	G	K	D	F
Density, g/cm^3	0.9693	0.8810	0.7112	0.7880	0.8333	0.9684
Refractive index	–	–	1.4345	1.4501	1.4709	–
Initial boiling point, °C	160	56	40	144	175	290
Distillation point, °C 10 vol % 50 vol % 90 vol %	–	–	 71 116 186	 162 192 218	 213 280 335	–
Yield of fractions, wt % Up to 160°C 160–230°C 230–350°C 350–400°C 400–480°C Above 480°C	 – 0.5 5.2 93.4 93.4 93.4	 11.9 19.2 19.5 49.4 49.4 49.4	–	–	–	 – – 4.9 9.3 27.9 57.9

* O1, heavy crude oil; O2, light crude oil; G, gasoline; K, kerosene; D, diesel; and F, fuel oil.

[1] GOST R 50.2.075-2010. State system for ensuring the uniformity of measurements. Crude petroleum and petroleum products. Laboratory methods for determination of density, relative density and API gravity. Moscow: Standartinform; 2011.

[2] GOST 2177-99. Interstate Standard. Petroleum products. Methods for determination of distillation characteristics. Moscow: Standartinform; 2006.

the higher the initial density of the petroleum product, the more noticeable were the changes occurring in its physicochemical characteristics (density, fractional composition, etc.).

The changes occurring in the parameters of a sample with changing treatment conditions were noted to be significantly affected by the nature of the sample. Increasing the pressure and the number of treatment cycles (up to 10) for Sample F led to a decrease in the initial boiling point and density and an increase in the yields of fractions whose initial boiling points were up to 400°C. Conversely, the yields of fractions boiling above 400°C decreased. For example, after one treatment cycle at a pressure of 20 MPa, the initial boiling point of Sample F decreased from 290 to 287°C, while the yield of fractions boiling up to 400°C increased from 14.4 to 14.8 wt %. Following 10 processing cycles, these parameters amounted to 273°C and 23.4 wt %, respectively. If the treatment was carried out at 50 MPa, then after 10 cycles, the initial boiling point decreased to 257°C, and the yield of fractions boiling up at 400°C increased to 29.2 wt %. No significant changes were observed with a further increase in the number of treatment cycles.

When treating light fractions (G, K, D), their initial boiling points also decreased with increasing pressure and number of treatment cycles. The dependence of the density of these samples on treatment pressure was similar to that for Sample F: increasing pressure led to a greater decrease in density. However, the dependence of the density of the light products on the number of treatment cycles differed significantly. Unlike Sample F, the density of the light petroleum products began to increase with repeated treatments (Fig. 2).

The number of treatment cycles after which an increase in density was detected depended on the nature of the sample (its initial density) and the treatment pressure. For example, at a treatment pressure of 30 MPa, the density of Samples G and K increased after 10 cycles. For Sample D, no significant change in density occurred following 10 treatment cycles at this pressure. After 10 treatment cycles, its density was 0.8293 g/cm^3, while after 20 cycles, the density was 0.8295 g/cm^3. However, when the treatment pressure for Sample D was increased to 40 MPa, an increase in its density was detected already after 10 treatment cycles (after 5 cycles, the density was 0.8298 g/cm^3; after 10 cycles, 0.8294 g/cm^3; and

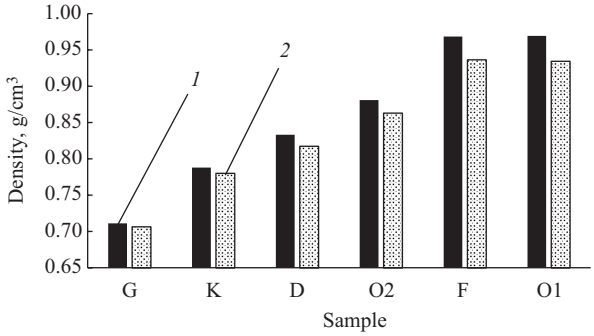

Fig. 1. Influence of cavitation treatment on the density of samples in five treatment cycles at 50 MPa: sample density (*1*) before and (*2*) after treatment

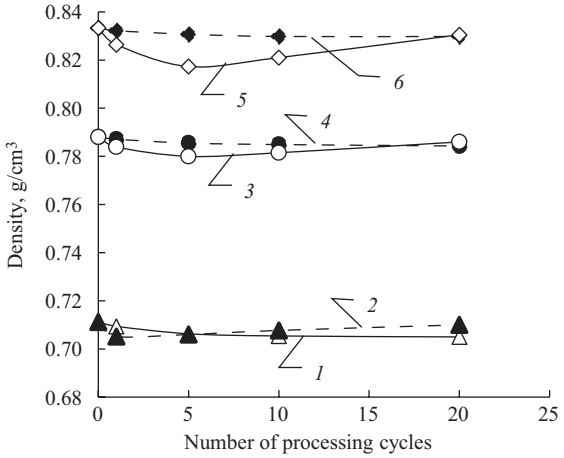

Fig. 2. Influence of conditions of cavitation treatment of light petroleum products on their density: Samples G (*1*, *2*), K (*3*, *4*), and D (*5*, *6*) at treatment pressures of 20 (*1*, *3*, *5*) and 50 MPa (*2*, *4*, *6*)

after 20 cycles, 0.8301 g/cm^3). At a treatment pressure of 50 MPa, an increase in the density of Sample D was noted after the fifth cycle.

Similar behavior with increasing number of treatment cycles was observed for the refractive indices of light petroleum products. For Samples G and K, increasing the number of treatment cycles to 10 (at a pressure of 30 MPa) resulted in a decrease in the refractive index to 1.4185 and 1.4408, respectively. After 20 treatment cycles, the refractive indices were 1.4205 and 1.4422, respectively. At a pressure of 50 MPa, the refractive index of Sample G after 5 processing cycles was 1.4205, and after 10 cycles, it increased to 1.4225.

Noteworthy is the effect of the number of treatment cycles on the fractional composition of light petroleum products and their distillation points. The initial boiling points of Samples G, K, and D decreased with both increasing pressure and increasing number of treatment cycles. When treating these samples at a pressure of 20 MPa, the 10, 20, …, and 95% distillation points of the samples also decreased with increasing number of cycles. If the treatment was carried out at higher pressures, then, after an initial decrease (after 1–5 cycles), subsequent treatment led to an increase in the 40%+ distillation point of the sample (Fig. 3).

For Sample D, an increase in distillation points was detected only at treatment pressures of 40 and 50 MPa; moreover, the higher the treatment pressure, the earlier this increase began. For example, if the treatment was carried out at 40 MPa, then, after 10 cycles, an increase in the distillation point was noted after distilling 60% of the sample. And if the treatment was performed at 50 MPa, then, after 10 cycles, the increase in the distillation point was observed after distilling 40% of the sample.

The effect of treatment conditions on the fractional composition of Samples G and K was similar. Moreover, an increase in the 40% distillation point was detected at a treatment pressure of 30 MPa.

The decreases in the densities, initial boiling points, and fractional compositions of the samples as a result of cavitation treatment can be explained by cracking reactions under the influence of heat released during the collapse of cavitation bubbles [17]. In the resulting lower-molecular-weight hydrocarbons, all of the above parameters are reduced. The interaction of long-lived radicals leads to the formation of products with a molecular weight higher than that of the feedstock. As manifested by an increase in the distillation points, this phenomenon is most noticeable after the treatment of light, low-boiling fractions. Although no such changes are observed following the treatment of dark petroleum products, an increase in density after cavitation treatment was noted for fractions boiling at temperatures above 480°C [14, 18].

Table 2 and Fig. 4 present data on the group hydrocarbon composition of Sample G and the number of carbon atoms in alkane molecules before and after treatment (5 cycles at a pressure of 50 MPa), confirming the occurrence of cracking reactions.

Table 2. Effect of treatment on the group hydrocarbon composition of Sample G after 5 treatment cycles at a pressure of 50 MPa

Hydrocarbons	Group hydrocarbon composition of Sample G, wt %	
	Before treatment	After treatment
Total alkanes	75.0	73.9
n-Alkanes	22.6	25.8
iso-Alkanes	20.0	18.0
Cycloalkanes	32.4	30.1
Arenes	22.5	22.6
Not identified	2.5	3.5

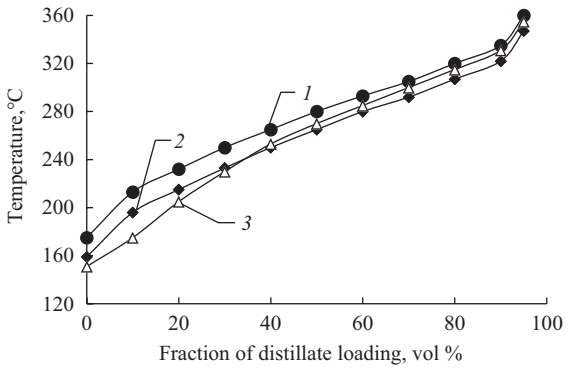

Fig. 3. Influence of the number of treatment cycles (pressure 50 MPa) on the fractional composition of Sample D: (*1*) initial sample and the sample after (*2*) 10 and (*3*) 20 treatment cycles at a pressure of 50 MPa

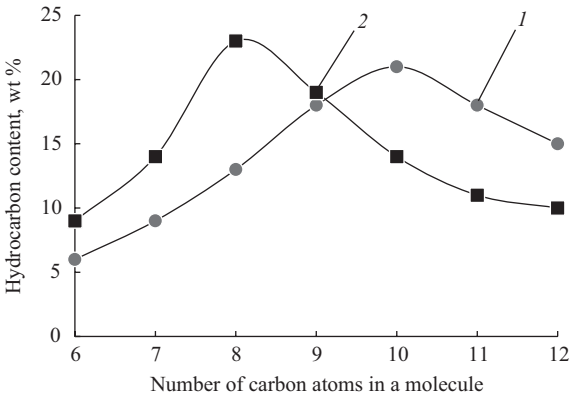

Fig. 4. Distributions of alkanes by number of carbon atoms in Sample G (*1*) before and (*2*) after treatment in 5 cycles at a pressure of 50 MPa

It is of particular significance that cavitation treatment was shown to alter the ratio of normal alkanes, isoalkanes, and naphthenes in the samples. While the fraction of *n*-alkanes increased, the contents of isoalkanes and naphthenes decreased. Following cavitation, the content of saturated hydrocarbons with 6–8 carbon atoms in a molecule increased, while the content of structures with 10–12 carbon atoms in a molecule decreased. No changes in the content of arenes were observed.

CONCLUSIONS

The results confirm the occurrence of cracking reactions during cavitation treatment of hydrocarbons. Depending on the treatment conditions and the initial characteristics of the sample, these reactions form products having lower and higher molecular weights than the feedstock. The results also suggest that arene hydrocarbons exhibit the greatest stability under cavitation conditions, while isoalkanes and naphthenes are more susceptible to degradation.

Authors' contributions

A.I. Nikolaev—development of the idea and concept of the study, formulation of the purpose and objectives of the study, identification of research objects, analysis of literary sources and obtained results, writing and editing the text of the article.

B.V. Peshnev—analysis of literary sources, writing and editing of the text of the article, development of methodology, analysis and presentation of the results obtained.

A.N. Korolev—conducting experiments, analysis and processing of the experimental data obtained, editing the text of the article.

D.V. Nikishin—consulting on methodology and research, providing equipment and consulting on the instrument base, editing the text of the article.

The authors declare no conflicts of interest.

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