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RESEARCH ARTICLE

Coking of high-viscosity water-containing oil

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Abstract

Objectives. *A characteristic feature of oil production is an increase in the volume of high-viscosity bituminous oil. In Russia, technologies based on the use of water vapor are used for their extraction. The use of such technologies leads to a large amount of water in the product stream from the production well. Preparation of oil for processing involves its stabilization, desalination, and dewatering. Since the densities of the extracted oil and the water contained in it are comparable, traditional preparation schemes for processing of high-viscosity bituminous oil are ineffective. One of the possible solutions to the problem involving such oil in the fuel, energy, and petrochemical balance is to use a coking process at the first stage of its processing. This aim can be achieved by studying the influence of the process conditions of coking high-viscosity water-containing oil on the yield and characteristics of the resulting products.*

Methods. *Coking of oil with a density of 1.0200 g/cm^3 at 50°C and with 18 wt% water content was carried out in a laboratory installation in a "cube." A hollow cylindrical apparatus was used as a reactor and was placed in a furnace. The temperature and pressure in the reactor were maintained at 500–700°C and 0.10–0.35 MPa, respectively.*

Results. *An increase in the coking process temperature results in an increase in the amount of gaseous products, a decrease in the amount of the coke generated, and a higher dependence of the amount of liquid products on temperature with a maximum yield at 550–600°C. The process temperature also affects the composition of liquid products. At a lower temperature, the amount of gasoline and kerosene fractions in liquid products is higher. With an increase in pressure, a higher amount of gaseous products, coke, and low-molecular-weight hydrocarbon fractions in liquid products could also be obtained. The characteristics of the coke produced in the coking process are similar to those of commercially produced grades. It is noted that when coking water-containing oil, up to 98% of the emulsion water goes with liquid products, and the remaining amount of water remains in the formed coke.*

Conclusions. *Results showed the possible application of the coking process at the initial stage of processing high-viscosity bituminous oil. In this case, the dewatering stage is significantly simplified since the technological scheme of delayed coking allows the separation of the gasoline fraction from water.*

Keywords: *coking, high-viscosity bituminous oil, watered oil, oil refining, oil refining method*

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НАУЧНАЯ СТАТЬЯ

Коксование высоковязкой обводненной нефти

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Аннотация

Цели. *Возрастание доли высоковязкой и битуминозной нефти является характерной особенностью современной нефтедобычи. В России при ее добыче применяют технологии, основанные на использовании водяного пара. Применение таких технологий приводит к тому, что продуктовый поток, выходящий из добывающей скважины, может содержать большое количество воды. Подготовка нефти к переработке предполагает ее стабилизацию, обессоливание и обезвоживание. Вследствие того, что плотность добываемой нефти и содержащейся в ней воды сопоставимы, традиционные схемы подготовки к переработке высоковязкой и битуминозной нефти являются малоэффективными. Одним из возможных решений проблемы вовлечения такой нефти в топливно-энергетический и нефтехимический баланс является использование на первом этапе ее переработки процесса коксования. Для реализации этой идеи необходимо изучить влияние условий коксования высоковязкой обводненной нефти на выход и характеристики образующихся продуктов.*

Методы. *Объектом исследования была нефть с плотностью при 50 °С 1.0200 г/см³, содержащая 18 мас. % воды. Коксование проводили на лабораторной установке в «кубе». В качестве реактора использовался пустотелый цилиндрический аппарат, размещаемый в печи. Температура в реакторе варьировалась от 500 до 700 °С, давление от 0.10 до 0.35 Мпа.*

Результаты. *Проведенные исследования показали, что при возрастании температуры коксования выход газообразных продуктов увеличивается, образование кокса уменьшается, а зависимость выхода жидких продуктов имеет экстремальный характер с максимумом в области температур 550–600 °С. Температура процесса влияет на состав жидких продуктов. При более низкой температуре в жидких продуктах выше содержание бензиновой и керосиновой фракций. Повышение давления приводит к возрастанию выхода газообразных продуктов, кокса и содержания в жидких продуктах низкомолекулярных фракций углеводородов. Образующийся в процессе кокс по своим характеристикам близок к промышленно выпускаемым маркам. Отмечено, что при коксовании обводненной нефти до 98% водной эмульсии уходит с жидкими продуктами коксования, и лишь небольшое количество воды остается в образовавшемся коксе.*

Выводы. *Представленные результаты показывают возможность использования процесса коксования на начальном этапе переработки высоковязкой и битуминозной нефти. В этом случае существенно упрощается проведение стадии ее обезвоживания, так как в технологической схеме замедленного коксования предусмотрено отделение бензиновой фракции от воды.*

Ключевые слова: *коксование, высоковязкие и битуминозные нефти, обводненные нефти, переработка нефти, способ переработки нефти*

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INTRODUCTION

In recent years, a characteristic feature of oil production is an increase in the total volume of high-viscosity bituminous oil, which contains a high amount of resin–asphaltene substances (RAS) and has high density and viscosity values. The technologies of extracting such oil include (1) the use of solvents, such as water, to displace fossil hydrocarbons (also known as cold recovery) and (2) in situ combustion where oil viscosity is reduced as a result of increasing the temperature and creating the pressure needed to displace the formed oil through gaseous combustion products [1–11]. In Russia, technologies based on the use of water vapor are used in the extraction of high-viscosity bituminous oil [8–11]. However, the use of water or water vapor to displace oil results to the inclusion of up to 95% water in the product stream from the production well.

Preparation of oil for processing involves its stabilization, desalination, and dehydration [12, 13]. Dehydration is based on the difference in the densities of petroleum hydrocarbons and water. However, due to the comparable densities of resin–asphaltene substances and water, their separation by gravitational method is almost impossible, and the high viscosity of RAS prevents the effective use of demulsifiers for the destruction of oil–water emulsions. As a result, traditional preparation schemes for processing of high-viscosity bituminous sands are ineffective [14].

A possible solution to the problem of involving such oil in the fuel, energy, and petrochemical balance is the use of a coking process at the first stage of its processing [15]. Currently, this process is widely used to increase the extent of oil refining, thereby obtaining distillate fractions from heavy oil residues (fuel oil, cracking residues, and tar). At the same time, the main components of such residues are RAS, and the technology involves the use of water vapor to prevent coking of the tubular furnace.

MATERIALS AND METHODS

Herein, a stable, high-viscosity oil–water emulsion (not amenable to dehydration) was subjected to a coking process, and its characteristics are presented in Table 1. The effects of the coking process conditions on the yield and characteristics of the resulting products were also investigated.

The density and paraffin content of the oil–water emulsion correspond to the fifth type of oil (bituminous) and to the first type (low paraffin) based on the classification in GOST 31378-2009[1] and OST 38.01197-80[2], respectively. However, the amount of water (more than the 1.0 wt % standard for the third type) and solids (more than 0.05 wt %) in the emulsion does not make it possible to be considered as a commodity product (GOST 31378-2009) for processing.

Coking was carried out in a laboratory installation in a “cube”[3] [16]. A hollow cylindrical

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[1] GOST 31378-2009. Interstate Standard. Crude Petroleum. General Specifications. Moscow: Standartinform; 2019.

[2] OST 38.01197-80. Industry standard. Oil of the USSR. Technological Indexing. Moscow: Ministry of Oil Refining and Petrochemical Industry of the USSR; 1980.

[3] Bikbulatova A.M. *Etapy stanovleniya i razvitiya otechestvennogo proizvodstva neftyanogo koksa metodom zamedlennogo koksovaniya: Na primere Novo-Ufimskogo NPZ* (*Stages of formation and development of domestic production of petroleum coke by the method of delayed coking: a case study of the Novo-Ufimsky refinery*): Cand. Sci. Thesis. Ufa; 2002. 104 p. (in Russ.).

Table 1. Characteristics of oil-water emulsion

Characteristics	Value
Density, 50°C, g/cm^3	1.0200
Dynamic viscosity at 50°C, MPa·s	22557
Molecular weight, g/mol	520
Content of mechanical impurities, wt %	0.72
Water content, wt %	18.0
Resin and asphaltene content, wt %	42.0
Paraffin content, wt %	1.0

apparatus was used as a reactor, which was equipped with a "pocket" for a chromel–alumel thermocouple and a nozzle for installing a pressure gauge. The raw materials were loaded into the reactor, and the connections of the components of the installation were checked for tightness. The reactor was heated to a set temperature using an electric furnace. The coking process temperature and pressure were varied from 500 to 700°C and from 0.10 to 0.35 MPa, respectively. During the experiment, the temperature and pressure in the reactor were monitored, as well as the volume of waste gas, which was recorded using a GSB-400 gas meter (*NII Electromera*[4], Russia). At the end of the experiment, the amount of the liquid products and coke formed was determined.

The hydrocarbon content in waste gas was determined by gas adsorption chromatography using a 3700 chromatograph (*Chromatec*, Yoshkar-Ola, Russia) that was equipped with a thermal conductivity detector. Conditions of the analysis are as follows: the current of the detector bridge was 90 mA, the carrier gas was nitrogen with a flow rate of 25 mL/min, the temperature of the column thermostat was at 70°C; the column length and diameter were 7 m and 2 mm, respectively, and the chromatographic phase was Al_2O_3 with a fraction of 0.15–0.25 mm and promoted with a 5% NaOH solution. To increase the sensitivity and simplify the calibration and processing of chromatograms, a heated reaction column for the conversion of hydrocarbons to hydrogen was installed in the gas line of the chromatograph, which was between the chromatographic column and the detector. The temperature, length, diameter, and nozzle of the reaction column were 850°C, 0.2 m, 5 mm, and copper oxide and iron chips (arranged sequentially), respectively. Registration of the detector signal and processing of chromatograms were carried out using the hardware and software complex EKOKHROM.

For liquid products, their fractional composition and pycnometric density were determined using an ARNP-1 apparatus (*Promkhimpribor*, Moscow, Russia) according to the GOST 2177-99[5] and GOST 3900-85[6] methods, respectively. Furthermore, the ash content of the carbon material was determined according to GOST 22692-77[7], and the mass fraction of the volatile substances was measured in accordance with GOST 22898-78[8].

RESULTS AND DISCUSSION

The output indicators of the coking process are shown in Table 2.

It can be seen that with an increase in temperature, the yield of gaseous products increases, the formation of coke decreases, and the dependence of the amount of liquid products on temperature increases with a maximum yield at 550–600°C. An increase in the pressure leads to an increase in the yield of gaseous products and coke but a decrease in the yield of liquid products.

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[4] https://electromera.ru/. Accessed October 23, 2021.

[5] GOST 2177-99. Interstate Standard. Petroleum products. Methods for determination of distillation characteristics. Moscow: Kodeks; 2001 (in Russ.).

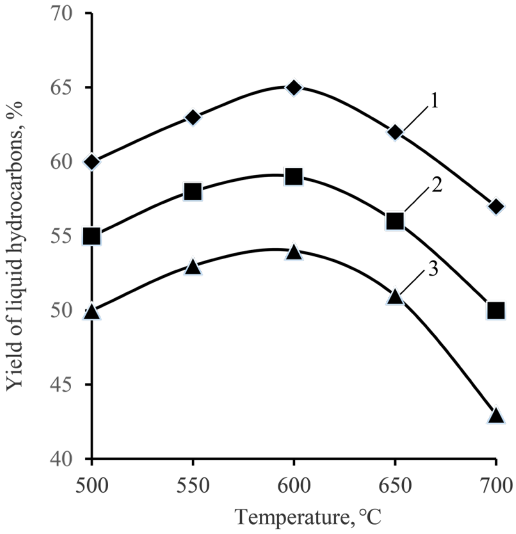
[6] GOST 3000-85. Interstate Standard. Petroleum and petroleum products. Methods for determination of density. Moscow: Standartinform; 2006 (in Russ.).

[7] GOST 22692-77. Interstate Standard. Carbon materials. Method for determination of ash. Moscow: IPK Izd. standartov; 2001 (in Russ.).

[8] GOST 22898-78. Interstate Standard. Low-sulphur petroleum cokes. Specifications. Moscow: IPK Izd. standartov; 2004 (in Russ.).

It is noted that ~98% of the emulsion water goes with liquid products and was subsequently easily separated by gravity, and the rest (~2%) was "encapsulated" by the resulting coke.

The effect of the coking process conditions on the yield of liquid hydrocarbons is shown in the figure.



Effect of the coking process conditions on the yield of liquid products. Coking pressure is at (1) 0.10 MPa, (2) 0.20 MPa, and (3) 0.35 MPa.

The characteristics of the products were only determined for those obtained under conditions where maximum yield was achieved, i.e., at 500°C (maximum coke yield) and 600°C (maximum yield of liquid products). In addition, the density of liquid products (hydrocarbons) varied from 0.8977 to 0.9346 g/cm^3, and it increased with an increase in temperature (from 500 to 600°C) and a decrease in pressure. The change in density is due to a change in the fractional composition of liquid hydrocarbons (Table 3).

It can be seen that at a lower temperature and a higher pressure, the amount of gasoline and kerosene fractions in liquid products is higher, which correlates with the densities of these products. The characteristics of the coke residue from the coking process are presented in Table 4.

In terms of density and amount of volatile substances and moisture, the resulting residues meet the GOST 22898-78 "Low-sulfur petroleum cokes. Technical specifications" requirements for coke. Due to a high amount of impurities in the raw materials, as well as the fact that they have not been desalted, the ash content of the resulting coke is significantly higher than that allowed for cokes used for structural purposes (no more than 0.8 wt %).

Table 2. Influence of coking process conditions on product yield

Process conditions		Output, wt %		
Temperature, °C	**Pressure, MPa**	**Gas**	**Liquid**	**Coke**
500	0.10	1.7	79.2	19.1
	0.20	6.5	72.8	20.7
	0.35	8.4	68.5	23.1
550	0.10	2.2	82.8	15.0
	0.20	7.0	76.4	16.6
	0.35	8.9	72.1	19.0
600	0.10	2.9	85.2	11.9
	0.20	7.7	79.0	13.3
	0.35	9.7	74.9	15.4
650	0.10	5.1	83.2	11.7
	0.20	11.4	75.8	12.8
	0.35	14.6	70.9	14.5
700	0.10	10.3	78.4	11.3
	0.20	19.9	67.8	12.3
	0.35	25.5	60.7	13.8

Table 3. Influence of the coking process conditions on the fractional composition of liquid products

Conditions of carbonization		Fraction content, vol %			
Temperature, °C	Pressure, MPa	Petrol (T_{bp} – 180°C)*	Kerosene (180–240°C)	Diesel (240–350°C)	Residue
500	0.10	12.4	34.3	28.8	24.5
	0.20	14.7	38.9	27.7	18.7
	0.35	25.0	40.0	25.2	9.8
600	0.10	11.3	26.4	30.3	32.0
	0.20	13.9	30.0	29.3	26.8
	0.35	22.3	31.5	27.3	18.9

* T_{bp} is the initial boiling point.

Table 4. Influence of the coking process conditions on the characteristics of the coke residue

Conditions of carbonization		Density of coke calcined at 1300°C, g/cm^3	Ash content, wt %	Mass fraction of volatile substances and moisture, wt %
Temperature, °C	Pressure, MPa			
500	0.10	2.10	2.51	10
	0.20	2.11	2.54	9
	0.35	2.12	2.57	8
600	0.10	2.11	2.82	9
	0.20	2.12	2.85	8
	0.35	2.13	2.87	7

Coking gases contained hydrogen (20 ± 5 vol %), methane (45 ± 5 vol %), and C_2–C_4 hydrocarbons. An increase in the temperature and pressure during the coking process led to an increase in the amount of hydrogen in the gas and a decrease in the amount of methane. The amount of the remaining components did not change significantly.

A characteristic feature of the resulting gas is the significantly low amount of alkenes. In the coking gases of residues from straight runs, the amount of unsaturated hydrocarbons can reach 50%, whereas in our case, it did not exceed 7 vol %. Obviously, this is due to the composition of the raw materials: low content of alkanes and high content of resins and asphaltenes that have polycyclic aromatic structures.

CONCLUSIONS

The results show the possible application of the coking process at the initial stage of processing high-viscosity bituminous oil. In this case, the dehydration stage is greatly simplified because the technological scheme of delayed coking allows the separation of the gasoline fraction from water. Also, the results have shown that with an increase in the coking process temperature, the yield of gaseous products increases, the formation of coke decreases, and the dependence of the amount of liquid products on temperature increases with a maximum yield at 550–600°C. The coking process temperature also affects the composition of liquid products. At a lower temperature, the amount of gasoline and kerosene fractions in liquid products is higher. In addition, an increase in pressure leads to an increase in the yield of gaseous products and coke and to an increase in the amount of low-molecular-weight fractions of hydrocarbons in liquid products. The characteristics of the coke formed from the coking process carried out in this study are comparable to those that are commercially produced. It is noted that during coking of water-containing oil, up to 98% of emulsion water goes with liquid products, and the remaining amount of water remains in the formed coke.

Authors' contributions

A.I. Nikolaev – formulation of the problem, planning and conducting experimental studies, determination of the physicochemical characteristics of the research objects and resulting products, processing the data obtained, preparation of the data obtained for publication.

B.V. Peshnev – formulation of the problem, planning and conducting experimental studies, processing the data obtained, preparation of the data obtained for publication.

E.V. Egorova – determination of the physicochemical characteristics of the research objects and the resulting products, processing the data obtained, preparation of the data obtained for publication.

The authors declare no conflicts of interest.

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