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## CONDITIONS OF THE HEAVY OILS GENERATION AND THEIR DISTRIBUTION IN THE TERRITORY OF UKRAINE

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The article examines the conditions of formation, transformation mechanisms, and spatial distribution of heavy oils in the major petroleum-bearing regions of Ukraine, including the Dniprovsko–Donetska Depression, the Precarpathian area, and the Crimean region. The aim of the study is to determine the conditions of heavy-oil accumulation and to characterize their distribution across Ukraine. It has been established that heavy oils, which are defined by increased density and a high content of resin–asphaltene components, are formed primarily through degradation processes affecting light and medium crude oils. This research integrates field observations, laboratory experiments, and theoretical concepts to evaluate the contributions of biodegradation, degassing, leaching of water-soluble fractions, and sulphate-induced oxidation to the formation of heavy oils. Particular emphasis is placed on the role of sulphate reduction and polysulphide formation, which intensify hydrocarbon oxidation and accelerate the transition of oils into heavier forms. Chromatographic analysis of n-alkanes from fields with different geological settings demonstrates systematic depletion of light hydrocarbons, shifts in carbon-number distribution maxima, and an increase in sulphur-bearing compounds. Experimental interaction of n-alkanes with sulphate and sulphide ions at low temperatures (40 °C) simulates natural hypergene processes and confirms the catalytic function of hydrogen sulfide in promoting hydrocarbon oxidation through polysulfide intermediates. The results highlight region-specific factors controlling heavy-oil formation, including burial depth, reservoir hydrochemistry, gas saturation, and the influence of meteoric waters. This integrated analysis enhances the understanding of heavy-oil evolution in Ukrainian basins and provides a conceptual framework for predicting the distribution of viscous hydrocarbon accumulations, which is crucial for optimizing exploration strategies and developing technologies for their efficient recovery.

*Key words:* heavy oils, biodegradation, sulphate reduction, polysulphides, hydrocarbon oxidation, n-alkanes distribution, chromatographic analysis, Dniprovsko–Donetska Depression, Precarpathian region, Crimean region.

**Introduction.** The important direction of the further development of oil production industry is the studying of hardly recoverable hydrocarbons, among which the significant part belongs to degraded oils with density more than 870 kg/m<sup>3</sup>. By the data of J. Hunt [5], B. Tisso and D. Vette [12] they are formed in the result of the destruction process (degradation), which includes: degassing, biodegradation (oxygen and sulphate oxidization) and leaching of the water-soluble components of oil.

**Research analysis.** Biramzhe and M. Bestuzhev [4] suppose that degassing and leaching of hydrocarbons change only the fractional composition of oils, but not their chemical composition. Their chemical composition undergoes the influence of outer agents-oxygen and sulphur. Sulphuring changes the oil composition and the chemical structure of its components. The experiment was carried out, during which oil distillates were heated to 150-170°C with 5 % sulphur powder during 5 hours in a cylinder with a mixer and a cooler. The following conclusions were drawn: a great number of asphaltenes were formed; distribution of sulphide compounds in the products, obtained in the laboratory, is analogous to natural ones in sulphide oils.

At studying the problem of the sulphur compounds origin in oil it is very important to investigate the process of the sulphur genesis.

H. Smith [11] considers two ways of sulphur formation under natural conditions. The first way is as follows: complex organic sulphur-bearing compounds, aminoacids, for example, which are like D-cysteine  $\text{HSCH}_2\text{CH}(\text{NH}_2)\text{CO}_2\text{COOH}$  or D-metione  $\text{CH}_3\text{S}(\text{CH}_2)\text{CH}(\text{NH}_2)\text{CO}_2\text{H}$ , are disintegrated forming molecular sulphur and hydrogen sulphide.

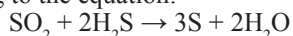
These data are proved by J. Hunt [5]: generation of  $\text{H}_2\text{S}$  takes place from organic compounds of sulphur, in the result of their disintegration in the source rocks at 130-150°C.

The second way is based on bacterial restoration of  $\text{SO}_4^{2-}$  in the presence of hydrocarbons, as a nutrient medium for microorganisms *Viebrio desulfuricans* with formation of hydrogen sulphide. Schematically this process looks like that:



Hydrogen sulphide, in its turn, interacts with  $\text{SO}_4^{2-}$ , that leads to formation of elementary sulphur.

S. Kushnir [8] calculated thermodynamic parameters of the process of the ion  $\text{SO}_4^{2-}$  restoration by various types of organic compounds. The obtained data show that all the reactions are endothermic, so they cannot be used by bacteria as a source for procuring energy. Thus, in the zone of hypergenesis an important role is played by not only biological, but purely chemical restoration of sulphates by hydrocarbons. All the obtained data show, that the ions  $\text{SO}_4^{2-}$  can really be reduced in formational waters to  $\text{H}_2\text{S}$  at the temperatures lower than 150°C, and the first product is not  $\text{H}_2\text{S}$ , but  $\text{SO}_2$ . Simultaneous presence of  $\text{H}_2\text{S}$  and  $\text{SO}_2$  in the system makes possible to form free sulphur according to the equation:



Thermodynamic calculations are given also by L. Anisimov [2]. They prove a principal possibility of formation of  $\text{H}_2\text{S}$  without bacteria participation, when  $\text{CaSO}_4$  interacts with the organic compounds, spread in the nature, at low temperatures.

Possibility of chemical reduction of ions  $\text{SO}_4^{2-}$  is closely connected with chemical activity of hydrocarbons, which increases in the following sequence: alkanes < arenes < naphthenes < carboxylic acids < carbo-hydrates [3].

**Oxidation.** The changes of components during the given process are evident, but geochemical medium at great depths is reducing, so the oxidation of the oil pools is connected only with the zones, which are in the contact with free air or formational sulphate waters.

The investigations of R. Biramzhe and M. Bestuzhev [4] show, that in the process of oxidation pitches and asphaltenes cannot form. Having analyzed the literary and their own experimental data they came to the conclusions: the influence of oxygen on the mixture of oil components goes selectively – at first polar and polycyclic components are oxidized, then aromatic ones, and saturated hydrocarbons are oxidized in the last place. Oxidation is accompanied with a number of secondary reactions, among which a special place belongs to the reaction of condensation of oxidized products. And it leads to the formation of heterocyclic compounds with high molecular mass.

**Biological action.** Chemical composition of oils can also be greatly changed under the influence of specific bacteria.

A. Jobson and others [6] gave the data of influence during 21 days of clean and mixed cultures of bacteria on oil of North-Cantal field, Saskatchewan (Canada); its density increased during this period from 827 to 1046 kg/m<sup>3</sup>.

The experiment data show that biodegradation goes in the following way: n-alkanes, isoprenoid alkanes, cycloalkanes and aromatic compounds [5, 6, 12].

R. Biramzhe and M. Bestuzhev [4] contend that biological actor of oil transformation is limited with only specific cases: during the given process only chemical composition is changed, but not the structure of oil molecules, for example-three-cycled naphthenes are not formed from monocycled ones, etc.

Literary review made possible to do the analysis of main mechanisms of oils degradation, which led to their density increasing at the expense of augmentation of the pitchy-asphaltene components part. And in the case of the light hydrocarbon migration and washing out the water-soluble ones and biodegradation this increasing possesses relative character, because the common quantity of pitchy-asphaltene matters in oil remains constant at decreasing the hydrocarbons common part.

The aim of this study is to determine the geological, geochemical, and hypergene conditions that control the formation, transformation, and spatial distribution of heavy oils in the major petroleum-bearing regions of Ukraine, and to identify the key degradation and oxidation processes responsible for their evolution.

**Materials and Methods.** *Source Materials and Study Objects.* Samples of oils from the Semenivka (Crimea), Bohdanivka (DDD), and Kokhanivka (Precarpathian) fields were analyzed to identify degradation mechanisms and conditions of heavy oil formation.

*Extraction and Preparation of n-Alkanes.* n-Alkanes were isolated using a urea complexation method, allowing selective extraction of straight-chain hydrocarbons.

*Chromatographic Analysis.* Flame-ionization detection was used with the following settings: evaporator 350°C, detector 300°C, program 60°C → 5°C/min → 320°C (20 min hold), nitrogen carrier gas (60 mL/min), two packed SE-30/INZ-600 columns. Identification was based on retention time correlations with standard mixtures.

*Laboratory Modeling of Hypergene Processes.* Model experiments involved contacting n-alkane mixtures with aqueous  $\text{SO}_4^{2-}$ ,  $\text{SO}_4^{2-} + \text{S}^{2-}$ ,  $\text{S}^{2-}$ , and elemental sulfur at 40°C for 300 days under anoxic conditions. Ion concentrations:  $\text{SO}_4^{2-} = 200 \text{ g/dm}^3$ ,  $\text{S}^{2-} = 10 \text{ g/dm}^3$ .

*IR Spectroscopy.* IR spectra were recorded to detect oxidation products. Significant absorption bands occurred at 1560–1620  $\text{cm}^{-1}$  (oxidized groups), 1660–1680  $\text{cm}^{-1}$  (quinoid structures), and 1700–1740  $\text{cm}^{-1}$  (esters). Strongest effects occurred with joint  $\text{SO}_4^{2-} + \text{S}^{2-}$  systems, confirming polysulfide formation.

Results were compared with natural chromatograms, depth, sulfur content, gas saturation, and alkane distribution maxima to evaluate biodegradation, degassing, leaching, and sulfate oxidation roles in heavy-oil formation.

**Results and discussion.** In Ukraine the fields of heavy oils are located in Dnieper-Donets depression (DDD), the Precarpathians and in the Crimea. The data of investigations of their physical-chemical parameters, the regularities of distribution and conditions of their bedding are given in the works [7, 9, 14]. By the results of these works the following regularities can be singled out:

- in DDD the pools of heavy oils are spread mainly in Middle Carboniferous deposits, the exception is Reshetnyaky field (Jurassic, 530 m);
- in the Precarpathians the main pools of heavy oils are concentrated in Jurassic deposits, the exception is Pavlivka field (Devonian, 960 m);
- heavy oils of the Crimea are met only in Miocene deposits of the Kerch peninsula.

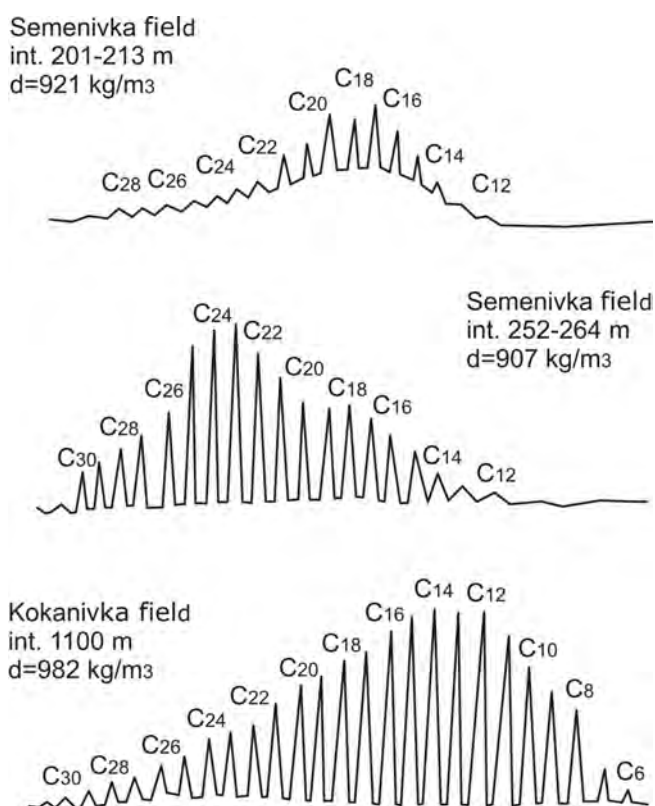
The range of the bedding depths of heavy oils is wide enough: from 100 m (Voykove field, the Crimea) to 4137 m (Kulychykhyne field, DDD). Practically a half (48 %) of the known fields is bedded at depth to 2000 m – these are the pools of the Kerch peninsula, of the Outer zone of Precarpathian deep, and also the separate pools of the Inner zone of the same deep and DDD.

Gas saturation for heavy oils is not high, and for 62 % of the fields does not exceed 50  $\text{m}^3/\text{m}^3$ .

Except the establishing of the common regularities of spreading we studied physical-chemical parameters, component composition, fractional content (table) and distribution of n-alkanes of heavy oils, which belong to the different oil-and-gas-bearing regions of Ukraine, are at different depths and vary among themselves by density and sulphur content [1].

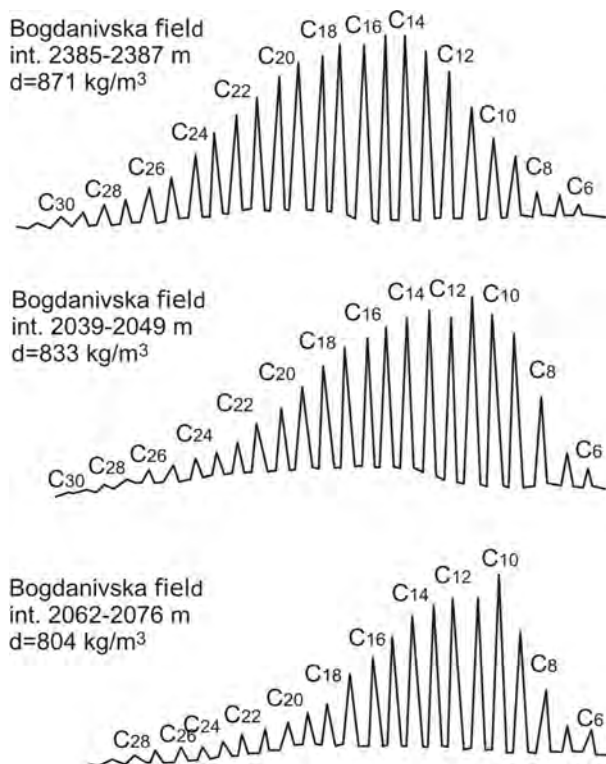
N-alkanes were extracted from oil by the method of complex-forming with carbamide with the following studying on chromatograph: flame-ionized detector, temperature of evaporator -350°C temperature of detector - 30°C, programming of thermostat - 60°C - 5°C/min - 320°C - 20 min - 320°C, gas-bearer nitrogen (the velocity of current - 60 ml/min), two packed columns - 10 % SE-30 on INZ-600. Qualitative identification of the chromatogram peaks was done by the graph of dependence of the n-alkanes time of retention (mixture: C<sub>8</sub>, C<sub>10</sub>, C<sub>12</sub>, C<sub>15</sub>, C<sub>16</sub>, C<sub>19</sub>, C<sub>24</sub>) upon the number of atoms of carbon in carboniferous skeleton.

In DDD we studied oils of Bogdanivka field. They belong to different oil-and-gas-bearing horizons and varies among themselves by meanings of density and sulphur content. Their chromatograms are shown in fig. 1. In fig. 2 the chromatograms of the n-alkanes distribution of Semenivka field (the Crimea) and Kokhanivka one (the Precarpathians) are given. On Semenivka field oils belong to two oil-and-gas-bearing horizons.



**Fig. 1. Chromatograms of the degraded oils of Semenivka and Kokhanivka fields**  
 Semenivka int. 201-213 m, d = 921 kg/m<sup>3</sup>; Semenivka int. 252-264 m, d = 906,6 kg/m<sup>3</sup>; Kokhanivka  
 int. 1100 m, d = 982 kg/m<sup>3</sup>

The analysis of the n-alkanes distribution in oils of Semenivka field and their fractional content (table 1) shows that they are strongly degraded. Taking into consideration a small depth of bedding of the investigated oils and low content of sulphur in them it is possible to state, that during their transformation the processes of biodegradation and degassing of the field prevail (gas saturation is 2,4 m<sup>3</sup>/m<sup>3</sup>). Besides these factors the influence of oxygen of the free air is also significant, because the field, which is situated nearer to the surface, has higher density, and maximum of the n-alkanes distribution is displaced in the direction of hydrocarbons with a longer carboniferous skeleton (fig. 1).



**Fig. 2. Chromatograms of the degraded oils of Bogdanivka field in DDD**  
 Bogdanivka int. 2385-2387 m, d = 871 kg/m<sup>3</sup>; Bogdanivka int. 2039-2049 m, d = 833 kg/m<sup>3</sup>;  
 Bogdanivka int. 2062-2076 m, d = 804 kg/m<sup>3</sup>

Table 1

**Physical-chemical parametres of the investigated oils**

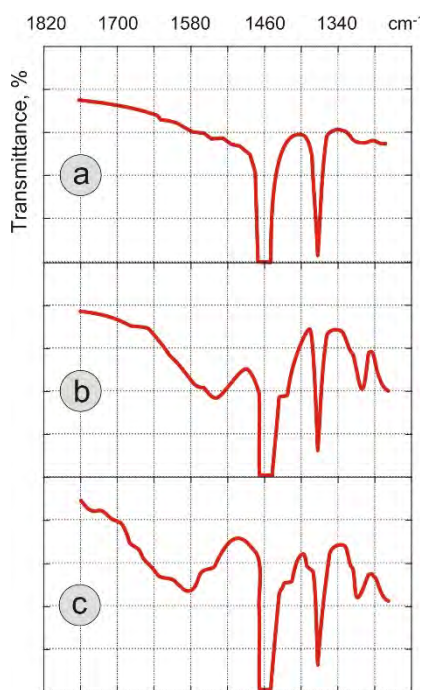
Field name	Age	Bedding depth, m	Density, kg/m <sup>3</sup>	Potential content of the boiling away fractions, %				Component composition, weight %			
				200°C	250°C	300°C	350°C	paraffin	sulphur	pitches	asphaltenes
Semenivka (the Crimea)	Karagan IV group of strata	201-214	921,3	-	1	30	53	0,24	0,16	16,1	traces
		252-264	906,6	1	17	39	56	0,73	0,16	15,5	traces
Bogdanivka (DDD)	C <sub>2</sub>	2385-2387	871,5	28	38	49	74	0,70	0,82	11,5	2,0
	-"	2039-2049	833,3	35	52	69	82	1,67	0,10	2,1	traces
	-"	2062-2076	804,6	54	70	81	90	0,55	0,05	1,4	traces
Kochanivka (the Precarpathians)	urassic	1100	982,0	6	18	30	36	1,62	6,13	23,0	17,5

Oils of Bogdanivka and Kokhanivka fields are bedded much deeper and have high sulphur content. It makes possible to state that in this case the process of sulphate oxidation plays an important role. It is proved by the fact that with increasing of sulphur content in oils the density also increases (table 1), the part of light components decreases, maximum of the n-alkanes distribution is displaced in the direction of hydrocarbons with a longer carboniferons skeleton (fig. 2).

During studying the process of oil degradation because of sulphate oxidation we made a number of laboratory investigations with different forms of sulphur. The mixture of n-alkanes was studied. This mixture without air contacted during 300 days at temperature 40°C with the solutions that contained  $\text{SO}_4^{2-}$  in one case; in the second –  $\text{SO}_4^{2-} + \text{S}^{2-}$ ; in the third –  $\text{S}^{2-}$ ; in the fourth – S (elementary sulphur). The solutions were prepared using formational water with the ions content (mg/cm<sup>3</sup>):  $C_{\text{Ca}^{2+}} = 118,65$ ;  $C_{\text{Mg}^{2+}} = 28,80$ ;  $C_{\text{Cl}^-} = 8,86$ ;  $C_{\text{HCO}_3^-} = 482,04$ ; for  $\text{SO}_4^{2-}$  and  $\text{S}^{2-}$  we fixed concentration (g/dm<sup>3</sup>):  $C_{\text{SO}_4^{2-}} = 200$ , and  $C_{\text{S}^{2-}} = 10$ .

During the investigations we obtained the following results:

1) On IR-spectra of the products of oxidation of the n-alkanes mixture with sulphate-ion (fig. 3, b) the increasing is shown in comparison with the initial mixture (fig. 3, a) of the stripes of absorbtion 1560, 1580, 1620, 1710 cm<sup>-1</sup>.

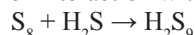


**Fig. 3. IR-spectra of the n-alkanes mixture**  
a) initial; b) oxidation with  $\text{SO}_4^{2-}$  ion; c) oxidation of  $\text{SO}_4^{2-}$  ions in presence of  $\text{S}^{2-}$

2) Joint presence of ions  $\text{SO}_4^{2-}$  and  $\text{S}^{2-}$  leads to the essential change of the interaction character in the system sulphate water-hydrocarbons. On IR-spectra the increasing of the stripes of absorbtion 1560, 1580, 1620, 1660-1680, 1700-1720, 1740 cm<sup>-1</sup> (fig. 3, c) is observed in comparison with the initial one. And in the given case the stripes 1660-1680 cm<sup>-1</sup> are shown more intensively, that corresponds to the chinoid structures, and additionally the stripe 1740 cm<sup>-1</sup> appears, that indicates to the presence of complex ethers.

3) IR-spectra of the hydrocarbons mixture, which contacted with water that contained free sulphur or sulphide-ion, retained without changes. This result lets to content that sulphur and sulphide-ion do not take part in the oxidation process independently, they only catalize this process. It is proved by the data by W.Toland [13], who states that sulphate-ion oxidizes hydrocarbons more rapidly in the presence of  $H_2S$ .

So on the base of experimental and literary data it is possible to state, that the main role of hydrogen sulphide is in the reaction of interaction with sulphur [5, 10]:



During this reaction polysulphides are formed, which are stronger oxydizers than sulphate-ion. It hastens and deepens the process of hydrocarbons degradation.

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## УМОВИ ФОРМУВАННЯ ВАЖКИХ НАФТ ТА ЇХ РОЗПОДІЛ НА ТЕРИТОРІЇ УКРАЇНИ

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Стаття розглядає умови формування, механізми трансформації та просторовий розподіл важких нафт у головних нафтоносних регіонах України, включаючи Дніпровсько–Донецьку западину, Передкарпаття та Кримський регіон. Метою дослідження є визначення умов накопичення важких

нафт та характеристика їхнього поширення в межах України. Встановлено, що важкі нафти, які характеризуються підвищеною густиною та високим умістом смолисто-асфальтенових компонентів, утворюються переважно внаслідок процесів деградації, що впливають на легкі та середні сорти сирої нафти. Дане дослідження інтегрує польові спостереження, лабораторні експерименти та теоретичні моделі з метою оцінити внесок біодеградації, дегазації, вилугування водорозчинних фракцій та сульфат-індукованого окиснення у формування важких нафт. Особливу увагу приділено ролі сульфатредукції та утворення полісульфідів, які інтенсифікують окиснення вуглеводнів і прискорюють перехід нафтових систем у важкі форми. Хроматографічний аналіз n-алканів із родовищ, що належать до різних геологічних обстановок, демонструє систематичне збіднення на легкі вуглеводні, зміщення максимумів розподілу за кількістю атомів карбону та зростання концентрацій сірковмісних сполук. Експериментальна взаємодія n-алканів із сульфат- та сульфід-іонами за низьких температур (40 °C) моделює природні процеси гіпергенезу та підтверджує каталітичну роль сірководню в інтенсифікації окиснення вуглеводнів через полісульфідні проміжні продукти. Отримані результати підкреслюють регіонально-специфічні чинники контролю формування важких нафт, серед яких глибина залягання, гідрохімічні властивості пластових вод, рівень газонасичення та вплив метеорних вод. Представлений інтегрований аналіз поглиблює розуміння особливостей еволюції важких нафт в українських осадових басейнах і формує концептуальну основу для прогнозування локалізації скупчень в'язких вуглеводнів, що має вирішальне значення для оптимізації стратегії пошуків та розроблення технологій їх ефективного видобутку.

*Ключові слова:* важкі нафти, біодеградація, сульфатредукція, полісульфіди, окиснення вуглеводнів, розподіл n-алканів, хроматографічний аналіз, Дніпровсько–Донецька западина, Передкарпатський регіон, Кримський регіон.

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