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REVIEW ARTICLE

Features of oil disperse systems and changes in their properties under low-energy wave action

Nadezhda A. Pivovarova✉
Astrakhan State Technical University, Astrakhan, 414056 Russia
✉ *Corresponding author, e-mail: nadpivov@mail.ru*

Abstract

Objectives. To analyze the properties, dispersion structure, and intermolecular interactions in oil disperse systems (ODSs) of various component compositions, and to consider the possibilities of controlling phase transitions in an ODS to achieve the most favorable dispersion properties for carrying out technological processes of refining hydrocarbons, upgrading petroleum products, or recycling oil waste. Phase transitions are proposed to be controlled by low-energy wave action (low-frequency ultrasound, constant magnetic field with low induction).

Results. The following features of oil disperse systems are considered and substantiated with literature data and the results of our own research: multiplicity of phases; diversity of hydrocarbon components and heteroatomic compounds contained in them; the nature of intermolecular interactions (the absence of charge interactions and the presence of charge–polarization and exchange spin-spin interactions); spin activity or paramagnetism of oil, petroleum products, and their various components; homolytic dissociation of high-molecular-weight and heteroatomic organic compounds, resulting in an increase in paramagnetism; as well as the presence of trace elements contained in organometallic compounds and in salts dissolved in emulsified water. An equation is presented for intermolecular interactions in ODSs, in which a decisive role is played by the exchange interactions caused by the presence of spin and spin-polarized molecules. Two-component models are described for the shells of complex structural units of the oil disperse system, their structure, and their interaction with the dispersion medium. Methods for controlling phase transitions and dispersity of ODSs are shown. Special attention is paid to low-energy wave technologies (ultrasonic waves with a frequency of 20–100 kHz with an intensity of up to 0.4 W/cm^2 and a constant magnetic field with an induction of less than 0.4 T). Examples are given for the intensification of such technological processes as separation of water–oil emulsions, removal of mechanical impurities, atmospheric and vacuum distillation, selective purification of oil fractions, degassing of heavy fuel oil to remove hydrogen sulfide, visbreaking, and others. Positive results are demonstrated for the application of low-energy technologies for reduction of the viscosity and pour point of oil and petroleum products in oil refining equipment. The study proposes a mechanism of the influence of a constant magnetic field on the flow of petroleum product or hydrocarbon raw material.

Conclusions. The study of the features of ODSs is key to the qualified colloid-chemical approach to processes of production, transportation, and processing of hydrocarbon raw materials. They are seen from the standpoint of ODS theory on the basis of the consideration of the dispersion structure and phase transitions in ODSs. Low-energy technologies in the petroleum industry are an effective tool for resource saving in various processes and optimization of their parameters without significant material costs.

Keywords
oil disperse systems, asphaltene paramagnetism, complex structural unit, dispersity of oil systems, low-energy wave action, magnetic field, ultrasound

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ОБЗОРНАЯ СТАТЬЯ

Особенности нефтяных дисперсных систем и изменение их свойств при низкоэнергетических волновых воздействиях

Н.А. Пивоварова✉

Астраханский государственный технический университет, Астрахань, 414056 Россия

✉ *Автор для переписки, e-mail: nadpivov@mail.ru*

Аннотация

Цели. Проанализировать особенности свойств, дисперсного строения, межмолекулярных взаимодействий в нефтяных дисперсных системах (НДС) разнообразного компонентного состава, а также рассмотреть возможности управления фазовыми переходами в нефтяной системе для достижения наиболее благоприятных дисперсных свойств при проведении технологических процессов переработки углеводородного сырья, облагораживания нефтепродуктов или утилизации нефтяных отходов. В качестве инструмента регулирования фазовыми переходами используют низкоэнергетические волновые воздействия (низкочастотный ультразвук, постоянное магнитное поле с невысокой индукцией).

Результаты. Рассмотрены и аргументированы литературными данными и результатами собственных исследований такие особенности НДС, как мультидисперсность; многообразие углеводородных компонентов и гетероатомных соединений, входящих в них; природа межмолекулярных взаимодействий (отсутствие зарядовых взаимодействий, зарядово-поляризационные и обменные спин-спиновые взаимодействия); спиновая активность или парамагнетизм нефти, нефтепродуктов и различных их компонентов; гемолитическая диссоциация высокомолекулярных и гетероатомных органических соединений, приводящая к росту парамагнетизма; присутствие микроэлементов, входящих в состав металлорганических соединений и в виде солей, растворенных в эмульгированной воде. Приведено уравнение межмолекулярных взаимодействий в НДС, решающую роль в которых играют обменные взаимодействия, обусловленные присутствием спиновых и спинполяризованных молекул. Представлены двухкомпонентные модели оболочек сложных структурных единиц НДС, их строение и взаимодействие с дисперсионной средой. Показаны способы управления фазовыми переходами и дисперсностью НДС; особое внимание уделено волновым низкоэнергетическим технологиям (с использованием ультразвуковых волн с частотой 20–100 кГц с интенсивностью до 0.4 Вт/см^2 и постоянного магнитного поля с индукцией меньше 0.4 Тл). Даны примеры интенсификации таких технологических процессов, как разделение водонефтяных эмульсий, очистка от механических примесей, атмосферная и вакуумная перегонка, селективная очистка масляных фракций, дегазация мазута от сероводорода, висбрекинг и др. Показаны положительные результаты применения низкоэнергетических технологий для снижения вязкости и температуры застывания нефти и нефтепродуктов в оборудовании нефтепереработки. Предложен механизм влияния постоянного магнитного поля на поток нефтепродукта или углеводородного сырья.

Выводы. Изучение и учет особенностей НДС позволяет реализовать квалифицированный коллоидно-химический подход к процессам добычи, транспортировки, переработки углеводородного сырья с позиций теории НДС, основанный на анализе дисперсной структуры и фазовых переходов в НДС. Применение низкоэнергетических технологий в нефтяной отрасли — эффективный инструмент для ресурсосбережения в различных процессах и оптимизации их параметров без существенных материальных затрат.

Ключевые слова

нефтяные дисперсные системы, парамагнетизм асфальтенов, сложная структурная единица, дисперсность нефтяных систем, низкоэнергетические волновые воздействия, магнитное поле, ультразвук

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INTRODUCTION

Hydrocarbons are an invaluable resource for producing fuels, lubricants, construction and road materials, electrical insulating materials, and solvents. They are also a source for the production of a variety of petrochemical products. It is difficult to overestimate the importance of petroleum products as a source of energy for a wide variety of industries, household needs, and transportation. Over the past 150 years, a colossal infrastructure has been created focused on the consumption of oil as a fuel source. The number of vehicles on Earth powered solely by internal combustion engines has reached 1.5–2 billion [1].

The deep refining of hydrocarbon feedstocks is performed using various physical and physicochemical processes, catalysts, high temperatures and pressures, complex process equipment, and so on. However, increasing the depth of oil refining remains a pressing issue, given that fossil hydrocarbons are a finite resource. This task is further complicated by the ongoing trend toward deteriorating hydrocarbon feedstock quality: increasing levels of sulfur and organometallic compounds, mechanical impurities, resins and asphaltenes; increasing density and viscosity; and decreasing potential for light fractions.

The Energy Strategy of the Russian Federation to 2050[1] defines the need to increase oil refining depth to 90% by 2030 and to fully meet the demand of the domestic market in petroleum products, and to produce motor fuels with the highest environmental standards. This requires the implementation of new technological solutions for hydrocarbon processing.

According to Academician P.A. Rehbinder, the founder of the school of physicochemical mechanics of disperse systems: “Optimization of the parameters of technological processes should be based on the most favorable content of physicochemical, thermal, and mechanical factors—changes in the chemical composition, including the addition of active substances, temperature, and the effects of external forces; optimal processes should always be complex. The task of the new field of knowledge, combining a number of problems of rheology, colloid chemistry, molecular physics, mechanics of materials, and the technology of their production, consists, first of all, in establishing the mechanism and patterns of the process of formation, deformation, and destruction of structures in order to control these processes” [2].

A qualified colloid-chemical approach to the production, transportation, and processing of hydrocarbon raw materials from the standpoint of the theory of oil disperse systems (ODSs) considering the dispersion structure and phase transitions in ODSs opens up significant opportunities for improving the efficiency and resource saving of various processes. It will also optimize their parameters without significant material costs. Scientists from Professor Z.I. Sunyaev’s scientific school “Physicochemical Foundations and Technologies of Oil Disperse Systems” continue to study the non-linear behavior of mixtures of hydrocarbon raw materials and petroleum products, accompanied by synergistic and antagonistic effects, and to investigate nonadditive effects in hydrocarbon feedstock processing [3–7].

The theoretical concepts and applied research of this school are based on the postulate of the relationship between the chemical and structural-group composition of the dispersed phase and its dispersity and physicochemical properties. The dispersed phase is considered to be a supramolecular structure: a complex structural unit (CSU) with characteristic composition, structure, properties, and internal interactions [8].

The discovery of the paramagnetic activity of molecules containing unpaired electrons reveals the nature of intermolecular interactions in CSU, and determines many features of the behavior, structure, and properties of CSE and ODS [9].

R.Z. Safieva’s review presents a systematic analysis of concepts regarding ODSs. Various approaches to accumulating and analyzing knowledge about ODSs (analytical, colloid-chemical, modeling, and chemoinformatic) were demonstrated. Each of these allows for the acquisition of digital data for the development of an information-based approach—petroinformatics. Concepts of the structure and properties of ODSs are based on the modern view of phase transitions involving asphaltenes and the understanding of the processes occurring within them when their conditions of existence change. These concepts provide the key to controlling phase transitions during hydrocarbon processing or to improving the quality of petroleum products [10].

The methods for influencing ODSs to control their structure can be divided into two groups: the introduction of new components into the system (additives, solvents, dopants, etc.) and wave action (mechanical or electromagnetic) of varying intensity. Mechanical actions are exerted by acoustic waves (including ultrasonic), cavitation, vibrating jets, pulsed

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[1] Order of the Government of the Russian Federation of April 12, 2025, No. 908-r. “Energy Strategy of the Russian Federation for the Period up to 2050.” http://static.government.ru/media/files/LWYfSENa10uBrrBoyLQqAAOj5eJYlA60.pdf. Accessed November 22, 2025 (in Russ.).

impacts, cavitation mills, and elastic low-frequency vibrations generated by the column of pumped liquid. Electromagnetic actions are performed by constant and alternating electromagnetic fields. An alternating magnetic field is characterized by its frequency (from infrared to microwave and radiation).

In terms of intensity, wave actions can be conditionally divided into weak actions, which change the balance of intermolecular interaction forces, dispersion composition, and structure of an ODS, but do not affect the structure of molecules, and strong actions, which lead to a change not only in the dispersion composition but also in the molecular composition, i.e., cause chemical reactions [11].

Among the wave actions of interest are weak, or low-energy, wave actions (LWAs), produced by ultrasonic and constant magnetic fields. Their energy, as a rule, is significantly lower than the bond energies in hydrocarbons and heteroatomic compounds in ODSs.

The aim of this paper is to analyze features of the structure, properties, and intermolecular interactions of ODSs of various compositions and the possibility of controlling them using low-energy wave actions. The characteristics of ODSs and their differences from classical colloids are discussed.

Multiplicity of phases in oil disperse systems

One of the features of ODSs is the multiplicity of phases: the ability of two or more phases (solid, liquid, and gaseous) to coexist in a single system. Examples of such ODSs include crude oil and unstable gas condensate, which contain dissolved gases, mineralized water, mechanical impurities of rocks, etc. The gas content in reservoir oil can reach 300–500 m^3/m^3 or more. Its typical value for most grades of oil is 30–100 m^3/m^3.[2] For gas condensate, this value can exceed 600 m^3/m^3. For example, the unstable gas condensate of the Astrakhan gas condensate field contains 55–75 vol % dissolved gases. The water content in oil and the content of mineral salts also vary widely: up to 90 wt % and up to 10 g/dm^3, respectively [12].

An even greater variety of dispersed inclusions is typical of waste oils, oil-containing waste, and sludge, in which the concentration of dispersed impurities, both inorganic and organic, varies over wide ranges.

The variety of hydrocarbon components and heteroatomic compounds contained in ODSs

Another characteristic of ODSs is the diversity of hydrocarbon components and the heteroatomic compounds which they contain. They differ in their properties, structure, shape, and molecular size. For example, more than 140 different hydrocarbons were detected using chromatography in the 28–180°C gasoline fraction obtained at the Astrakhan Gas Processing Plant.[3]

Oil contains over 1000 identified organic compounds, but a much larger number remains unknown. Fourier-transform ion cyclotron resonance mass spectrometry has demonstrated that oils contain tens of thousands of compounds. The data obtained was processed in the form of a two-dimensional diagram corrected to the Kendrick mass using a method based on the idea that the oil system is characterized by continuous distribution of the molecular weights of the original components. A similarly complex structure is characteristic of heavy fractions and residues of oil in which molecules are complex hybrid structures containing saturated and unsaturated bonds. They are heteroatoms of sulfur, nitrogen, oxygen, and other elements. It was reported [10, 12, 13] that approximately 30 metals have been found in the organic compounds of oil.

Intermolecular interactions in ODSs: the absence of charge interactions and the presence of charge–polarization and exchange spin–spin interactions

Another feature of ODSs is the absence of charge interactions between molecules in them. Conventional colloid chemistry represents electric charge as force centers, the main reason for the formation of colloidal particles. These concepts of Coulomb interactions are erroneously transferred to hydrocarbon media. In ODSs, there are no charged molecules, cations, and anions, easily traced by measurements of electrophoresis and the permittivity of petroleum products [9]. Among the variety of organic compounds in raw hydrocarbon materials and petroleum products, there are molecules containing functional groups of an acidic or basic nature, which are more or less obvious dipoles. Organic acids contained in oil have weak polarity. According

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[2] Gas content (gas saturation) of reservoir oil. URL: https://neftegaz.ru/tech-library/ngk/148222-gazosoderzhanie-gazonasyshchennost-plastovoy-nefti/. Accessed February 08, 2019 (in Russ.).

[3] The analysis was conducted at the plant's central laboratory with the author's participation. The data have not been published.

to the results of potentiometric titration, the content of benzoic and humic acids in the studied high-paraffin oil was 0.14 mg KOH/g [14]. Petroleum acids, as well as phenols, were detected in the resinous oil of the Irkutsk oblast [15].

Organic acids were reported to participate in the sedimentation process [10]. The presence of high-molecular-weight tetrameric naphthenic acids (C_{80}) leads to the formation of naphthenate deposits during oil production and transportation.

It is also known that asphaltenes and resins are polar [3, 7, 8, 16, 17]. For example, it was shown [17] that, depending on the structure, molecular weight, number of aromatic cycles, and the presence of heteroatoms, the dipole moment of asphaltenes can vary widely from practically zero to 3.12 D.

Polarity was indicated as one of the causes of the self-aggregation of asphaltenes, with the π interaction between aromatic rings and the electron transfer being noted [18]. The solubility of asphaltenes in aprotic solvents with a low ionization potential was associated with the formation of donor–acceptor complexes [19].

Asphaltenes are insoluble in alkanes but dissolve well in light aromatic hydrocarbons (benzene, toluene), carbon disulfide, and carbon tetrachloride. All of the above solvents have a dipole moment equal to or close to zero (only toluene has a dipole moment of up to 0.37 D). On the other hand, asphaltenes are also soluble in polar substances such as pyridine and chloroform (2.2 and 1.15 D, respectively), but are insoluble in polar ethyl alcohol and ethyl ether (1.74 and 1.18 D, respectively).

The solubility of resins is significantly higher: from individual low-molecular-weight solvents (*n*-alkanes C_5–C_7, C_8, and others), petroleum ether, benzene, and toluene, which have no dipole moment, to alcohols, ethers, and organic acids, which have a dipole moment of 1.15 D and higher. Of course, resins are soluble in all substances in which asphaltenes dissolve.

The ambiguous behavior of resins and asphaltenes indicates that polarity is not a determining property in the interactions of the components of ODSs, charge interactions are absent, and charge–polarization forces are limited.

Paramagnetism of oil and petroleum products

A distinctive feature of ODSs is paramagnetism. According to F.G. Unger's theory [9, 20, 21], the intermolecular interactions in hydrocarbon environments are dominated by the exchange interactions caused by the presence of molecules with stable radicals (with uncompensated spin) and estimated by the number of paramagnetic centers (PMCs). This property is characteristic of a wide range of petroleum products: from gasolines (10^{15} spin/g) to asphaltenes, carbenes, and carboids (up to 10^{23} spin/g). The number of PMCs in oil residues was experimentally measured [22] to be $3 \cdot 10^{18}$ spin/g PMCs in heavy fuel oil of the Astrakhan gas condensate field and $4.2 \cdot 10^{19}$ spin/g PMCs in tar of the Syzran oil refinery. The content of free radicals in Venezuelan oils and residues was analyzed [23]. The results showed that, in heavy oils, the PMC content ranges from $2.4 \cdot 10^{17}$ to $1.31 \cdot 10^{19}$ spin/g, and in the vacuum residues of these oils, the PMC content increases by one to two orders of magnitude—up to $9.12 \cdot 10^{19}$ to $1.372 \cdot 10^{20}$ spin/g.

It follows that a dominant role in the formation of colloidal particles in ODSs is played by the exchange interactions of the spin origin. Along with spin-containing molecules with explicit spin moments, ODSs also contain spin-polarized molecules, which similar to charge dipoles contain electrons with opposite spins, but separated in space. This creates a spin dipole which plays a significant role in exchange interactions during the formation of the ODS structure [9].

Homolytic dissociation of components of an oil system

The number of PMCs in ODSs changes under the action of external factors (temperature, solvent) due to homolytic dissociation. Molecules with the lowest energy of forbidden reversible singlet–triplet transitions are separated into fragments. Each receives one electron from the broken bond, which leads to an increase in the number of free radicals. The phenomenon of homolytic dissociation was explored [20] during the dissolution and heating of asphaltene precipitates obtained by different methods. The NMR spectra of the samples differed depending on the nature of the solvent, the degree of heating, and the rate of removal of the precipitate from the solvent, even with the same feedstock. During dissolution or heating, the molecule passes into an excited state or undergoes cleavage of a weak bond to form a radical pair. The molecule becomes a triplet. Due to the positive sign of the interaction energy with solvent molecules (alkanes), these newly formed paramagnetic species precipitate along with the other paramagnetic species. When the precipitate is removed from the solvent, i.e., when paramagnetic materials are concentrated, conditions arise for a reverse triplet–singlet transition, i.e., for the recombination of radical pairs. The energy of such transitions is low and comparable to the energies of intermolecular interactions.

Similar conclusions were reached by M.Yu. Dolomatov in a study of the effect of heating the asphaltene concentrate from the second stage of propane deasphalting on its paramagnetic, electrically conductive, and viscous properties. It was shown that an increase in the number of PMCs in asphaltenes is associated with an intensification of the thermal decomposition of metastable compounds due to the rupture of weak carbon bonds [17, 23].

Heating of heavy Venezuelan oils to 160–170°C leads to an increase in the number of free radicals. For the vacuum residues of these oils, the process of homolytic dissociation continued until approximately 230°C [24].

Since a change in the number of PMCs affects the behavior of ODSs in various refining processes, B.P. Tumanyan proposed the introduction, along with other physical and chemical properties, of a new evaluation parameter: the paramagnetic activity factor of a system. This factor more comprehensively assesses the quality of oil systems for their competent preparation for refining [6].

Presence of trace elements in ODSs

One of the features of ODSs is also the presence of trace elements contained in oil which contribute to the overall picture of interactions in ODSs. More than 60 trace elements have been found in oils of various origins, including more than 30 metals [3]. The presence of transition *d*-metals in oils and petroleum products predetermines the existence of PMCs. Paramagnetism is most noticeable in petroporphyrins, in particular, in vanadyl porphyrin complexes, concentrated mainly in asphaltenes [25]. According to S.N. Khadzhiev, the content of transition metals in oil varies widely [26]. For example, in West Siberian oils of the Shaimskoe field, their concentrations (g/t) vary in the following ranges: Fe, 5.03–52.20; Mn, 0.02–0.64; V, 0.18–11.63; Ni, 0.41–17.06; and Cr, 5.3–32.7; at lower concentrations, Zn, Ti, Mo, Co, etc. are found. A correlation was noted between the contents of vanadyl porphyrins ultraheavy oil V(IV), sulfur, and nitrogen and the number of PMCs. The largest portion is found in resins and asphaltenes. The correlation between the concentrations of sulfur and vanadium was confirmed [16], but the relationship between the concentration of vanadium and the content of asphaltenes in tars proved to be inverse.

The electron paramagnetic resonance spectra of the samples of heavy and extra heavy Venezuelan crude oil and its vacuum residues showed an intense central signal related to free radicals and a multiplet due to the presence of porphyrin V^{+4} [24].

It should be noted that paramagnetism was also detected in oil-associated hardness salts in emulsified water globules. For example, the number of PMCs in the sediments of waters isolated from oil ranged from $9.59 \cdot 10^{19}$ to $1.61 \cdot 10^{20}$ spin/g [9, 21].

Equation of intermolecular interactions in ODSs

All the above features determine specific intermolecular interactions characteristic of ODSs, namely, the formation of stable associative combinations based on exchange interactions caused by the spin and spin-polarized properties of molecules. This approach to the formation of colloidal particles was proposed by F.G. Unger. The development of quantum-chemical concepts made it possible to define a pair interaction operator *U* as a set of expressions depending on the distance between the interacting particles and the nature of the bond [9]:

$$U(R) = \pm k e^{-k_0 R} \pm k_1 R^{-1} \pm k_2 R^{-2} \pm$$
$$\pm k_3 R^{-3} \pm k_4 R^{-4} \pm k_5 R^{-5} - k_6 R^{-6},$$

wherein *k* are coefficients that can be represented as constants or functions, not necessarily linear; *R* is the distance between particles; and *e* is the base of the natural logarithm.

In this equation, each term accounts for interactions of only a specific type. The contribution of each subsequent term to the overall balance of intermolecular interactions decreases as the absolute value of the exponent of the term increases. The first term is the operator of exchange interaction between radicals and/or triplet (or higher-order multiplet) paramagnetic molecules to form a homeopolar (covalent) bond. The second term is the operator of pair interaction between two ions and characterizes charge (Coulomb) interactions. The third term is the operator of interaction between a charge and a charge dipole, resulting in charge interaction and the formation of ordered associative combinations. The fourth term is the operator of resonant interaction, i.e., the interaction of a radical (a paramagnetic molecule, a spin-containing particle) with a spin-polarized diamagnetic molecule. As in the previous case, a free radical can form an ordered associative combination with a certain number of spin-polarized molecules based on exchange interactions. Exchange interactions can be transmitted throughout the volume of a liquid by analogy with charge interactions. The fifth term is multipole and reflects the charge nature of the dipole, quadrupole, and multipole moments. The sixth term is also multipole but determines the spin nature of the dipole, quadrupole, and multipole moments. The seventh term describes only the van der Waals attraction of particles; therefore, it has a negative sign and characterizes long-range forces. These forces are represented by various interactions: orientational, dispersion, and induction ones.

From the analysis of F.G. Unger’s equation, it can be concluded that the first, fourth, and sixth terms describe exchange interactions in oil systems. Moreover, the highest contribution is made by the potential of pair interaction between two free radicals. This is the energy of a covalent bond. It is such bonds that exist in hydrocarbon molecules. For example, the energies of carbon–carbon bonds in alkanes are known to reach 440 kJ/mol, while in molecules containing oxygen, sulfur, and nitrogen heteroatoms, they are somewhat lower, around 270–350 kJ/mol. Resonant interactions between a radical and a diamagnetic spin-polarized molecule determine their attraction or repulsion. In ODSs, such forces arise between the paramagnetic core (most often asphaltene) and the nearest layers consisting of diamagnetic molecules capable of spin multipole-multipole interactions of diamagnetic molecules. These molecules are located in the outermost layers of the solvation shell; i.e., they are furthest from the core.

It should also be noted that, in ODSs, there are donor–acceptor interactions in the form of hydrogen bonding. Strong hydrogen bonds (60–200 kJ/mol), intrinsic to water and aqueous solutions, are not typical of components of oil systems. Heteroatoms and arene fragments tend to form medium hydrogen bonds (4–6 kJ/mol). Weak hydrogen bonds (0.5–4.0 kJ/mol) are formed by almost all petroleum hydrocarbons. The contribution of hydrogen bonds to the potential energy of interaction of nonpolar and polar compounds can reach 99%. The energy of a hydrogen bond includes three components: the electrostatic energy of attraction, which prevails at large distances; the energy of polarization (orientation and induction interactions) and charge transfer, which manifests itself as the distance decreases and promotes the attraction of molecules; and the energy of repulsion. Thermal exposure favors the destruction of hydrogen bridges with a bond energy of up to 40 kJ/mol [9].

Quantum-chemical calculations and experimental studies by M.Yu. Dolomatov [27] showed that a significant contribution to the intermolecular interactions of molecules in ODSs with the participation of asphaltenes is made by hydrogen bonds. The formation of a charge-transfer complex, and the ionization potentials and electron affinity are consistent with the data on optical density, electron affinity, and electrical conductivity.

The second, third, and fifth terms of the equation describe Coulomb interactions. Since charged molecules are absent in oil systems, and charge dipoles are present in very small quantities, their role in the formation of an ODS according to the presented concepts [9, 21] can be neglected.

On the other hand, a study of the viscosity increase mechanism in crude oil using electrostatic potential analysis and molecular dynamics established electrostatic interactions between asphaltene fragments [28]. Asphaltenes were modeled as normal (neutral), acidic, and anionic and explain the interactions of the components in the system by the presence of positive and negative electrostatic potential on the surfaces of their molecules. The extensive nonpolar space on the surface of normal asphaltenes facilitates their interaction with crude oil molecules (non-Coulomb interactions).

Quantum-chemical calculations using density functional theory determined the energy of intermolecular interactions between fragments of asphaltene nanoparticles in the range from 33.85 to 113.42 kJ/mol [27]. The stability of nanocluster formation was explained by donor–acceptor interactions between naphthenic–aromatic fragments.

The complexity of the structure of asphaltenes and various approaches to their isolation from oil systems, instrumental and chemical analysis, and mathematical and quantum-chemical description give rise to different models of intermolecular interactions, each of which has its own limitations and disadvantages.

Two-component models of complex structural units of ODSs

All of the above-mentioned features of the composition of ODSs and the intermolecular interactions within them determine a certain ordered organization of ODSs which consist of structural elements. According to Z.I. Sunyaev, structural elements of ODSs are defined as sets of interacting elements of ODSs which retain their physicochemical characteristics and composition in space and time. Particles of the dispersed phase of ODSs are characterized by a specific structural organization. They determines the properties of the entire system and its susceptibility to various external influences. A particle of the dispersed phase is called a complex structural unit (CSU) [8].

As mentioned earlier, ODSs are multiphase and polydisperse systems. To a first approximation, they can be represented as systems containing two-component CSUs representing various dispersed inclusions. Figure 1 presents examples of the most common two-component models of CSUs of liquid ODSs.

An ODS consisting solely of hydrocarbons and various organic heteroatomic compounds (Fig. 1a) is a dispersion medium (diamagnetic hydrocarbons) and a CSU containing the heaviest components with unpaired radicals and surrounding layers arranged according to the exchange interaction potential. The composition of the core and the number of layers depend on the nature of the ODS.

Studies of the composition of CSUs by I.R. Khairutdinov and F.G. Unger [9, 29] using radioactive tracers showed that the solvate shells isolated from

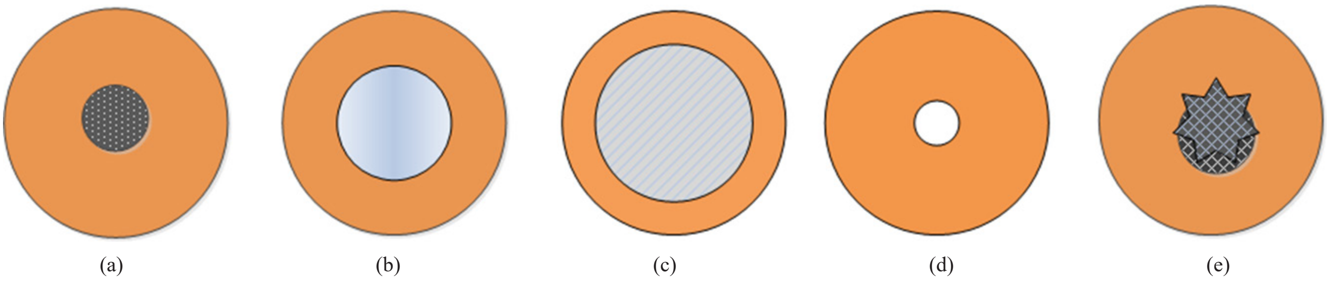


Fig. 1. Types of two-component models of complex structural units of oil disperse systems containing different dispersed phases in the dispersion environment:
(a) high-molecular-weight condensed aromatic compounds;
(b) water globule;
(c) high-molecular-weight paraffins;
(d) gas bubble;
(e) inorganic particles, mechanical impurities

residual petroleum products contain low-molecular-weight maltenes, petroleum acids, thiols, phenols, and other heterocyclic compounds with weak paramagnetic properties. Along with them, high-molecular-weight aromatic compounds and resins are also present, with strongly manifested paramagnetism. The cores in liquid ODSs may also contain dense high-molecular-weight formations representing spin-containing particles.

For heavy distillates and residual petroleum products, the composition of the CSU can be described by a hypothetical model in which the core consists of highly condensed aromatic compounds with high paramagnetism (asphaltenes). The surrounding layers, in order of decreasing paramagnetism, contain aromatic and heteroatomic components with a smaller number of condensed rings, then condensed naphthenic–aromatic compounds and paraffin–naphthenic hydrocarbons with polycycles and with signs of spin polarization. The dispersion environment consists of paraffinic or paraffinic–naphthenic hydrocarbons without signs of spin polarization [9].

The photocolorimetrically determined sizes of CSUs vary depending on the nature of the petroleum product and range from tens of nanometers for light distillate fractions to hundreds of nanometers for vacuum distillates and residues [3, 30].

In emulsified oil systems (inverse (water-in-oil) emulsions), the dispersed phase is water (Fig. 1b). Water-in-oil emulsions have been an object of study for decades. Surfactants are adsorbed at the interface, and the surface layer differs sharply from the properties of the inner and outer parts of the adjoining phases. This is because the molecules comprising this layer have an excess of surface energy. The interaction energy of the solvation layer with molecules of the dispersed phase depends on the composition of the dispersed phase and determines the size and stability of the system.

A study of the interfacial layer of water-in-oil emulsions showed that it consists predominantly of asphaltenes [31, 32]. A more detailed analysis of structural-group composition revealed that the concentration of asphaltenes in the interfacial layers is 6.7–7.2 times higher than that in oil, and the content of resins is 2–3 times lower [33]. Figure 2 shows water globules enclosed in a complex shell consisting of a first layer of asphaltenes adsorbed directly at the interface and a second layer of resinous substances adjacent to the asphaltenes as a result of exchange interaction. The shells prevent the coalescence of the water globules.

The dispersion medium in a water-in-oil emulsion is inhomogeneous. It consists of hydrocarbons and organic heteroatomic compounds and, in turn, forms a dispersion environment with higher dispersity. Here intermolecular interactions form a CSU similar to that described in Fig. 1a. The dispersities of emulsified globules of water and the oil phase can differ from several to thousands of times, depending on the composition of the emulsion. The closer the dispersities of the water globules and the oil phase, the more stable the emulsion. This is a manifestation of such a property of the dispersion medium as polydispersity. The particle sizes of the aqueous dispersed phase in water-in-oil emulsions range from hundreds to nine thousand nanometers [34]. Figure 2 illustrates the structure of the disperse system of a water-in-oil emulsion.

The following two-component model of an ODS (Fig. 1c) contains a CSU, the core of which is high-melting paraffins [6, 14, 35–39]. The associative behavior of *n*-alkanes in ODSs and its change under different temperature conditions were investigated using dynamic light scattering methods, as well as optical, electron, and atomic force microscopy [35]. The associates of *n*-alkanes are shown to have a shell,

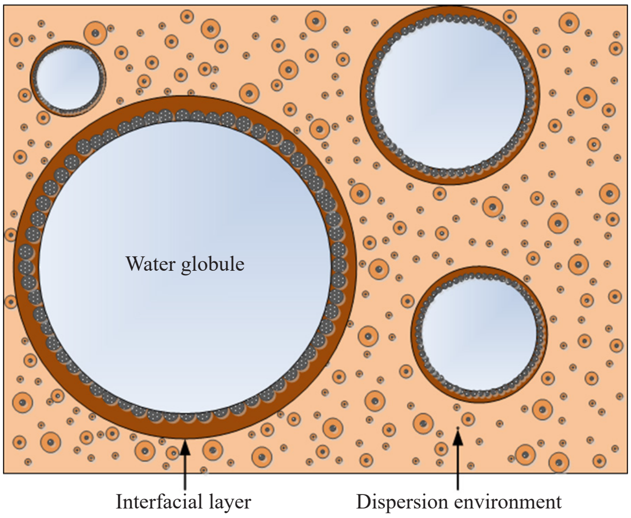


Fig. 2. Structure of the disperse system of a water-in-oil emulsion with the interface consisting of layers

a specific internal composition, and a structure. In the paraffinic oil system under investigation, the associates are preserved even at elevated temperatures. A study of the effect of paraffin concentration on the dispersed structure of hydrocarbon fluids at temperatures below the onset of crystallization established [38] that, at low concentrations (below 3 wt %), loose steric aggregates are formed. At higher concentrations, dense packings of gel-like structures of significant size are formed. The concentration of paraffins and resins and their ratio affect the process of coarsening of aggregates, the pour point, and the amount of asphalt–resin–paraffin deposits [39].

Using the example of the Tuymazy oil, it was shown [38] that asphaltenes and resins with a low content of paraffin fragments, high molecular weight, and polarity are adsorbed on paraffin crystals and form a dense packing of molecules.

A similar conclusion was made about the formation of stable associative complexes with an ordered structure in a mixture of resins and paraffin hydrocarbons and a decrease in the crystallization temperature of the system [36]. As in the case of water-in-oil emulsions, asphaltenes and resins are present at the interface, and the ODS containing paraffins in the CSU can be considered as a complex system with dispersity of different dimensions. Just as in the case of water-in-oil emulsions, where the oil medium acts as a dispersion environment for dispersed water globules, the ODS with the paraffin-containing CSU has an oil dispersion medium which is inhomogeneous and can itself be considered as an ODS with a dispersed phase particle size tens and thousands of times smaller.

Figure 1d presents the following two-component model of an ODS containing a gas bubble as the SCE (gas-in-oil emulsion) which can be formed during the dissolution of gas or steam, as well as during the distillation of hydrocarbon feedstock. Gas-in-oil emulsions are characterized by very thick solvation layers, although the bubbles of the gas phase are small in size. If such an emulsion was formed by condensation, the dispersed phase (CSU cores) contains dissolved hydrocarbon vapors, and if it was formed by dispersing, the dispersed phase may contain gases insoluble in the hydrocarbon environment. The foaming capacity of gas-in-oil emulsions depends on the content of natural surfactants, such as naphthenic acids and their salts, sulfur compounds, and phenols. High-molecular-weight components, such as asphaltenes and resins, paraffins, and other poorly soluble compounds, have a stabilizing effect [3, 6, 7, 40, 41].

The particle size of the gas dispersed phase varies widely depending on the hydrocarbon composition of the dispersion environment, the nature of the gas in the CSU core, and the conditions in which the CSU is located. For a gas-in-oil emulsion such as fuel oil containing dissolved hydrogen sulfide, the photocolorimetrically determined particle size of the dispersed phase ranges from 350 to 600 nm [42]. However, the dispersity of heavy fuel oil after degassing increases significantly, while the average particle size of the dispersed phase decreases several times.

Figure 1e presents a two-component ODS containing a CSU in the form of mechanical impurities—solid inclusions of inorganic nature. The sources of mechanical impurities are sand, clay, fine iron particles, rust, salts, and dust, which primarily enter the oil during production, storage, transportation, and refining. At the interface between the solid particle and the oil dispersion environment, the same forces determining the surface energy excess act leading to surfactant adsorption. The mechanisms of adsorption of asphaltenes at the interfaces between oil and silica-based nanofluid and oil and wetted sandstone were investigated [43]. The results of packet adsorption showed the emergence of a strong affinity of the silica nanofluid film. Thus, in this case, as in the previous ones, ODS is a complex system in which the particle of mechanical impurities is the core of the CSU with asphaltene shells, and the hydrocarbon phase is the dispersion medium. This hydrocarbon phase, in turn, is an ODS with higher dispersity, in which the particle size of the dispersed phase can vary by tens or hundreds of times depending on the oil composition, the nature of the impurities, and its extraction methods. For crude oil, the size of mechanical impurities varies from tens of nanometers to several millimeters.

Redistribution of CSU components between layers and dispersion medium

Another feature of ODSs is the redistribution of SCU components between layers and the dispersion medium under external influences [3, 6–8].

According to Z.I. Sunyaev's theory of controlled phase transitions, the geometric dimensions of the CSU and, accordingly, the physicochemical and technological properties of the ODS change under the combined action of external factors. As the core size changes, hydrocarbons are continuously redistributed between the macrophases, i.e., between the dispersed phase and the dispersion environment. Moreover, the manifestations of this process undergo extreme or multiextremal changes [3].

It is precisely because of phase transitions in ODS that many of their properties are not additive ("anomalous"). The viscosity of mixed petroleum products is known not to be additive, since the mixing of petroleum products is already an external influence which alters the balance of intermolecular forces in the system, leading to a redistribution of components between the dispersed phase and the dispersion environment. Moreover, a study of the viscosity of oil from the Ashalchinskoye field and the light fractions, vacuum distillates, and residue isolated from it established the absence of additivity even in cases where the components of the original ODS were first separated by distillation and then combined again.

When adding 5 vol % gasoline fraction to the oil distillate, we observed nonadditive changes in kinematic viscosity. As expected, the kinematic viscosity decreased, but not according to the additivity rule. This parameter was determined at two temperatures, 40 and 100°C, and the results showed inconsistent changes. For example, an increase in temperature decreases the viscosity of the original oil distillate by a factor of 6.9, while for the oil with gasoline additive, it decreased by a factor of 5.7. The activation energy for viscous flow of the oil with the additive was 13% lower than that of the original oil. A possible explanation is that, when the oil is diluted with gasoline, some of the solvation shells of the dispersed phase are transferred to the dispersion environment. Therefore, upon heating, the ability of such a system to further remove components from the CSU decreases when compared to the original ODS.

As already mentioned, the ability to control phase transformations in ODSs by changing the balance of intermolecular interaction forces and controlling the dispersity of ODSs allows the desired values of the macroproperties of hydrocarbon raw materials for efficient processing to be achieved, as well as those of petroleum products for improving their quality [3, 6, 7].

Control of phase transitions and dispersity of ODSs

Another way to control the dispersity of ODSs is the introduction of solvents, additives, and dopants to hydrocarbon raw materials and petroleum products, and compounding to obtain specified dispersion properties that determine optimal physicochemical characteristics [3–8].

E.A. Chernysheva developed an index I_{hc} of hydrocarbon composition of oils: the ratio of the content of paraffinic hydrocarbons to the content of aromatic hydrocarbons and to the sum of the contents of resins and asphaltenes. This index is proposed as one of the criteria for assessing the stability of ODSs. It can be used in a set of parameters for assessing the stability and compatibility of oils sent for processing along with data on the group hydrocarbon composition, the stability factor indicator, and the ratio of physicochemical parameters of oil, reflecting the structural features of the oil system [5].

Another way to control phase transitions in ODSs is by wave actions. These include ultrasonic and electromagnetic actions of medium and low intensity. The effects of magnetic fields and ultrasound on oil, petroleum products, and hydrocarbons were initially studied in the middle of the last century [45–48], and the number of such studies is currently growing. Of significant interest are low-intensity wave actions (LWAs). These include ultrasonic waves with a frequency of 20–100 kHz and an intensity of up to 0.4 W/cm^2 and a constant magnetic field with an induction of less than 0.4 T.

The advantages of LWAs are low energy consumption, low environmental risks, technical simplicity, and low capital costs. The energy of this type of wave actions is typically much lower than the bond energy in hydrocarbons and heteroatomic compounds of ODSs. This determines the range of their effect on the oil system, namely, on intermolecular interactions and phase transitions between the dispersed phase and the dispersion environment [3, 6, 11].

The ultrasonic action on oil systems has been explored in Russia and abroad [34, 47–49]. For many years, scientists at the Altai State University, under the direction of V.N. Khmelev, have been studying the effects of ultrasound on various phenomena and technological processes, as well as developing and creating ultrasonic devices [49–51].

At the Institute of Petroleum Chemistry, Siberian Branch, Russian Academy of Sciences, the effect of ultrasonic vibrations was experimentally and theoretically investigated under the direction of N.V. Yudina [14, 15, 36, 52, 53]. At this institute, the effect of magnetic fields on oil, petroleum products, and their components was also explored [54–57]. Scientists from the Ufa University of Science and Technology, headed by S.R. Alimbekova, proved that descaling of the surface of well equipment under the action of a constant magnetic field and an electromagnetic field is caused by the inhibition of the process of nucleation and growth of crystals, as well as by changes in their dispersity and polymorphism [58, 59].

Since 1995, a scientific school for exploring the effect of LWAs on oil systems in dynamic mode (on a liquid flow) has been formed at the Astrakhan State Technical University under the research leadership of the author of the article. The treatment with a constant magnetic field in dynamic mode was carried out in the range of induction of 0.1–0.4 T, and flow velocity in the active gap from 0.08 to 0.3 m/s. Ultrasound acting on the liquid flow had a frequency of 20–50 kHz and a power of 0.23–0.33 W/cm^3. When testing various hydrocarbon raw materials, petroleum products, used oils, and oil waste, practical recommendations were obtained for the adaptation and implementation of the results in industry [11, 22, 30, 42, 46, 60–67].

During atmospheric and vacuum distillation of stable gas condensate, crude oil, and heavy fuel oil, all pretreated in a constant magnetic field, the yield of light fractions increased by 1–6 wt %. During visbreaking of magnetically pretreated heavy fuel oil, the coke and gas formation decreased by a factor of 1.2–2.3. The yield of light petroleum products increased by 4–8 wt %. Low-energy treatment of water-in-oil emulsion showed that the degree of its separation increased by 20–30%. The degree of removal of mechanical impurities from LWA-pretreated gas condensate increased by a factor of 15. A significant, more than 80%, reduction in deposits from model mixtures on the heat exchange surface was also observed [11, 60, 61].

The purity reached 86% [42] after LWA during degassing of heavy fuel oil to remove hydrogen sulfide. The quality of LWA-pretreated diesel and marine fuel was improved, the fuel consumption decreased by 5%, and the carbon monoxide emissions in exhaust gases were lower [62, 63]. A positive effect of a magnetic field on the uniformity of the gas distribution in the adsorber for drying natural gas and an increase in the service life of zeolite were discovered [64]. The possibility of intensifying the selective purification of oils and the extraction of undesirable components from oil feedstock with *N*-methylpyrrolidone was demonstrated [65].

The treatment efficiency of magnetically pretreated oil-containing wastewater increased by a factor of 1.3–5.7 [66]. Laboratory and industrial tests of magnetic treatment of an amine solution showed a decrease in the foaming capacity of the absorbent, a decrease in foam stability, and more effective removal of mechanical impurities during filtration [67].

LWAs on a ODS lead to a change in its dispersity depending on the component composition of the system. If the ODS consists almost exclusively of hydrocarbons and various organic heteroatomic compounds, then a phase transition from the dispersed phase to the dispersion medium occurs, resulting in homogenization of the system. If the ODS is a more complex multiphase or polydisperse system, then the wave action influences different levels: homogenization in the hydrocarbon medium and the opposite effect for the coarse dispersed phase (water and gas emulsions, suspensions).

The effect of low-intensity and low-frequency ultrasound is exerted in both stationary and dynamic modes and leads to a change in the balance of intermolecular forces which organize the ODS. Even on prolonged exposure, a constant magnetic field of low induction under stationary conditions has virtually no effect. However, applying a magnetic field to a flow causes a noticeable change in its paramagnetic, dispersion, and physicochemical properties. In this case, it is possible that a synergistic effect of the energy of liquid motion and its structuring in a magnetic field is manifested. It was assumed [68] that shear stresses during liquid flow affect the viscosity of oil, the rate of paraffin deposit growth, and the rate of hardness salt deposit growth. They are a consequence of changes in the colloidal components of these systems, while the destruction of aggregates occurs even at low liquid velocity gradients, corresponding to low shear stresses.

We observed a 1.1–1.2-fold change in the number of PMCs in heavy fuel oil and its blends with tar when passing them at different flow rates through a setup consisting of a pump and transport tubes passing through a magnetic tunnel disconnected from the grid. When a magnetic field was applied, the number of PMCs increased 1.2–1.5 times [22]. Similar changes were observed for the dispersity of petroleum products of different compositions: diesel fraction, vacuum distillate, and heavy fuel oil [30].

Constant magnetic field and ODSs

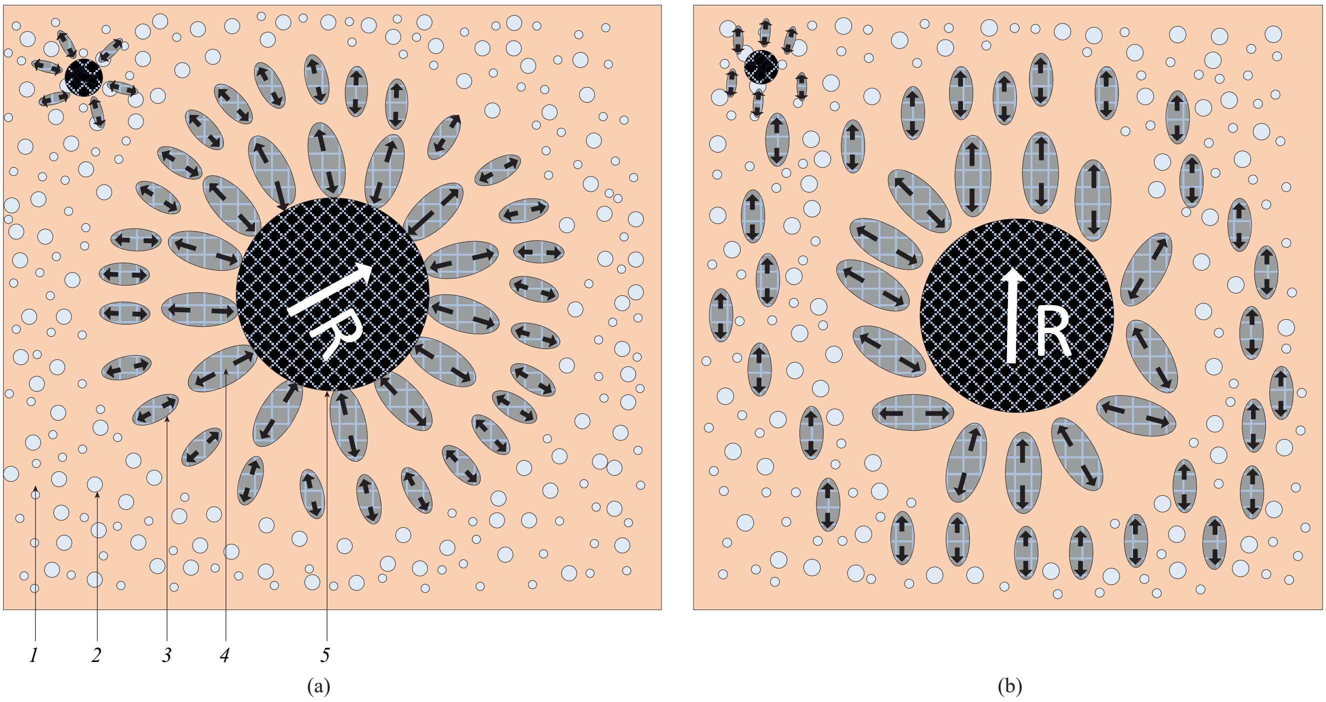
The mechanism of action of a constant magnetic field on an ODS consisting primarily of hydrocarbons and various organic heteroatomic compounds, some of which have spin properties, is as follows. Before exposure to a magnetic field, this system exhibits chaotically arranged spin dipoles, spin-containing particles and compactly organized CSUs, conventionally shown as two-layered (Fig. 3a). Under the action of a magnetic field, the paramagnetic nuclei are oriented along the field vector. During this movement, the shells are deformed, they move away from the core, partially detach, and pass into the dispersion medium (Fig. 3b). This leads to a decrease in the CSU size, i.e., an increase in the dispersity of the system as a whole and an enrichment of the dispersion medium.

For more complex multiphase, polydisperse, multicomponent oil systems, the basic two-component models of which were discussed above, the magnetic field affects both the coarse dispersed phase and the inhomogeneous dispersion medium.

As already mentioned, in almost all ODSs, surfactants containing PMCs and associated shells are present at the interface.

The interlayer of paramagnetic molecules is also subject to the magnetic field, causing the molecules to align along the magnetic induction vector. It also deforms the interlayer, and detaches it, transferring into the dispersion oil environment. As a result, the surface of the dispersed particle is exposed and becomes accessible for coagulation or coalescence. This phenomenon facilitates the removal of mechanical impurities, the separation of emulsions, or the degassing of the liquid.

The combination of wave action and chemical methods increases the efficiency of technological processes [11, 42, 57, 60, 61, 65, 69–72]. For example, during the degassing of heavy fuel oil to remove hydrogen sulfide, the purity increases significantly under the sequential action of ultrasound and a constant magnetic field on the flow. Such treatment increases



(a)

(b)

Fig. 3. Changes in the structure of an oil disperse system consisting of hydrocarbons and various organic heteroatomic compounds under the action of a constant magnetic field: (a) before and (b) after magnetic treatment. (*1*) Inhomogeneous neutral dispersion medium; (*2*) components of the dispersion environment from neutral molecules with compensated spins; (*3*) spin-polarized molecules with low paramagnetism; (*4*) spin-polarized molecules with increased paramagnetism; (*5*) the core consisting of spin-containing molecules with high paramagnetism

the yield of light fractions during the distillation of hydrocarbon feedstock, improves the separation of water and mechanical impurities from oil and gas condensate, and reduces the formation of deposits on heat exchange surfaces [11, 42, 60, 61].

Combined treatment has a more intensive effect on the viscosity and low-temperature properties of oil and the behavior of its components. The combination of wave action with reagent treatment increases the efficiency of oil preparation for refining. It decreases viscosity, and reduces the consumption of a demulsifier during oil dehydration, a hydrogen sulfide neutralizer during heavy fuel oil degassing, etc. [11, 57, 61, 65, 69–72].

CONCLUSIONS

An analysis of accumulated knowledge about the characteristics of ODSs shows that they are characterized by multiplicity of phases, as well as a diversity of hydrocarbon components and heteroatomic compounds, mineral impurities, and trace elements. Intermolecular interactions in ODSs are dominated by exchange spin-spin interactions caused by the paramagnetism of resins and asphaltenes, as well as the homolytic dissociation of high-molecular and heteroatomic organic compounds. Changes in the conditions of ODS existence and external influences lead to the redistribution of ODS components between the layers and the dispersion environment. Using two-component models of ODS shells, their interaction with the dispersion environment is explained, providing a key to understanding the possibility of controlling the structure, dispersity, stability, and other properties of oil systems.

Considering the specific features of ODSs is key to the efficient and optimal processing of hydrocarbon feedstock and the upgrading of petroleum products. Effective tools for controlling the dispersed state of oil systems include treatment in a constant magnetic field (magnetic induction no higher than 0.4 T), or ultrasonic treatment (20–50 kHz, 0.23–0.33 W/cm^3). These wave actions are low-energy, and the equipment required does not necessitate significant capital or operating costs. As demonstrated above, LWAs considerably improve the technological and, consequently, economic performance of processes, promoting rational use of hydrocarbon feedstock and petroleum products.

The author declares no conflict of interest.

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About the Author

Nadezhda A. Pivovarova, Dr. Sci. (Eng.), Professor, Department of Chemical Technology of Oil and Gas Processing, Astrakhan State Technical University (16/1, Tatishcheva ul., Astrakhan urban district, Astrakhan, 414056, Russia). Email: nadpivov@mail.ru. Scopus Author ID 7003930664, RSCI SPIN-code 5096-5935, https://orcid.org/0000-0001-5486-3141

Об авторе

Пивоварова Надежда Анатольевна, д.т.н., профессор кафедры «Химическая технология переработки нефти и газа», ФГБОУ ВО «Астраханский государственный технический университет» (414056, Россия, Астраханская область, городской округ город Астрахань, г. Астрахань, ул. Татищева, стр. 16/1). E-mail: nadpivov@mail.ru. Scopus Author ID 7003930664, SPIN-код РИНЦ 5096-5935, https://orcid.org/0000-0001-5486-3141

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