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Physicochemical Aspects of Primary Oil Processing Technology (Review)

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Abstract—The review presents theoretical concepts of petroleum disperse systems, options for controlling phase transitions, balances of intermolecular interaction forces, and means to achieve the active state of feedstock through various kinds of its treatment so as practical ways to optimize technological processes can be found. These principles are described in relation to the primary processing of crude oil, namely, the demulsification and straight-run distillation processes.

Keywords: petroleum disperse systems, intermolecular interactions, physicochemical technology, active (extreme) state of feedstock, straight-run distillation of crude oil

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Enhancement of the efficiency of technological processes and more complete use of the potential of petroleum feedstock are important tasks of refining. It is known that the following basic methods are practiced to achieve these goals: feedstock preparation, creation of new catalysts and improvement of their properties, and perfection of the engineering design of processes. One of the ways for preparing feedstock is the regulation of its disperse structure using various external factors.

It is known that crude oil is a complex multicomponent and multiphase mixture consisting of more than 1000 different hydrocarbons (HCs) and other compounds. The set of their chemical properties, structure, and intermolecular interaction forces in combination with certain external conditions determine physicochemical properties that uniquely characterize each oil sample. Although long-term occurrence of oil in the reservoir under static conditions or prolonged storage allows it to achieve a balanced stable state, a slightest external influence (temperature, pressure, blending with various components and additives) alters this balance. It has been established that the restructuring of the system begins in the microworld from the nanoscale level of individual molecules and a change in the balance of forces between the molecules and their rearrangement cause a change in the properties of the oil. A targeted influence on intermolecular contacts, which is one of the principles of the currently popular nanotechnology, makes it possible to control the properties of systems within the required boundaries with minimal energy costs.

A characteristic feature of petroleum systems is their microdispersity or, rather, their polymicrodispersity. The nanometer (colloidal) dimensions of the dispersed particles of petroleum systems (1–100 nm) result in a developed surface area, so that a great role is played not only by forces acting in the bulk, but also by surface phenomena at the boundaries of macromolecules and supramolecular structures.

These phenomena are studied by researchers from the scientific and academic school founded by Professor Z.I. Syunyaev in the 1970s at the Department of Oil Refining Technology of the Gubkin State University of Oil and Gas. Scientific ideas that formed the basis of the school are, first of all, the ideas of Academician P.A. Rehbinder, who is the founder of physicochemical mechanics as a new field of science [1]. He introduced the concept of surface activity as a rigorous thermodynamic characteristic of a substance into science.

Syunyaev proposed concepts and terms, such as petroleum disperse system (PDS), complex structural unit (CSU), extreme (active) state of feedstock, and physicochemical technology of oil refining, that are currently accepted by the scientific community and are widely used in scientific publications [2].

The physicochemical technology of crude oil processing is based on the theory of controlled phase transitions. According to this theory, technological processes and the quality of petroleum products can be controlled by affecting the kinetics of nucleation and development of a new phase and phase transitions during the preparation, transportation, and processing of petroleum stocks, and also in the preparation of marketable crude oil blends [3, 4].

The basis of the physicochemical technology is the principle formulated by Rehbinder: "The task of the new field of knowledge that unites a number of problems of rheology, colloid chemistry, molecular physics, the mechanics of materials, and the technology of their production primarily consists in establishing the mechanism and features of the processes of formation, deformation, and degradation of structures with the aim to control these processes" [1]. It is the ability to control phase transformations in petroleum systems and to affect the balance of intermolecular interaction (IMI) forces, the size of complex structural units and ultimately the basic macroproperties of PDS (resistance to phase separation, structural and mechanical strength, etc.), and the yield and quality of petroleum products that forms the basis of the physicochemical technology of oil refining.

BLENDING AS FEEDSTOCK PREPARATION METHOD

Preparation of feedstock for various technological processes is an important stage in the perfection of any technological process. There are different methods of feedstock preparation. First of all, these are dehydration and desalting of crude oil, hydrotreating and hydrofining of distillate and residual stocks, deasphalting, and other processes. Their effectiveness can be improved by using the physicochemical technology principles, namely, by controlling the balance of intermolecular interaction forces and phase transitions and promoting feedstock transition to an active state by various actions: introducing additives or using magnetic and electric fields, ultrasound, and mechanical or other effects [5–10].

The simplest and most affordable way to achieve the active state of PDS is blending crude oils and feedstock streams. At the same time, a nonadditive character of the properties of a blend should be taken into account so that to determine the most favorable (optimal) ratio of the components, which ensures their compatibility and best performance in the transportation, processing, storage, and creation of phase separation resistant commercial petroleum products [2]. Mixing does not give an arithmetic sum of the properties of the individual components, because they are independent disperse systems, and blending results in a new system with inherent physical intermolecular interactions and the corresponding balance of forces of these interactions.

Physical interactions exist in any petroleum system and are especially noticeable between molecules of macromolecular compounds even at ordinary (ambient) temperatures. As a result of these interactions, supramolecular structures composed of tens and hundreds of molecules depending on the conditions (temperature, pressure, presence of natural surfactants, etc.) are formed. Due to excess surface energy, these supermolecular entities (nuclei) are surrounded by an adsorption–solvation layer and, together with the nucleus, form a complex structural unit [2, 11]. Together they are the so-called "soft systems" [12].

External influence on the petroleum system is especially evident already at the first stages of primary crude oil treatment. It is known that the main process in refineries is dehydration and desalting of crude oil in an electric desalter. The hydrophobic disperse system in a desalter is subjected to the combined influence of several factors: temperature, pressure, electric field, wash water circulation, introduction of surfactants/demulsifiers, and alkalization. The optimal combination of these factors ensures the separation of the system into two phases (water–oil) with a flat interface [13–15]. In the case of addition of different demulsifiers, either enhancement or antagonism of their effects is possible. The maximum of the synergistic effect can be found by measuring the dielectric properties to reveal the highest value of the parameter $\varepsilon_1\varepsilon_2 = (\varepsilon_1)^2 \tan\delta_{\max}$ [16].

The physical interactions of certain types of molecules and compounds are clearly manifested at temperatures close to the pour points of a given petroleum system. However, the dispersion is also varied under high-temperature conditions of various kinds of treatment: distillation, cracking, and coking.

Polydispersity of crude oils (petroleum disperse systems) is the reason behind the nonlinearity of the change in their properties, which together with the multicomponent nature and homological proximity of the constituent components determines their high sensitivity to external factors. One of the impacting factors can be mixing with other crude oils and components. The mixing can be both controlled and spontaneous.

On the basis of a large experimental material it was shown that blends of crude oils differing in fractional and chemical composition, crude oil blends with gas condensates, and blends of gas oil fractions show both a nonadditive change in properties, depending on the ratio of components, and nonlinear behavior in straight distillation, catalytic cracking, coking, and other processes [17–19]. For example, in the case of compounding Yarega heavy oil with Usa paraffin-base oil, the viscosity of the blends change in an abnormal manner [10, 20, 21], which affects the pumping efficiency of the oil blends. Examples of analysis and calculation of the viscosity of blended oils are presented in [22–26].

Despite the fact that the structure of petroleum systems from the standpoint of modern theoretical concepts is described mainly at the level of qualitative changes, a wealth of empirical results obtained to date allows interpretation some of the phenomena observed in them. It is known that the blendingcaused nonlinearity of the change in properties is most pronounced for components that substantially differ in properties and for surfactant-containing products. An increase in the number of components by blending can lead to an increase in the number of extremums on the curves of the change in properties and a decrease in their intensity, i.e. the deviation of the properties of the system from the additive behavior will be reduced.

Chemical thermodynamics teaches that the general behavior of a system is determined by Gibbs free energy, which obeys the following equation for an ideal binary system under atmospheric pressure [17]:

$$G_{\mathrm{p}} = x'G_1^0 + RTx'\ln x' + (1 - x')G_2^0 + RT(1 - x')\ln(1 - x'), \quad (1)$$

where G_{p} is the free energy of 1 mole of an ideal binary mixture; G_1^0 and G_2^0 are the free energies under standard conditions (1 atm, 20°C) for the first and second components of the mixture, respectively; x' and $(1 - x')$ are the mole fractions of the first and second components, respectively; and $x'\ln x'$ and $(1 - x')\ln(1 - x')$ are respectively the concentration contributions to the Gibbs energy of the first and second components.

An analysis of this equation shows that the free energy, depending on the concentration of the components, changes nonmonotonically and has one minimum at a certain concentration of ideal substances, where the system is in an equilibrium and thermodynamically stable state.

Real petroleum systems are characterized by complex intermolecular interactions. Unger [27] supposed that they are based on the interaction between paramagnetic and spin-polarized molecules. It is difficult to mathematically describe such systems. Therefore, it should be emphasized that Eq. (1) can be used to describe only some cases of the behavior of a binary system. It is commonly known [28] that Gilbert N. Lewis proposed to replace $\ln x'$ and $\ln(1 - x')$ in Eq. (1) by $\ln\alpha_1$ and $\ln\alpha_2$, where α_1 and α_2 are the activities of the components, which are the products of the concentration multiplied by activity coefficients γ_1 and γ_2 characterizing the IMI in the system: $\alpha_1 = x\gamma_1$ and $\alpha_2 = (1 - x)\gamma_2$.

If proceeding from Eq. (1), the total free energy of a real system, G_{p}, can be written as follows according to these ideas [17]:

$$G_{\mathrm{p}} = xG_1^0 + RTx\ln\alpha_1 + (1 - x)G_2^0 + RT(1 - x)\ln\alpha_2. \quad (2)$$

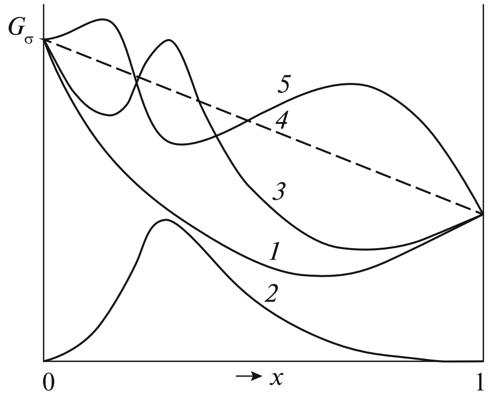

Fig. 1. Dependence of Gibbs free energy (G) on the concentration of a substance (x) in a binary mixture [17]: (*1*) free energy of the ideal mixture, (*2*) the free energy correction for the interaction between the unlike molecules of the two components, (*3*) the total free energy of the real binary mixture, (*4*) the additive mixing function of the binary mixture, and (*5*) increment or decrement in the free energy of the real binary mixture relative to energy of mixing.

Substituting for the activities α_1 and α_2 in Eq. (2), we obtain:

$$G_{\mathrm{p}} = xG_1^0 + RTx\ln x + (1 - x)G_2^0 + RT(1 - x)\ln(1 - x) + RTx\ln\gamma_1 + RT(1 - x)\ln\gamma_2. \quad (3)$$

Analysis of Eq. (3) shows that the value of free energy varies nonmonotonically with a few extremums.

Thus, it has been already noted that the first four terms of this equation correspond to the description of the Gibbs free energy of an ideal system. The last two terms take account of an additional contribution to the free energy, namely, the contribution of intermolecular interactions characteristic of real systems, i.e. real mixtures of petroleum products.

This component $\Delta G = RTx\ln\gamma_1 + RT(1 - x)\ln\gamma_2$ is often called additional or "excess" energy. This energy, in particular, explains the so-called synergistic effect as well.

Figure 1 shows the dependence of free energy on the concentration of one of the components in a binary mixture for ideal (Eq. (1)) and real (Eq. (2)) systems.

If the activity coefficient of an individual compound in the standard state is taken to be unity, i.e. assuming that there is no intermolecular interaction of dissimilar components, the "excess" free energy will be zero. That is, $\Delta G = 0$ for $x = 0$ or $x = 1$. According to Raoult's law, a function that passes twice through zero values has at least one maximum or minimum, i.e. takes a nonmonotonic form (curve *2*).

By physical meaning, Gibbs free energy has a certain absolute value; i.e., a function that maps intermolecular interaction energy passes through a maximum.

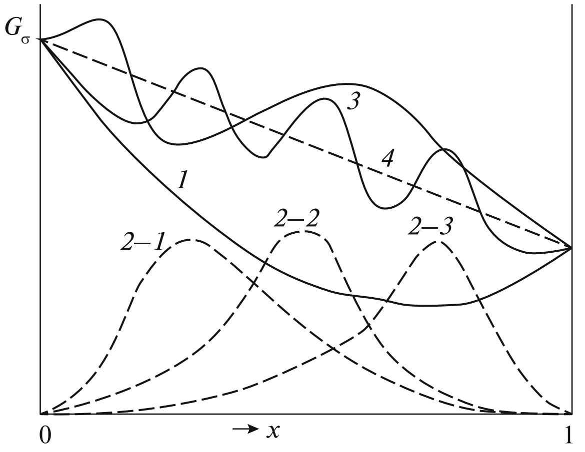

Fig. 2. Dependence of Gibbs free energy on the concentration of components in a ternary mixture [17]. (*1*) Free energy of the ideal mixture, (*2-1*, *2-2*, *2-3*) free energy corrections for the interactions between molecules of the pseudo-component with the third component, (*3*) is the total free energy of the real ternary mixture, and (*4*) the additive mixing function of the ternary mixture.

Therefore, the dependence of the free energy of the mixture on the concentration of components in it has a few extremums; i.e., the curve of change in the real free energy of a real binary mixture will have two minimums and one maximum (curve *3*). In any case, there will be a deviation from the additivity law shown by straight line *4*. The difference between the energies described by line *4* and curve *3* is the excess of the free energy over the equilibrium values (curve *5*).

In accordance with this, the physicochemical properties of such a mixture (viscosity, pour point, boiling point, resistance to phase separation, etc.) also vary nonmonotonically exhibiting several extremums.

Figure 2 shows the dependence of the free energy for a ternary mixture, in which a mixture of any two components is taken as one pseudo-component. The curve of change in the free energy and the physicochemical properties of such a system with the concentration of the third component has three maxima and four minima (curve *3*). The nonlinearity of the change in properties in the case of mixed components will be manifested to the greatest extent for the components that differ in chemical composition and in the concentration of surfactants that significantly change the intermolecular interaction within the component, as well as in the case of energetic impacts (electric or magnetic field, ultrasound, etc.) on the system.

Thus, when different petroleum products, for example, fuel oil and cracked residue are mixed, the nonlinearity of the change in the physicochemical characteristics of the blend will manifest itself to a greater extent than in the case of blending heavy fuel oil with another fuel oil, especially obtained from crude oils of similar compositions.

As the number of components in the blend increases, the number of extremum points on the curves of change in their properties increases. At the same time, the greater the number of components used to make a blend, the less is the deviation of the blend characteristics from the relevant additive values. In this regard, the traditional use of linear equations and additivity rules in technological calculations is justified to some extent. Nonetheless, when assessing blends of petroleum products that have certain individual properties, it is necessary to be guided by the above-discussed ideas that the properties of the petroleum system as a whole change as a polyextremum function and, in engineering practice, it is necessary to take into account a nonlinear character of changes in the properties for blends of crude petroleums and petroleum products differing in chemical composition.

SYNERGISTIC EFFECTS IN PETROLEUM SYSTEMS

The above-discussed physical (energetic) nature of the nonlinear change in the properties of real complex systems, depending on the mixture composition, can also help to reveal the physical essence of the so-called synergistic effect observed when petroleum systems are subjected to treatment. The effect is due to the excess of free energy over equilibrium values, which results in an increased activity of such mixtures in comparison with the additive value (see curve *5* in Fig. 1).

It was found that synergism manifests itself to a greater extent with unequal amounts of components. For example, in the case of distillation of a 30 : 70 mixture of paraffinic and aromatic crude oils, the yield of light fractions (up to 350°C) is higher by 3–5 wt % than the arithmetic average calculated according to the additivity rule [3], whereas the deviation is less for 50 : 50 mixtures.

If one of the components has a significant activity by its nature (for example, it is a surfactant), the synergistic effect will be manifested at low concentrations of this component (hundredths, thousandths of a percent).

Theoretical conclusions on the nonlinearity of changes in the properties of petroleum disperse systems are confirmed by practice. Oil blending can be considered as a process of changing the solvent composition with respect to the asphaltene portion with the establishment of a dynamic equilibrium between the possible reactions of formation of radicals and their recombination, which is manifested in a change in the paramagnetism of the system. As the group composition of blends changes, some amount of hydrocarbons is either released or retained as a result of a nonlinear change in the balance of intermolecular interaction forces. Thus, the group composition, as

Table 1. Change in the asphaltene stability index (ASI) and the yield of distillate fractions of surfactant-doped crude oil

Progalite concentration, wt %	ASI	Yield of fraction, wt %					
		IBP–180°C	180–240°C	240–350°C	∑IBP–350°C	350–500°C	resid >500°C
0	0.85	17.1	11.9	13.5	42.5	25.1	32.4
5×10^{-2}	0.93	17.3	11.6	14.8	43.7	24.2	32.1
5×10^{-3}	0.96	17.1	11.7	17.4	46.2	21.4	32.4
5×10^{-4}	0.88	17.2	11.0	16.6	44.8	22.3	32.9

well as paramagnetism and the yield of light fractions in the distillation of petroleum compositions, vary as a polyextremum function depending on the ratio of the components in the mixture [11].

The synergism and antagonism effects should be taken into account when preparing and processing blended feedstocks.

The boiling (evaporation) processes are significantly affected by surface phenomena [29]. It was found that the introduction of a surfactant (Progalite) in an optimal concentration into crude oil leads to an increase in the kinetic stability of the system and, accordingly, an increase in the recovery of light distillates by distillation (Table 1) because of the redistribution of hydrocarbons between the coexisting phases. The optimum Progalite concentration in this case is $5 \times 10^{-3}\%$. The recovery of light fractions increases in comparison with the initial oil from 42.5 to 46.2% [29], that is, by 3.7%, a value that exceeds the precision of the measurement method (2%). The existence of relation between the stability characteristics and the yield of fractions confirms that they are due to the same cause, the change in the balance of intermolecular interaction forces in the system.

It was shown in [30] that the mixing of model systems (toluene–hexane and α-methylnaphthalene–hexadecane), fuel fractions, crude oils, or oil residues is characterized by a change in surface properties and, as a consequence, a change in the fractional composition of petroleum systems upon distillation in comparison with calculated, expected additive values.

Chigareva et al. [31] showed that in the distillation of petroleum systems, the dependence of the vapor bubble diameter (d) on the composition of a mixture is closely related to the distribution of components between the coexisting phases and the surface layer. They analyzed the mechanism of the influence of the mixture composition on the vapor bubble diameter through interfacial tension σ at the phase boundaries. The average vapor-bubble departure diameter varies in accordance with a change in the σ value.

The statistical processing of cine records of boiling of a water–n-butyl alcohol or a water–isoamyl alcohol binary mixture used as an example showed a nonmonotonic change in d with the alcohol concentration.

The most affordable and often unavoidable way to affect the state of PDS is to mix crude oils and feedstock components. Note that the basis for any kind of treatment of a petroleum system is the principle of regulating phase transitions by varying the balance of IMI forces and affecting the particle size of the dispersed phase. The theoretical expression of this postulate is the Kelvin–Thomson equation [32]. It can be assumed that by dispersing the system via extremely reducing the radius of the CSU nucleus, it is also possible to affect the vapor bubble size during boiling, namely, to decrease this radius and thereby intensify the liquid–vapor phase transition process.

The Kelvin–Thomson equation relates the partial pressure over a flat and curved surface, in particular, to a droplet radius r characterizing the degree of curvature (the smaller the value of r, the greater the surface curvature):

$$RT\ln\left(\frac{p_r}{p_0}\right) = (2\sigma V)/r,$$

R is the universal gas constant; T is the absolute temperature; V is the partial molar volume; and p_r and p_0 are the partial vapor pressure above the droplet of radius r and the planar surface, respectively.

For example, for a droplet of water with $r = 10^{-5}$ cm (σ = 73 dyne/cm, V = 18 cm^3/mol), the $\frac{p_r}{p_0}$ ratio is 0.01; i.e., the pressure increases by 1%; and for a droplet of $r = 10^{-6}$ cm, $\frac{p_r}{p_0} = 1.11$; i.e., p_r increases already by 11%.

Thus, by altering the size of the disperse particles of a petroleum disperse system, it is possible to affect the results of its refining, in particular, straight distillation.

Mixing of oils and components can have both desirable and undesirable consequences. Although some of the physicochemical properties of the system can vary according to the additivity rule, most of the parameters vary nonlinearly and in a polyextremum manner. This concerns viscosity, rheological properties, resistance to phase separation, the potential recovery of distillates by distillation, etc.

A vivid example of the undesirable consequences is the problems of incompatibility of crude oils upon

Table 2. Results of thermogravimetric study of fuel oils and their optimal mixture

Sample	Endothermic peak area, mm^2	Amount of evaporated substance, 10^{-3}, kg	Onset evaporation temperature, K	Enthalpy of vaporization, kJ/kg
M-1	900	0.10	513	1386.6
M-2	850	0.10	503	1439.6
Optimal blend M-3	1000	0.12	487	1290.5

mixing. They can appear in the form of "diminution of volume by mixing" (as large as several percent) or, in case of significant incompatibility, in the form of phase separation in the system (stratification)—precipitation of asphaltene–resin–wax (ARW) deposits. With the exception of processes based on the phase separation (electric desalting, deasphalting, dewaxing, separation, etc.), this is highly undesirable or even dangerous for production. The consequence is the uncontrolled formation of a poorly soluble deposit layer during the transportation of oils in tankers, trouble in the operation of an electric desalter, clogging of transmission pipelines, fouling in heat exchangers of crude preheat trains and in contact devices of atmospheric–vacuum pipestills, etc. Ultimately, this can lead to unscheduled outage of equipment and to significant economic loss and environmental damage. For a large refinery, a shutdown of main high-margin units can lead to a decrease in the production rate of petroleum products by several thousand tons per day, and, accordingly, to losses of several million dollars [33]. The problem of the incompatibility of crude oils and oil products and the stability of their blends has been explored by many investigators [34–43].

From the point of view of colloid chemistry, the manifestation of incompatibility indicates that the dispersed structure of a petroleum system is in a suboptimal state. In this state, the feedstock potential cannot be fully realized, the recovery of valuable fractions by distillation is lower, the quality of the distillates is worse because of insufficient separation, the refining depth is smaller, and there are other negative consequences. In that case, part of the light distillates (up to 7–8%) is entrained in dark products and part of vacuum fractions (up to 10–12%) remains in the tower bottoms [2, 11].

For proper blending of crude oil and gas condensate, it was proposed [44, 45] to reconcile the physicochemical characteristics of target fractions with the standardized properties of commercial petroleum products. This problem in the cited study was solved by a computational method using characteristics of the desired fractions, such as density, viscosity, and pour point, that most fully determine the additive, rheological, and low-temperature properties of commercial petroleum products.

To estimate the IMI energy by distillation in the case of mixing feedstock components, the initial samples of atmospheric residues of two crude oils differing in composition (Usa paraffin-based oil and aromatic, resinous West Siberian oil) were studied by the thermogravimetric method [46]. These residues (fuel oils M-1 and M-2, respectively, as well as fuel oil obtained from a 30 : 70 blend of the crude oils (sample M-3)) subjected to vacuum distillation showed an increase in yield of distillates by 7% relative to the calculated yield. Examination of these samples on a differential thermal analyzer (Table 2) made it possible to reveal the cause of this phenomenon.

With constant heat input, the increase in the yield of the vapor phase during the distillation of the modified feedstock can be explained by a decrease in the activation energy of the molecules to overcome the boundary potential barrier. The most direct and reliable way to estimate the intermolecular interaction energy is to calculate it from the enthalpy of vaporization (1290.5 kJ/kg vs 1386.6 and 1439.5 kJ/kg for the initial components). The blend M-3 (with the optimum composition of 30 : 70) is characterized by a minimum value of the phase transition enthalpy, and the onset evaporation temperature of the same blend is lower by 26 and 16 K than that of M-1 and M-2, respectively (see Table 2).

In summary, it should be pointed out that the parameters of the system have a significant effect on the occurrence and results of processes, since most of the petroleum refining processes are accompanied by phase transitions. The availability of methods for timely recognition of the nucleating dispersed-phase particles, the kinetics of their development, and understanding of their behavior makes it possible to predict the properties of petroleum systems over wide ranges and also to increase the efficiency of technological processes. It is important to assess the dispersed state of feedstocks in advance for finding the optimal component ratio in the blended feedstock from the point of view of dispersion properties (and process results) and to select the intensity of feedstock treatment (additive and surfactant concentrations, intensity of wave, magnetic field, etc. impacts). Thus, the petroleum refining processes can be intensified.

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