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Effect of the Conditions of Low-Frequency Acoustic Treatment on the Stability of Oil-Water Emulsions of Oil from the Ignyalinskoye Deposit

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Abstract

The effect of the integrated action of low-frequency acoustic treatment, demulsifiers and the temperature factor on the stability and structural-mechanical properties of oil-water emulsions of paraffin low-resin oil from the Ignyalinskoye deposit is investigated. After low-frequency acoustic treatment of emulsions, water phase droplets are dispersed, and their number increases. Acoustic processing and introduction of demulsifier do not lead to the destruction of oil-water emulsions (10 mass %) stabilizing them due to an increase in the fragmentation of the dispersed phase. The integrated action is an effective method of the destruction of 50 mass % emulsions. After integrated processing, the size of the droplets of disperse phase in oil systems increases. Acoustic treatment of oil, oil-water emulsions (10 mass %) is accompanied by a quantitative increase in resin-asphaltene components. However, significant changes are observed only in the structural-group composition of asphaltenes.

Keywords: oil-water emulsions, low-frequency acoustic treatment, resin-asphaltene components

INTRODUCTION

One of the essential problems which are often encountered during oil mining and transportation is the formation of complicated and extremely stable emulsions. At present, the major oil deposits in the world enter the medium or late period of operation, and well stream watering increases. The presence of emulsified water in commercial oil causes a number of difficulties during processing: pressure restriction, decrease in productivity, pipeline corrosion, pump failure, poisoning of the catalysts at oil processing plants, etc. The presence of various sites with high shear rates or turbulence zones at the deposits leads to the formation of inverted emulsions of the water-in-oil type, with their stability depending on the nature of oil fluid and the amount of stratum water [1, 2]. The presence of natural surface-active substances in oil (asphaltenes, resins, carboxylic acids), as well as solid particles represented mainly by salts, sand, clay, iron oxides and high-molecular paraffins, promotes stabilization of water-oil emulsions. In addition, the introduction of the technologies of acid hydraulic fracturing and physicochemical methods for increased oil recovery caused such problems as higher stability and higher electric conductance of the formed wateroil emulsions [3, 4]. An increase in the watering of mined oil has also a substantial effect on the net cost of oil preparation using traditional methods and causes the need to introduce new energysaving technologies. To prevent economic losses arising during the preparation of commercial oil, it is necessary to separate water-oil emulsions into two separate phases in the process known as demulsification before oil transportation or processing.

It is established that it is difficult to separate stable emulsions completely at the field using only the technology of gravitational sedimentation in a horizontal dehydrator. Under the action of electric field, collisions and coalescence between emulsified water drops would be enhanced, thus promoting the separation of the dispersed water phase from the continuous oil phase. However, dehydration is accompanied by the problems concerning the requirements to the electrostatic dehydrator because of a frequent drop of the electric field voltage and low efficiency of demulsification [5, 6].

During the recent years, the use of electric fields with a frequency higher than 1000 Hz and high alternating voltage, as well as the action with low-frequency acoustic fields are attracting extensive attention of researchers by higher demulsification efficiency, lower consumption of electric energy, and better stability of performance characteristics during the treatment of water-in-oil emulsions [7–12].

The application of the technology of low-frequency acoustic treatment (LFAT) is a promising direction in dehydration of water-oil emulsions and is urgent due to simplicity, availability, efficiency and practicability [13].

The goal of the present work was to study the effect of the integrated action of LFAT, demulsifying agents and the temperature factor on the stability and structural-mechanical properties of water-oil emulsions in high-paraffin low-resin oil from the Ignyalinskoye deposit.

EXPERIMENTAL

The effect of LFAT duration and the temperature factor on the stability of water-oil emulsions (10 and 50 mass % of water) in low-resin high-paraffin oil from the Ignyalinskoye deposit (the Irkutsk Region) was studied by means of the Bottle Test [14]. Paraffin content is 2.0 mass %, resin content is 7.9 mass %, asphaltenes content is 0.1 mass %. Emulsions were formed by mixing oil and stratal water for 10 min at room temperature using an overhead drive mixer PE-0118 (Russia, power 150 W, rotation frequency 3000 r.p.m.).

Low-frequency acoustic treatment of oil and water-oil emulsions was carried out in a laboratory set-up which was an analogue of a commercial jet submersible vibrator VEMA-0.3, in the steady mode at room temperature [14]. The vibration system of the set-up was composed of the vibrating confuser (activator), elastic elements and the motor part; it was immersed into the liquid medium. The energy which is necessary to maintain stable reciprocating vibratory movements of the activator is imparted into the system by the magnetic field. The limiting magnetic field strength (*H*) in the air gap of the device is $H = NI/\delta_{\min} = 1000 \cdot 2/(1 \cdot 10^{-3}) = 2 \cdot 10^6 \text{ A/m}$ where *N* is the number of turns of electromagnet coil; *I* is the current in the electromagnet coil, A; δ_{\min} is the minimal air gap of the electromagnet, m.

Under the action of LFAT, the liquid system (LS) is affected by the mechanical motion of a flow of LS, shear movement between the layers of the LS, alternating stress (up to 3 atm), acoustic, electric and magnetic fields; the velocity of submerged jets up to 20 m/s, acceleration of the working activator up to 100g, the frequency of the major action 50 Hz [13, 15]. The low-frequency acoustic treatment destroys the colloid dispersed structure existing in the oil system, which leads to substantial changes in the structural-mechanical properties [16, 17].

During experiments, the samples of time-stable water-oil emulsions 200-250 mL in volume were subjected to LFAT for 0.5-10 min at the external field voltage of ~100 V. Then the treated emulsions were thermostated for 1 h at 20 and 50 °C.

RESULTS AND DISCUSSION

The viscosity-temperature dependences obtained using the SX-800 device of INPN Kristall (Russia) for the samples of initial oil and emulsion (10 mass %) after LFAT for 5 and 10 min are shown in Fig. 1.

One can see that the dynamic viscosity (η) of initial oil increases smoothly with a decrease in temperature, while for oil treated for 5 min, a sharp jump is observed below -15 °C (at -20 °C, η increases by a factor of 1.8). An increase in treatment time to 10 min, quite contrary, proceeds with some decrease in viscosity.

The behaviour of viscosity within the negative temperature range for the emulsion (10 mass %) is in many aspects similar to the change in η of oil, however, an increase in the time of LFAT causes an increase in η within the entire temperature range: after treatment for 5 min, viscosity increases insignificantly and grows sharply after LFAT for 10 min (by a factor of 1.5 at -20 °C). This may be connected with the fact that the initial period involves the destruction of



Fig. 1. Dependence of dynamic viscosity on temperature for oil of the Ignyalinskoye deposit (a) and emulsion (10 mass %) after LFAT (b).

the colloid dispersed structure existing in the emulsion, due to the dispersion of water drops and the crystal structure of paraffin hydrocarbons (PH). The transfer of PH from the dispersed phase into the dispersing medium is accompanied by a substantial decrease in shear strain and viscosity. With an increase in the time of LFAT, the solubility of PH in the dispersing medium decreases sharply, which leads to a substantial increase in viscosity. This may be due to the origination of inertial (short-range) coagulation, and due to the effect of the vibrational strengthening of the structure, which is characterized by a sharp increase in efficient viscosity [18].

Any kind of physical action on the structured oil system with the dispersed phase represented both by molecular crystals of PH and by the associates of resinous-asphaltene components leads to the transformation of crystallization-related structured and associative formations and, as a consequence, to a strong change in the structural-rheological characteristics [19–21].

The features of structure formation processes in oil systems under low-frequency acoustic action were studied with the example of the changes in the activation energy of viscous flow ($E_{a(vf)}$), which was calculated using Frenkel–Eyring equation: $\eta = A \exp (E_{a(vf)}/(RT))$

Here η is dynamic viscosity; *A* is a pre-exponential factor which includes (in the latent form) the dependence of viscosity on other structural parameters, in particular, molecular mass; $E_{a(vf)}$ is the energy barrier for the fluctuation transition (the activation energy of viscous flow); R is the universal gas constant; T is absolute temperature [22, 23].

Semi-logarithmic dependencies of η on reciprocal temperature for initial oil and for the emulsion (10 mass %) before and after LFAT are shown in Fig. 2. The viscosity – temperature dependencies for the samples under investigation look linear, without inflection points, insignificantly differing from each other in the slopes, which is manifested as different $E_{a(vf)}$ values.

Table 1 shows $E_{a(vf)}$, solidification temperature (T_s) and η at -20 °C. Solidification temperature after LFAT of initial oil for 5 and 10 min increases by 13.4 and 10.3 °C, respectively, and for emulsion – by 0.5 and 7 °C, respectively. The viscosity of oil increases by 80 and 37 %, respectively, the viscosity of the emulsion increases by 3 and 54 %, respectively. The maximal increase in $E_{a(vf)}$ by 15.7 % is observed after LFAT of oil for 5 min and by 17.7 % after LFAT of the emulsion for 10 min.

Comparing $E_{\rm a(vf)}$ values of oil and emulsion before and after treatment, one may note that the destruction of the colloid dispersed system formed during LFAT requires more energy than initial oil.

This conclusion is confirmed by the results of the experiment on the investigation of LFAT effect on the aggregative stability of water-oil emulsions with different content of the aqueous phase. The field-treated emulsions are characterized by high stability both at 20 °C and after heating to 50 °C. With an increase in treatment time from 0.5 to 5 min, the stability of emulsions with water concentration close to critical (50 mass



Fig. 2. Temperature dependence of viscosity η in Arrhenius coordinates for initial oil (a) and emulsion (10 mass %) (b) after LFAT.

%) is conserved, in spite of the presence of large water globules (Table 2, Fig. 3). However, the release of the free aqueous phase does not occur both without heating and after heating to 50 °C.

The minimal size of drops (d_{\min}) in emulsions vary within the range 1.1–3.5 µm are only weakly dependent on LFAT time and temperature, while the maximal size (d_{\max}) decreases substantially with a n increase in the time of treatment: from 67.2 µm in the initial emulsion (10 mass %) to 18– 24 µm after LFAT for 3–5 min. The low-frequency treatment of the emulsion (50 mass %) for 1 min is accompanied by a substantial increase in \mathcal{A}_{\max} by a factor of 4.7, longer LFAT, quite contrary, causes dispersing of the dispersed phase to the size equal to d_{\max} of initial emulsion (see Table 2).

The introduction of a commercial demulsifying agent (DA) of TND-A grade (Russia) before

TABLE 1

Solidification temperature $(T_{\rm s})$, dynamic viscosity (η) and the activation energy of viscous flow $(E_{\rm a(vf)})$ of oil and emulsions (10 mass %) before and after LFAT

Sample	T _s , °C	η at -20 °C, mPa·s	$E_{ m a(vf)},$ kJ/mol
Oil:	-56.0	270	39.1
LFAT for 5 min	-42.6	490	46.2
LFAT for 10 min	-45.7	370	45.1
Emulsion (10 mass %):	-52.0	350	37.7
LFAT for 5 min	-51.5	360	39.1
LFAT for 10 min	-45.0	540	44.4

or after LFAT in the concentration of 120 g/t of oil into the emulsion (10 mass %) did not cause the destruction of the oil-containing system at 20 °C and at 50 °C.

After the addition of DA into the emulsion (50 mass %), layering process starts at room temperature, with the release of the free aqueous phase, however, heating causes a substantial increase in the rate of emulsion separation and the amount of separated water (Fig. 4).

After integrated action (10 min LFAT + DA) on the emulsion (50 mass %) the volume of water released after settling increases by 13 mass %. Temperature rise to 50 °C has a negative effect on layering causing a decrease in the amount of water phase by 9 %. This may be due to the fact that heating causes an increase in the reactivity of resins and asphaltenes that are present in the

TABLE 2

Size of drops in emulsions (10 and 50 mass %) before and after LFAT (thermostating for 1 h at 50 $^{\circ}\mathrm{C})$

Emulsion sample	Water	Vater content in emulsion			
	10 mas	10 mass %		50 mass %	
	Size of drops, µm				
	$d_{_{ m min}}$	d_{\max}	$d_{_{ m min}}$	d_{\max}	
Without LFAT	2.1	67.2	1.7	32.0	
LFAT for 0.5 min	1.8	71.7	2.2	131.6	
LFAT for 1 min	2.1	40.0	3.5	149.6	
LFAT for 3 min	2.2	17.8	2.9	21.0	
LFAT for 5 min	1.9	20.5	1.1	14.3	
LFAT for 10 min	0.9	23.9	2.1	31.6	



Fig. 3. Microphotographs of emulsions (50 mass %) after LFAT for 5 min and thermostating at 20 and 50 °C.

armouring shells of the drops of dispersed phase, which prevents further coalescence of water drops.

The fractions of asphaltenes, resins, and oils were isolated from oil and emulsion samples (10 and 50 mass %) under investigation. It was demonstrated that the content of asphaltenes and resinous components in oil-containing systems increases with an increase in treatment time (Fig. 5). This may be due to the fact that treatment caus-

es the destruction of the solvate shell of water globules of the existing oil colloid dispersed structure due to the liberation of asphaltenes and resinous components occluded with them.

CONCLUSION

The effect of the integrated action of low-frequency acoustic treatment, demulsifying agent



Fig. 4. Kinetics of water release from emulsion (50 mass %) with the addition of DA and after integrated treatment (LFAT + DA) at 20 (a) and 50 °C (b).



Fig. 5. Effect of LFAT time on the content of asphaltenes (a) and resinous (b) components extracted from oil and emulsions (10 and 50 mass %).

and temperature factor on the stability and structural-mechanical properties of water-oil emulsions of high-paraffin low-resin oil from the Ignyalinskoye deposit was investigated. It is demonstrated that acoustic treatment does not lead to the destruction of emulsions containing 10 mass % water but instead it stabilizes the emulsions by increasing the degree of scattering of the dispersed phase.

The treatment of oil and emulsions (10 mass %) is mainly accompanied by an increase in solidification temperature, viscosity and the activation energy of viscous flow, which is the evidence of an increase in their stability. Low-frequency acoustic action on emulsions in the case of water concentration close to the critical level (50 mass %) leads to the partial destruction of water without its release in the free form. A complex emulsion is thus formed, with the drops of both the minimal (1–2 μ m) and the maximal (130–150 μ m) size. To destroy them, it is necessary to use additional methods of oil system treatment (heating to higher temperatures, variation of demulsifyier type and its concentration, an increase in settling time).

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