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RHEOLOGICAL BEHAVIOR OF OILS IN A MAGNETIC FIELD

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The influence of magnetic treatment parameters on the rheological properties of oils has been investigated. It has been noted that the strongest effect on these properties is produced by the regime of magnetic treatment — steady-state or flowing, the volume rate of oil flow, and the induction value of the magnetoactivator magnetic field. The optimal regimes of magnetic treatment have been determined. It has been shown that treatment of oil with a magnetic field promotes a decrease in the solidification temperature and the amount of asphalt-resin deposits.

At present, the most acute problems in oil production are paraffinization of wells and exhaust lines, as well as a high accident rate of pipelines. This is due to the complete absence of pipes with a protective coating and the insufficient number of applied inhibitors preventing corrosion and the formation of asphalt-resin-paraffin deposits (ARPD). Providing the necessary decrease in the rate of corrosion and ARPD, the inhibitors are irrevocably lost; they require special dosing equipment and considerable working expenses. An alternative technique is magnetic treatment (MT) of pumped liquids, which in the last few years has proved to be good in the oil fields of Russia, Vietnam, China, and other countries owing to the low cost of MT devices and the practically complete absence of working expenses [1–6].

Magnetoactivators found the widest practical use for controlling ARPDs and deposits of salts on the walls of oil wells [1, 2, 6]. Settling in the process of evaporation on the walls of pipes, rods, and on other parts of the well equipment, mixtures of salts, paraffin, and resin-asphalt substances (RAS) decrease the flow rate of oil in the pipes, hinder the operation of pumps, and considerably shorten the intervals between cleanings. Magnetoactivators equipped with centering elements are fixed in any part of the tubing string (TS) of the well below the zone of intensive deposition of ARPDs. The application of magnetoactivators decreases the rate of corrosion of pipelines 2–10 times, increases the intercleaning period of operation of TSs by a factor of more than 10, and enables the effect of MT to last for up to 72 h [1, 4, 5].

However, production tests in a number of oil fields revealed both positive effects and negative consequences in using magnetoactivators for combating oil deposits [2]. The absence of theoretical substantiation of the mechanism of magnetic actions on oil dispersive systems (ODS) often makes it impossible to predict in good time the possible effect of this action and design magnetoactivators with optimal parameters. The scientific explanation of results obtained in practice is limited by the insufficient theoretical treatment of the problem of the action of magnetic forces (MF) because of the complexity of the structural and energy transformations proceeding in substances with different structures at the micro- and macrolevels.

Several qualitative theories that form the basis of the mechanism of MF action on ODSs are known. These are: "colloidal" hypotheses, according to which the MF acts on colloidal para-, dia-, and ferromagnetic particles; "ionic" hypotheses, in which the main role is played by the ions contained in water; and "water" hypotheses, according to which the MF acts on water proper. The mechanism of MF action on water systems is reduced to the change in the bonding of microimpurities with liquid medium molecules. The "water" theories became fairly widely known as applied to the MF action on the physicochemical properties of oil and water-oil liquid media [7–9]. They are based on the destruction in the MF of aggregates of iron-containing particles. However, one cannot assign the only and determining role in the processes proceeding in oils subjected to a magnetic force to the microimpurities of iron, which is one of the kinds of mineral composition of mechanical impurities in oils. According to the "colloidal theory," the mechanism of MF action on substances is based on the different behavior of diamagnetic and paramagnetic molecules

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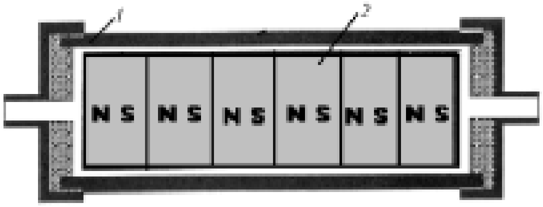

Fig. 1. Diagram of the MAZh magnetoactivator: 1) case; 2) magnetic system consisting of six high-efficiency magnets based on alloys of rare-earth metals Nd–Fe–B providing a magnetic induction amplitude of up to 0.8 T on external magnet poles.

in the external MF [4, 5]. As applied to oil systems, according to this theory, the resins and asphaltenes present in oil are substances of the paraelectric, paramagnetic, and diamagnetic type. The presence in oil of ferromagnets (iron, vanadium, nickel, etc.) also complements the complex spatial interactions of diamagnetic and paramagnetic components of oil in the MF.

The notion about the mechanism of MF action on multicomponent oil systems should take into account the features of oil as the object of investigation, i.e., the content of resins, asphaltenes, and paraffin hydrocarbons, as well as the presence of the water and gas phases and microimpurities [2, 3].

A specific feature of ODSs is their tendency toward a change in the degree of dispersion under the influence of external factors (temperature, pressure, chemical additions, and physical fields), since the supermolecular formations present in ODSs exhibit thermodynamic instability under thermal and dynamic actions. The structural transformations proceeding in ODSs under the action of an MF can be followed by a restructuring of the associates formed from RASs and paraffin hydrocarbons due to the change in their dimensions or their decomposition into components, as well as due to the fact that one or several components simultaneously leave the associates [10, 11].

The influence of the rheological properties on the hydrodynamic characteristics as applied to the flows of high-viscosity and viscoelastic media was investigated by many researchers, beginning from the 1920s. A great contribution to the development of this direction has been made by R. Bingham, M. P. Volarovich. M. Reiner, and P. A. Rebinder [12–15]. By the end of the 1960s, a large volume of experimental and theoretical information about the presence of anomalous rheological properties in different types of (non-Newtonian) liquids, and also in various hydrocarbon systems, including petroleum derivatives, had been accumulated. It was proposed that the anomaly of viscosity of colloidal solutions is due to their structure formation [16, 17]. Because of the necessity of solving problems concerning the preparation, transportation, and refining of high-viscosity and high-solidification oils, for which anomalies of the rheological properties are characteristics, the issues concerning the mechanisms of structure formation in oil systems under the action of external factors and the interrelationship of the structure formation with rheological properties are still burning.

The aim of the present work is to investigate the influence of the alternating MF created by magnetoactivators based on permanent magnets on the rheological properties of oils with a different content of paraffin hydrocarbons and resin-asphaltene substances.

Experimental. As objects of investigation, we chose 16 samples of highly paraffineous oils of the West Siberian region of Russia that are characterized by a higher content of paraffin hydrocarbons (PH), (over 5 mass %), a low density (ρ_{20}), and high solidification temperatures (T_s). Two samples of the highly resinous oils of the Urals region, which contain about 30 mass % of RASs and have a high density (0.93–0.95 g/cm^3) and a low solidification temperature, are high-viscosity and low-fluidity oils.

Magnetic treatment was carried out by means of an MAZh magnetoactivator made at the Siberian integrated chemical works (Seversk, Russia), which is analogous in technical characteristics to the magnetoactivators used in the oil production [18]. In this magnetoactivator, a system of six annular magnets was used, which made it possible to obtain in the clearance between it and the casing several zones with alternating directions of the radial MF. Despite the small dimensions (160 mm long, 40 mm in diameter), the application of composite hard-magnetic materials based on alloys of rare-earth metals neodymium–iron–boron provides a magnetic induction amplitude on the internal pole concentrators of up to 0.6 T, and on the external ones — up to 0.8 T (Fig. 1).

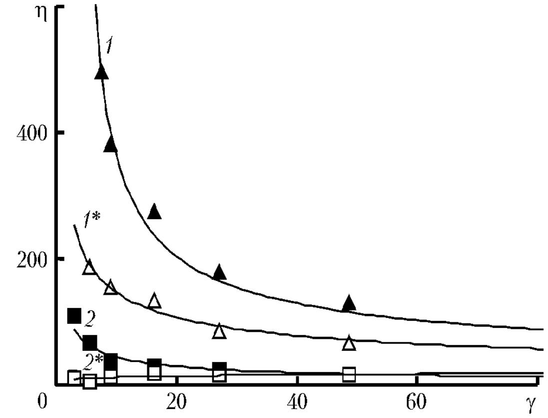

Fig. 2. Viscosity-temperature dependence before and after (*) magnetic treatment of highly paraffineous oils of the oil fields: 1) Ostaninskoye; 2) Samotlorskoye. η, mPa·sec; γ, sec^{-1}.

In the course of the laboratory experiment, the oils under investigation were passed through a magnetoactivator in a teflon pipe of diameter 4.5 mm at room temperature at a volume rate of oil flow $W = 3\ \text{cm}^3/\text{min}$ (the time during which the oil stayed in the working zone of the magnetoactivator was about 4 sec). The rheological characteristics of the oil before and after the MT were determined on a VPZh-2 capillary viscosimeter and a Reotest 2.1 rotational viscosimeter. At rates of shear from 3 to 80 sec^{-1} the values of the critical shear stress (τ_{sh}) and dynamic (η) and kinematic (ν) viscosities were determined.

The solidification temperatures of the oils were determined according to State Standard (GOST) 20287-91. The features of the process of sedimentation in the MF were investigated on the facility developed by the "cold finger" method, which in the last few years has been widely used in laboratory practice for determining the quantity of oil sediments formed [19]. The facility consists of a cooled metal rod (the temperature of the rod is $+10^{\circ}\text{C}$), which is a model of a TS or an oil pipeline placed in a heated oil sample (the oil temperature is $+30^{\circ}\text{C}$). In the course of the experiment, the quantity of ARPDs formed in 60 min on the "cold" rod is determined gravimetrically.

The inhibiting ability (S) of the MF to prevent the ARPD formation in oil was calculated by the formula

$$S = \frac{m - m_{\text{m.t}}}{m} \cdot 100\% .$$

The reproducibility of all experimentally determined rheological parameters was estimated from the results of 3–5 concurrent experiments. The deviations of successive measurements from the arithmetic mean value do not exceed 3.1% for dynamic viscosity, 3.5% for kinematic viscosity, 2.4% for the solidification temperature of oil, and 4.6% in determining the quantity of ARPDs.

Results and Discussion. The rheological dependences of dynamic viscosity on the rate of shear $\eta = f(t)$ at 20°C for the highly paraffineous oils of the Ostaninskoye and Samotlorskoye oil fields before and after the MT are given in Fig. 2. The rate of shear strongly influences the fluidity of the oils, and they exhibit the properties of non-Newtonian liquids at rates of shear of up to 100 sec^{-1} (Fig. 2, curves 1, 2). The magnet-treated oils, when flowing, exhibit Newtonian properties, the rheological curves are partially rectified, and there is a marked decrease in the values of the critical shear stress and effective viscosity (Fig. 2, curves 1^*, 2^*, see Table 1).

The alternating MF, depending on the configuration of the field lines, the number of polarity reversals, the intensity, and the time of action, can produce a different effect on the oil. We performed a set of experiments to investigate the influence of magnetization regimes (steady-state and flowing ones), the MT time or volume rate of the oil flow and the magnetoactivator magnetic induction on the rheological properties of the oils.

The influence of magnetization regimes on the kinematic viscosity of the oil was considered with the example of the highly paraffineous oil of the North Kalinovoye oil field and the highly resinous oil of the Russian oil field (Fig. 3). Treatment in a stationary MF for 5 h leads to a destruction of the crystal paraffin lattice of the highly paraf-

TABLE 1. Inhibiting Ability (*S*) and Temperature Effect (ΔT) of Magnetic Treatment of Highly Paraffineous Oils

Oil field	PH, mass %	RAS, mass %	τ_{sh} (before/after MT), Pa	E_{act}, (before/after MT), kJ/mole	*S*, %	ΔT, °C
			RAS less than 5 mass %			
Urengoiskoye	20.0	1.7	260.8/163.0	55.8/49.9	1.0	0.5
Chkalovskoye	24.0	1.9	94.5/65.2	59.3/47.9	2.4	1.0
Chertalinskoye	16.3	3.0	260.0/202.1	52.0/49.5	9.6	4.5
North Ostaninskoye	16.0	4.4	123.9/81.5	36.6/32.2	7.7	1.5
			RAS from 5 to 15 mass %			
South-Tambaevskoye	16.0	10.7	94.5/19.6	30.9/34. 8	16.5	13.5
Sobolinoye	8.4	10.1	91.3/19.6	23.8/19.0	19.3	7.0
Samotlorskoye	9.2	12.3	32.6/3.3	12.6/7.0	25.0	21.0
Urmanskoye	9.5	11.1	29.5/28.0	23.2/20.4	82.0	7.5
Shirotnoye	10.8	15.6	29.3/13.0	18.2/16.4	65.6	8.0
Kalinovoye	6.4	14.6	52.2/22.8	28.5/22.8	45.4	8.5
			RAS over 15 mass %			
North Kalinovoye	7.1	19.2	16.3/71.7	10.1/13.5	–16.0	6.2
Gerasimovskoye	5.1	19.3	21.2/24.9	22.9/28.2	–16.7	17.0
Cheremshinskoye	7.8	19.6	16.3/29.3	7.8/11.7	–20.5	12.5

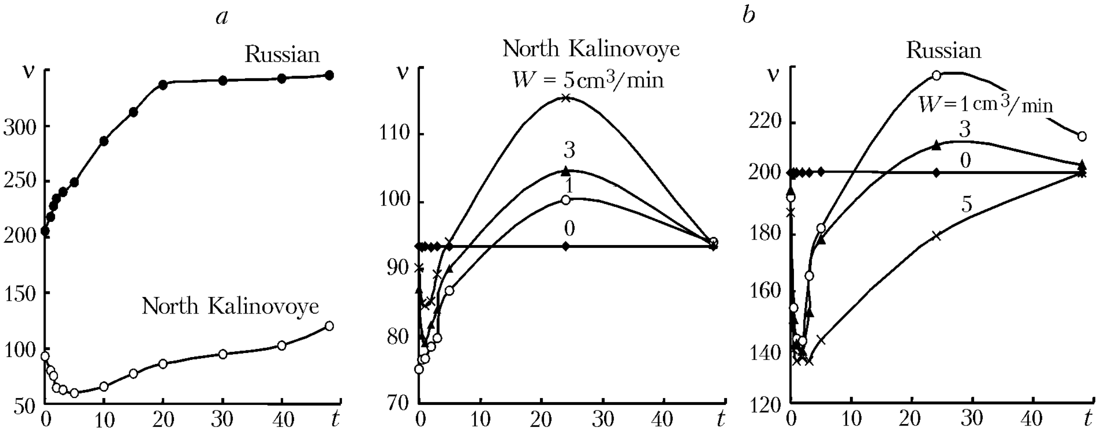

Fig. 3. Influence of the MT time under stationary conditions (a) and of the oil-flow volume rate *W* (b) on the kinematic viscosity of the highly paraffineous oil of the North Kalinovoye oil field and of the high-viscosity oil of the Russian field. ν, mm^2/sec; *t*, h.

fineous oil (Fig. 3a) and to a decrease in viscosity by a factor of 1.5. Further increase in the MT time markedly decreases its efficiency. For the highly resinous oil, treatment in the steady-state regime for up to 20 h increases the viscosity by 60–65% (Fig. 3a). In this case, the increase in viscosity may be associated with the considerable strengthening of the structure due to the interaction and coagulation of the associates of RASs. Further MT for 20–48 h practically does not lead to any change in the viscosity of the highly resinous oil.

We have investigated the influence of the volume rate *W* of the oil flow on the kinematic viscosity ν of the highly paraffineous and highly resinous oils as they are flowing through the magnetoactivator MF (Fig. 3b). For both samples, upon MT the viscosity first decreases and then, in several hours, it gradually regains its original values and increases to values much higher than the original ones. After 24–48 h, the viscosity decreases to values close to the original ones. It has been established that the features of the rheological behavior of oils with a different content of PH, resins, and asphaltenes in the MF is determined by the value of *W*. For the highly paraffineous oil of the North-

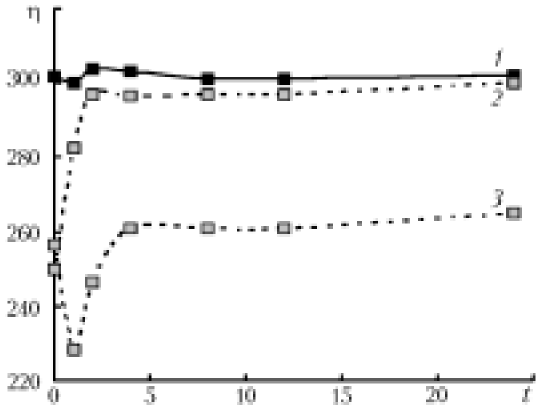

Fig. 4. Time change in the dynamic viscosity (at $\gamma = 9\ \text{sec}^{-1}$) of the high-viscosity Taimurazinskoye oil field treated in magnetoactivators with a different magnetic induction: 1) oil without MT; 2) in magnetoactivator M1 (0.4 T); 3) in magnetoactivator M2 (0.8 T). η, mPa·sec; t, h.

Kalinovoye oil field (Fig. 3b), with decreasing flow rate W the efficiency of magnetic action increases (at $W = 1$ cm^3/min ν decreases by 20% at most). The viscosity 5–24 h after MT exceeds the original values by 20%, and after 48 h its complete recovery is observed.

For the highly resinous oil of the Russian oil field, upon MT the viscosity decreased by 50% independent of the volume rate W, producing an appreciable effect only on the relaxation properties of the oil (Fig. 3b). The higher the value of W, the longer the viscosity relaxation time: at $W = 5$ cm^3/min the relaxation time is 48 h, whereas at $W = 1$ cm^3/min it is only about 10 h. However, additional experimental studies have shown that a further increase in the volume rate W leads to a sharp decrease in the MT efficiency. Therefore, in the subsequent experiments MF-treatment of samples of different compositions was carried out at $W = 3$ cm^3/min.

The rheological properties of the oils are also strongly influenced by a constant MF. In the experiment, the highly resinous oil of the Taimurzinskoye oil field was treated in magnetoactivators with different values of magnetic induction (Fig. 4). It has been established that with increasing magnetic induction in the magnetoactivators a more considerable decrease in the viscosity is observed. For instance, upon treatment in the magnetoactivator M1 (0.3–0.4 T) the viscosity decreased by 17% at most (Fig. 4, curve 2), and in M2 (0.6–0.8 T) — by 25% (Fig. 4, curve 3). For the oil sample treated in M1, η regained its original value after 1 h, and after passing through the magnetoactivator M2 complete recovery of η was not observed for 25 h. Further investigations have shown that the period of complete recovery of the rheological properties of the oils can vary between 3 and 5 days [20, 21].

The state of any thermodynamic system depends on the energy ratio between the intermolecular interaction and the thermal motion; therefore, for the oil system the activation energy of the viscous flow E_{act} and the associativity (structuring) parameter of the liquid turn out to be interrelated. To investigate the features of the structure-formation processes in flowing oil systems, we used the Frenkel–Eyring equation of the viscosity-temperature dependence:

$$\eta = A \exp\,(-E_{\text{act}}/RT)\,,$$

where E_{act} is the mean excess energy of reacting ("active") molecules as compared to the other ("inactive") molecules; $\exp\,(-E_{\text{act}}/RT)$ characterizes the portion of "active" molecules in the reaction mixture that changes dramatically with temperature [22, 23]. To calculate E_{act}, we plotted the logarithmic temperature dependence of the dynamic oil viscosity $\ln \eta - 1/T$ in the 20–60°C temperature range (Fig. 5). The quantity E_{act}, which is determined by the slope of the tangent to the curvilinear dependence, characterizes the strength of the bonds in associative complexes in each structural state. Usually for oils of different compositions the value of E_{act} is varied from 3 to 60 kJ/mole [20, 23, 24].

For highly paraffineous oils, the plot, as a rule, represents two sections corresponding to certain types of structure formation and separated by an inflection point. This point corresponds to the phase-transition temperature at which there occurs a destruction of the crystal structure of paraffin hydrocarbons that is characteristic of oils under

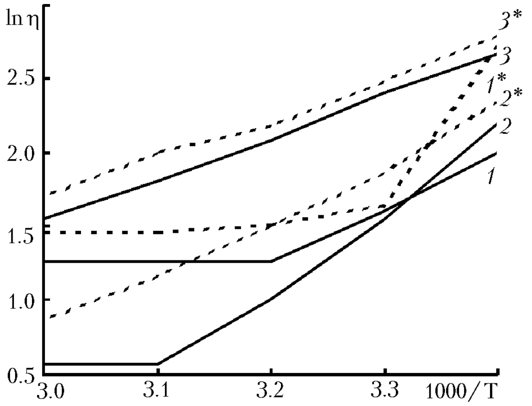

Fig. 5. Logarithmic dependence of the dynamic viscosity of highly paraffineous oils on the inverse temperature: 1) North Kalinovoye oil field; 2) Cheremshinskoye; 3) highly resinous oil of the Russian oil field in semilogarithmic coordinates before and after (*) MT.

these conditions — for the highly paraffineous oils it is 40 and 50^{o}C, respectively (Fig. 5, curves 1, 2). The change in the value of E_{act} is due to the stepwise change in the degree of association of molecules in the oil system and is indicative of the phase transitions that have taken place [23].

Upon MT, the shape of the viscosity–temperature curves is determined by the composition of the resinous components of oils. For highly paraffineous oils with a high content of nonpolar resins, two sections of the curve are preserved, but the inflection point shifts by 10–20^{o}C into the region of low temperatures (Fig. 5, curve 1^{*}). For magnet-treated highly paraffineous oils with a high content of nonpolar resins (Fig. 5, curve 2^{*}) and highly resinous oils before and after MT the plot is represented by only one linear function without an inflection point (Fig. 5, curves 3, 3^{*}).

The results of the investigation of the influence of MT on the rheological properties of 13 samples of paraffineous oils with a different content of PH and RASs are given in Table 1. The oils of the Chkalovskoye, Urengoiskoye, Chertalinskoye, and North Ostaninskoye oil fields are highly paraffineous with a content of PH of over 16% and a small content of RASs. For the oils of this group, upon MT the structure-formation processes proceed in the supermolecular structure and are accompanied by destruction or the formation of the crystal lattice of PH. In so doing, there is a marked decrease in the critical shear stress (τ_{sh}) characterizing the limiting load that the oil system endures in the range of small rates of shear and describing the transition of the oil from the state of rest to the state of flow. Thus, for the oil of the North Ostaninskoye oil field there is a decrease in τ_{sh} by 50% and a more insignificant decrease in E_{act} — by 5–15%.

An increase in the content of RASs in highly paraffineous oils may lead to both positive and negative effects of MT. Oils with a content of RASs of the order of 5–15% (oils of the South-Tambaevskoye, Urmanskoye, Samotlorskoye, Shirotnoye, and other oil fields) upon magnetic treatment are characterized by a decrease in the main rheological parameters, and oils with a content of RASs over 15%, such as the oils of the North Kalinovoye and Cheremshinskoye oil fields, are characterized by their increase.

For many years, the application of MF in the oil production and pipeline transport was restricted mainly to controlling the ARPD formation on the walls of oil equipment. Simultaneously, the question of the influence of a constant MF on the solidification temperature was considered [1, 3–5]. However, the investigations made are uncoordinated and do not explain why the effect of using magnetoactivators in particular oil fields is insufficient or, in individual cases, negative. To establish the relation between the MT efficiency and the content in the oils of paraffin hydrocarbons, resins, and asphaltenes, we made experimental investigations of the influence of the MF on the process of oil-deposit formation and the solidification temperature of highly paraffineous oils with a different content of RASs and PH. We estimated the inhibiting ability of the MF to prevent the oil-deposit formation on the metal surface of equipment and the depressing effect by the decrease in the solidification temperature (see Table 1).

Treatment of highly paraffineous oils with a high content of PH and a low content of RASs proves to be ineffective — no marked change in the solidification temperature and quantity of ARPD is observed. Upon MT of

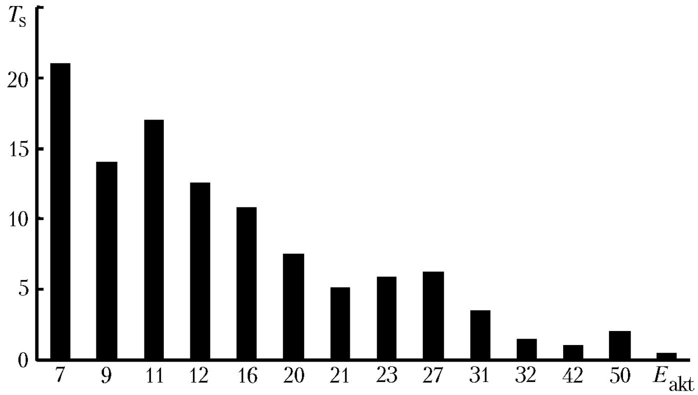

Fig. 6. Change in the solidification temperature of the highly paraffineous oils upon MT depending on the viscous flow activation energy. T_s, ^oC; E_{act}, kJ/mole.

highly paraffineous oils with a content of RASs over 5%, usually there is a decrease in T_s by 6–20°C. In so doing, MF-treatment of oils with an RAS content from 5 to 15% leads to a marked decrease in the deposit formation (the degree of inhibition S is of the order of 60–80%). In a number of oils with an RAS content over 15%, upon MT the quantity of the deposit formed increases (negative values of S in Table 1).

The correlation analysis has revealed that there exists a relation between the solidification temperature of magnet-treated highly paraffineous oils and E_{act}. Figure 6 shows the dependence of the depressing effect of a decrease in T_s on the value of E_{act} for magnet-treated highly paraffineous oils, according to which the efficiency of magnetic treatment appreciably decreases as E_{act} increases to values higher than 30 kJ/mole.

On the basis of extensive and detailed investigations with the use of various methods, we have shown that in MT of oils of different compositions an important role is played by the paramagnetic and diamagnetic components — asphaltenes and resins that are present in the oil. The application to a structured oil system containing paraffin hydrocarbons, resins, and asphaltenes of an alternating magnetic field of the magnetoactivator leads to destruction of the crystal structure of paraffin hydrocarbons. The decrease in viscosity upon MT is due to the more labile and active RASs — polar resins, in whose structure reaction-active components are present. And the RASs thereby simultaneously perform the function of a structural-mechanical barrier on the surface of dispersed-phase particles, increasing the depth and rate of its destruction and impeding the formation in the oil of the crystal structure of paraffin hydrocarbons. For oils in the composition of whose resins barely active and weakly polar substances prevail, upon MT an increase in the values of the rheological characteristics can be observed [10, 24].

CONCLUSIONS

1. From the results of the MT of highly paraffineous and highly resinous oils, it may be concluded that the efficiency of magnetic action is strongly influenced by such characteristics as magnetoactivator magnetic-field induction, the regime of MT — steady-state or flowing, as well as the MT time or the volume rate of the oil flow. We have determined the optimal regimes of experimental studies of the influence of MT on the oil. Magnetic treatment was carried out with the aid of an MAZh magnetoactivator with a magnetic field induction from 0.6 to 0.8 T in a dynamic regime with a volume rate of 3 cm^3/min.

2. Upon magnetic treatment, there is a change in the activation energy of the viscous flow accompanied by a stepwise change in the degree of association of molecules, a shift of the phase-transition point into the region of lower temperatures in one case and the absence of a phase-transition point in the other.

3. The efficiency of magnetic treatment — the decrease in the critical stress of shear, the effective viscosity, the solidification temperature, and the quantity of the oil deposit formed — largely depends on the content in highly paraffineous of paraffin hydrocarbons, resins, and asphaltenes. Magnetic treatment produces a positive effect on oils with a content of paraffins not exceeding 16% and resinous-asphaltene substances of the order of 5–15%.

4. Extensive studies of the rheological behavior of oils of different composition in the MF makes it possible to deepen and widen our understanding of issues considering the influence of physical fields on different structured systems, including the oil colloidal-dispersive systems investigated by us.

NOTATION

A, pre-exponential factor; E_{act}, activation energy of viscous flow, kJ/mole; m, quantity of deposit formed in oil without MT, g; $m_{\text{m.t}}$, quantity of deposit formed upon MT, g; R, universal gas constant; S, inhibiting ability; T, absolute temperature, K; T_s, solidification temperature of oil, $^{\text{o}}$C; ΔT, change in the solidification temperature of oil upon MT, $^{\text{o}}$C; t, time, h; W, volume rate of oil flow, cm^3/min; γ, rate of shear, sec^{-1}; η, dynamic viscosity, mPa·sec; ν, kinematic viscosity, mm^2/sec; ρ_{20}, oil density at 20$^{\text{o}}$C, g/cm^3; τ_{sh}, critical shear stress, Pa. Subscripts: act, activation; s, solidification; m.t., magnetic treatment; sh, shear.

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