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Chapter

Applied Problems in the Rheology of Structured Non-Newtonian Oils

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Abstract

The rheology problems of non-Newtonian oil, accompanied by the physical phenomena of formation and destruction of coagulation structures and aggregates, significantly affecting the flow are considered and analyzed. Also are considered issues of hydrodynamic interaction of particles leading to the formation of disordered structures, which significantly change the rheological properties of non-Newtonian oil. It has been noted that the formation of coagulation structures depends on energy dissipation, viscosity, stress or shear rate, and the size of the particles forming the structure. With increasing asphalt-resin content in the oil, the probability of particle collision increases, increasing the rate of formation of various disordered structures up to a framework that nullifies the rate of oil flow. Applied problems of rheology, including dissolution kinetics of asphalt-resinous substances in aromatic hydrocarbons and improving rheological properties of the oil, rheology of structured non-Newtonian oils in gas lift method of production, as well as possible ways to create new technologies for processing non-Newtonian oils were considered.

Keywords: rheology, non-Newtonian oil, coagulation structures, models, asphalt-resinous substances, kinetics, technology

1. Introduction

The world has huge reserves of heavy oils that show non-Newtonian properties, and their use is limited by a lack of efficient extraction, transport, and refining technologies.

Non-Newtonian oils are characterized by a fairly high content of asphalt-resinous substances and paraffinic compounds that are prone to coagulation structures and aggregates. This factor significantly affects the rheological properties of the oil, primarily its effective viscosity and diffusion, making its transport and storage difficult. At the same time, there are many methods of improving the rheological properties of oils required in the production, transportation, and refining of the latter. They are associated with improving temperature conditions, as well as creating efficient technologies using various chemical reagents that reduce the surface tension and viscosity of rheological fluids and many other factors that enable intensification of the extraction, transport, and refining processes. The flow of highly viscous fluids and dispersed systems with a high content of dispersed particles is characterized by certain

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complexities, mainly related to the non-linearity of the disordered structure and the properties of the carrier phase. Accordingly, the rheology of such systems, at present, can be classified as:

- the rheology of highly viscous systems (oils, greases, gels, oils, heavy refinery fractions, etc.) which do not obey Newtonian fluid flow laws [1–4] $\tau = -\eta \partial U / \partial x$

- the rheology of dispersed systems (oil suspensions and emulsions) is characterized by the formation and collapse of coagulation structures due to the hydrodynamic interaction of different particles, whereby the rheological flow properties depend on particle content and properties as well as on shear stress and elasticity limit [5–7]

- a special class of problems related to the rheology of non-Newtonian oils or oil dispersion systems containing, in addition to solid-phase particles, liquid water droplets and gas bubbles, asphaltenes, resins, and, at low temperatures, paraffin particles dissolved in oil [8–10]. The presence of this spectrum of particles of various varieties and nature leads to the formation of highly complex coagulation structures, subsequently transforming into aggregates, clusters of aggregates, up to the formation of a viscoelastic framework [6, 11–13].

Analysis of the literature studies [1, 6, 11, 13–15] showed that the most widespread in engineering practice are rheological empirical or semi-empirical equations. For heavy viscoplastic oil, s the following rheological equations are the most acceptable: Bingham mod: $\tau = \tau_0 + \eta \bullet \gamma$; Ostwald-de-Ville model: $\tau = k_0 \bullet \gamma^n = k_0 \bullet \gamma^{n-1} \bullet \gamma$; Hershel-Bulkley model (Hershel-Bulkley): $\tau = \tau_0 + k_0 \bullet \gamma^n$. In practical calculations, various modifications of these equations using are possible.

2. Formation of disordered coagulation structures and aggregates in oils

The flow of oil disperse systems is accompanied by various physical phenomena of hydrodynamic interaction, and collision, resulting in the formation of coagulation structures and aggregates, in which energy dissipation plays an important role. Hydrodynamic interaction of particles among itself is observed at their high concentrations when the distance between the particles allows them to collide: $l \approx 80 a^3 \sqrt{\rho_d / C_m}$. The probability of a particle collision is determined by many factors: the number of particles itself per unit volume, their shape and properties, the properties of the medium, the flow velocity, and temperature, on which important parameters such as diffusion coefficient and viscosity depend. In viscous fluid flow, the dissipative function is determined as

$$\varphi_D = 2\eta_C \left(\frac{\partial V_x}{\partial x} \right) + 2\eta_C \left(\frac{\partial V_y}{\partial y} \right)^2 + \eta_C \left(\frac{\partial V_x}{\partial y} + \frac{\partial V_y}{\partial x} \right)^2 \tag{1}$$

The energy dissipated in the volume of the liquid is expressed by the formula

$$-\frac{dE}{dt} = \int \frac{\eta_C}{2} \left(\frac{\partial V_1}{\partial x_1} + \frac{\partial V_k}{\partial x_i} \right)^2 dv \tag{2}$$

Energy dissipation per unit mass of flow is defined as follows

$$\varepsilon_R = -\frac{dE}{dtdm} = \frac{\eta_C}{2}\left(\frac{\partial V_1}{\partial x_r} + \frac{\partial V_k}{\partial x_i}\right)^2 \tag{3}$$

(where)

$$dm = \rho_C dv$$

For non-Newtonian oils, expression (Eq. (3)) will be written as

$$\varepsilon_R = -\frac{dE}{dtdm} = k_0\left(\frac{\partial V_1}{\partial x_r} + \frac{\partial V_k}{\partial x_i}\right)^{n+1} \tag{4}$$

where $k_0 = \frac{\eta_C}{2}\left(\frac{\partial V}{\partial y}\right)^{n-1}$ – consistency coefficient. In the one-dimensional case for viscoplastic oil flow, by applying expression (Eq. (4)), we have

$$\varepsilon_R = k_0\dot{\gamma}^{n+1} \tag{5}$$

Coagulation structures are formed by intermolecular bonds between the particles, moreover if liquid interlayers remain between the particles, then the thickness of these interlayers significantly affects the strength of the coagulation structure. Aggregate-unstable oil systems are characterized by the inconstancy of the medium due to continuous structuring and changes in the physical properties of the particles, i.e. changes in volume and size of asphaltene particles caused by their interaction, collision, coagulation, and crushing at a certain concentration in an enclosed volume [16]. The connection between the structure and viscosity of oil dispersal systems as well as the features of their non-Newtonian flow is explained by a change in structure resulting from the emergence and collapse of aggregates from asphaltene particles in the presence of resins. Oil structured systems containing high molecular paraffin crystals, resins, and asphaltene particles at very low laminar flow velocities or in absence of flow form a chain or in the extreme case a continuous grid (frame) between itself and the structure of the porous medium. **Figure 1** shows the formation of a framework of asphaltene particles contained in oil [5, 9].

In [17] it is noted that real oil disperse systems are classified by activation energies into two structural groups differing by the nature of intermolecular interaction of particles in an oil disperse medium. These groups are distinguished from each other by their asphaltene and resin content and, they can be classified into immobile with low asphaltene content, and, interacting with high asphaltene content. **Figure 2** illustrates the characteristic changes in activation energies for the two groups.

A large amount of asphalt-resin substances in the oil reduces the activation energy and creates more favorable conditions for the formation of coagulation structures. The change in mass of non-deformable nanoaggregates is defined as
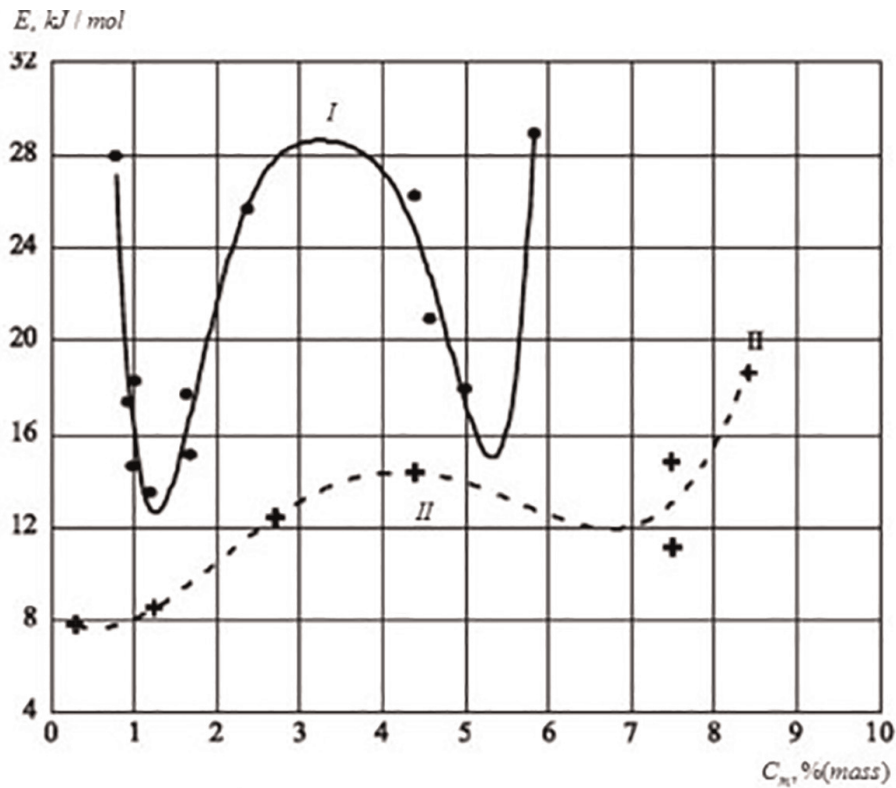
$$\frac{dm}{dt} = (m_\infty - m)\omega \tag{6}$$

$$t = 0, m = m_0$$

where ω is the frequency of particle collision.
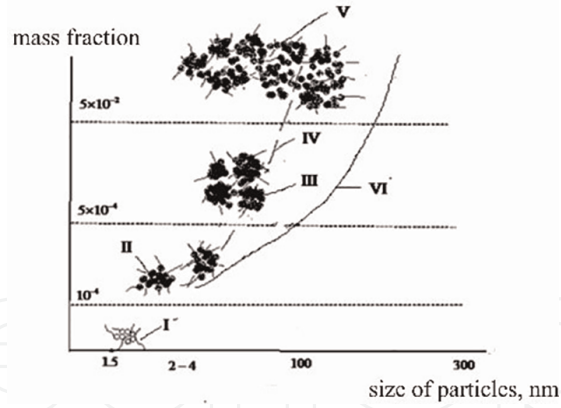
Figure 1.
Asphaltene particle aggregation in oils: I- single molecules and particles; II- nanoaggregates; III- clusters of nanoaggregates; IV- unstable suspension; V- viscoelastic framework; VI- stable emulsion with toluene.

Figure 2.
Dependence of activation energy on asphaltene content for the stationary group (I) and the interacting group (II).

The solution to this equation will be represented as

$$m = m_\infty - (m_0 - m_\infty) \exp(\omega t) \qquad (7)$$

If assume that $m_\infty >> m_0$, we will get

$$m = m_\infty[1 - \exp(-\omega t)] \qquad (8)$$

where m_∞- is the limiting steady mass of the aggregate, m_0 - is the initial mass of the aggregate.

Assuming the spherical shape of nanoaggregates and imagine that, $m = \frac{\pi}{6}a^3\rho$, the size of the nano-aggregates, will be defined the following form, considering (Eq. (7)),

$$a_g = a_{g\infty}\left[1 - \exp\left(-C_0\phi_0\left(\frac{\varepsilon_R}{\nu_c}\right)^{1/2}t\right)\right]^{1/3} \qquad (9)$$

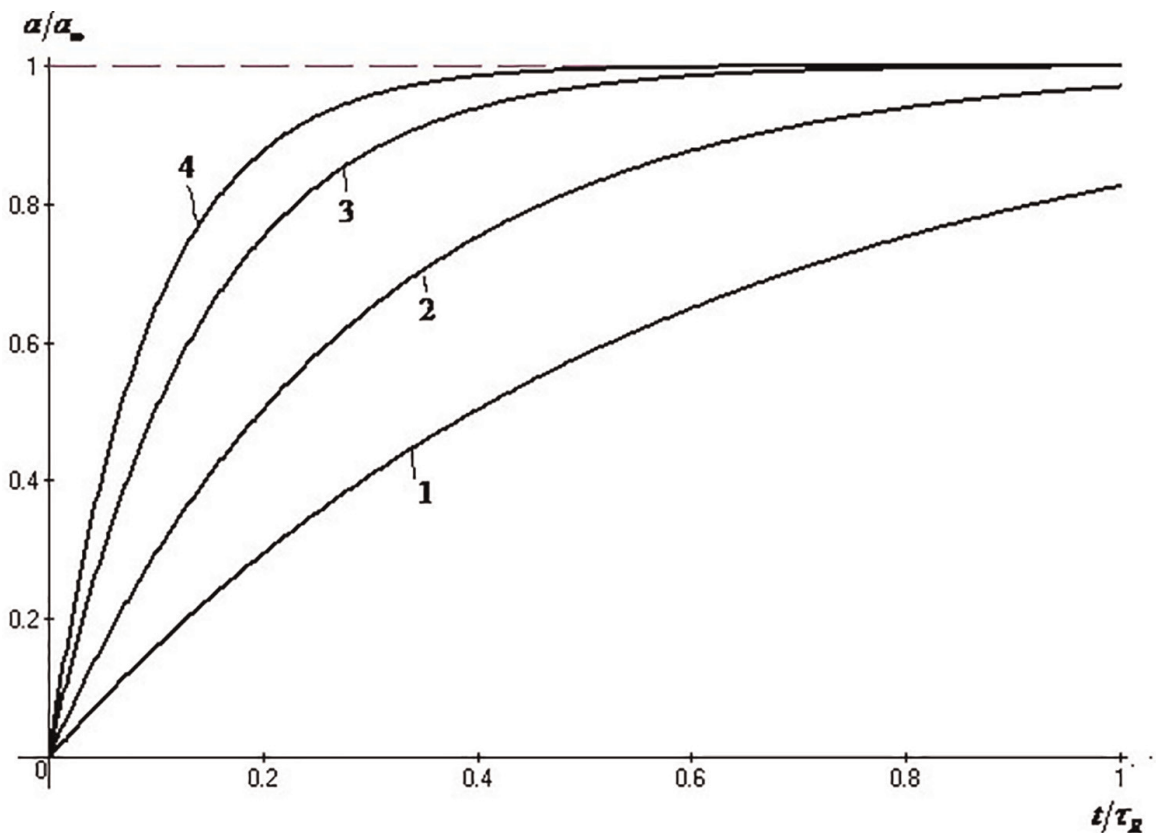For laminar flow, the formation of aggregates is expressed by an equation of the form.

$$\alpha_g = \alpha_{g\infty}[1 - \exp(-8\pi DN_0\alpha_0 t)] \qquad (10)$$

According to **Figure 3**, the size of the nano-aggregates fluctuates in range and the maximum size of the framework is limited by the presence of pore or tube walls. The frequency of collisions between asphaltenes increases as the rises of volume fraction of asphaltene particles. The relaxation time for turbulent flow is defined by the expression $\tau_R = (\nu_C/\varepsilon_R)^{1/2}$ and for laminar flow $\tau_R = 3\nu_C/(8kTN_0)$, which leads to a rapid reaching of the final aggregate size. With increasing oil viscosity, for both laminar and turbulent flows, the collision frequency of asphaltene particles decreases, which inhibits the rate of nanoaggregate formation.

Maxwell's rheological equation for a viscoelastic fluid in substantive derivatives is written as [3]

$$\lambda\left(\frac{\partial\tau}{\partial t} + U\frac{\partial\tau}{\partial y}\right) + \tau = \eta_c\bullet\gamma \qquad (11)$$

$$t = 0, \tau = \tau_0, \bullet\gamma = 0$$

Figure 3.
Variation of nano-aggregate size over time depending on the content of asphaltene particles in the oil: $1 - \varphi = 0.05$; $2 - 0.1$; $3 - 0.2$; $4 - 0.3$..

A special form of equation (Eq. (11)), is given in the following form

$$\lambda \left(\frac{\partial \tau}{\partial t} + U \frac{\partial \tau}{\partial y} \right) + \tau = 0 \tag{12}$$

The solution of Eq. (12) can be represented as

$$\tau = C_1 f(y - Ut) \exp\left(-\tau/\lambda\right) \tag{13}$$

Substituting this solution into (Eq. (13)) we obtain the identity. Here $\lambda = \eta_c/G$ - is the relaxation time by Maxwell, U - is the rate of displacement of the deformation front, $f(y - Ut)$- is the function defining the deformation displacement front in the framework, y- is the coordinate, G- is the modulus of shear elasticity, $\dot{\gamma} = d\gamma/dt$ - is the rate of shift, y- is the shifting gradient, τ_0 - is the ultimate shift stress or the yield stress.

Moreover, if $\tau \leq \tau_0$, then $\dot{\gamma} = 0$. The complete solution of equation (Eq. (13)) will be

$$\tau = C_1 f(y - Ut) \exp\left(-\tau/\lambda\right)\tau_0 \tag{14}$$

Otherwise, the equation can be represented in logarithmic form

$$\ln \tau = \ln \tau_0 - t/\lambda + \ln\left(C_1 f(y - Ut)\right), \tau_0 = \eta_c \dot{\gamma} \tag{15}$$

Obviously, the value t/λ in equation (Eq. (15)) characterizes the deformation of the viscoelastic framework in time and depends on the velocity or pressure gradient. In approximation, this dependence can be presented as $t/\lambda = t\dot{\gamma}/Wef\left[\left(gradP/(gradP)_0\right)^n\right]$, (where $We = \lambda\dot{\gamma}$) is the Weissenberg number).

As can be seen from **Figure 1**, the reduction in asphalt-resin content while dissolving them in aromatic hydrocarbons inhibits the formation of various disordered structures.

3. Mechanism and kinetics of dissolution of asphalt-resinous substances in aromatic hydrocarbons

The presence of asphalt-resin compounds in high-viscosity oil, in heavy oil products (tar, bitumen), and in waste oil water has a negative effect on oil extraction, transport, treatment, and refining processes. Due to their high adhesion and cohesion capacity, asphalt-resin compounds adhere and precipitate on equipment surfaces, forming deposits of a certain thickness. In refining processes at high temperatures, asphalt-resin substances form heavier remains (coke) which deposit on the surface of water droplets in oil emulsions, and form adsorption layers which complicate the coalescence and aggregation of water droplets, thereby inhibiting the stratification and separation of the oil–water system. The presence of asphaltenes in oil determines its rheological properties through the formation of coagulation structures - nanoaggregates, nanoaggregate clusters, and eventually a viscoelastic framework, which worsens the rheological properties of the fluid and complicates oil filtration in the porous reservoir, its production, and transport. As noted above, the usage of different reagents prevents the formation of certain structures in the oil volume,

which leads to a decrease in its effective viscosity and an improvement in rheological properties. According to the work, asphalt-resin substances are very soluble in hexane and aromatic hydrocarbons (gasoline, toluene, xylene) and poorly soluble in alcohols and esters.

The problem of dissolving asphaltenes is an important one for improving the rheological properties of oil and for processes of cleaning waste oil water from asphalt-resin compounds by liquid-phase extraction.

Along with asphalt-resinous substances, crude oil contains some by-products: for-mat the ion water, solid phase, and other impurities that also affect the rheological properties of the oil. Usually, the formation water in an oil volume is in a dispersed form, that is, in other words, the form of droplets, which under different flow conditions can form coagulation structures (flocculus) that significantly affect rheological properties. The separation of water from oil, or a significant reduction in the water content of oil, also has a positive effect on the rheological properties of the last.

The present section will be considered only improving the rheological properties of the crude oil by partial dissolution of asphalt-resinous substances in aromatic hydrocarbon. Experimental studies have shown that the best characteristics of mixability and solubility in heavy oil asphaltenes have been obtained with aromatic solvents and solvent mixtures at different temperatures. It has been discovered that temperature and solvent properties are the physical parameters that most affect the dissolution phenomenon of asphaltenes in oil. The aromatic hydrocarbon content is a key chemical property of solvent blends that improves their solubility properties. Works, [17–19] presented the degree of asphaltene deposition depending on the toluene content for different oilfields.

It has been noted that the degree of asphaltene deposition using a solution of 65–69% normal heptane and toluene decreases to 30–35% (wt.). In works [21–23], is offered a model of the kinetics of asphaltene-resin substances dissolution in hexane and in a mix of hexane and benzene for the Russian oils.

The presented model has an empirical character, although satisfactorily describes the experimental data

$$\alpha = 1 - \exp\left(-kt^n\right) \qquad (16)$$

where α - dissolution degree, k - dissolution rate constant, n- index of degree $n = 0.5 - 1.05$. Moreover, the kinetics of the process is not built on the phenomena of mass transfer under certain flow conditions, but is regarded simply as a heterogeneous process.

In work [22] for the convenience of determining the kinetic parameters, the exponential model is decomposed into a McLaren series, which reduces the non-linear model to a two-parameter Langmuir model. Many experimental data and kinetic curves of dissolution of asphaltenes in aromatic hydrocarbons are given in [21–23].

The most widespread model of particle dissolution kinetics is the membrane model, according to which in each phase there are fixed or moving laminar boundary layers directly adjacent to the particle boundary, in which the transport is carried out by molecular diffusion. Therefore, there is a scheme for a stationary unit particle in which the whole boundary layer is considered as an area where turbulent pulsations do not excite, and the variation of the solute concentration is characterized by a linear dependence. Unlike laminar flow, in turbulent flow the buffer sublayer plays this role. At intensive mixing in the apparatus, due to the appearance of developed isotropic turbulence, the kinetic curves may differ from the linear dependence, because of the

complex distribution profile of the turbulent diffusion coefficient and flow velocity in the boundary layer. Dissolution of asphaltene particles belongs to mass-exchange processes and, depending on the hydrodynamic flow regime, is characterized by different mechanisms. Especially, in the fixed-film model, there is a thin boundary diffusion layer at the interface, in which the entire concentration gradient of the substance is concentrated, and the transfer through this layer is carried out only by molecular diffusion. The turbulent boundary layer model describes the mass transfer between a fixed boundary and a turbulent fluid flow with a complex turbulent diffusion coefficient profile. According to this model, the concentration of substance in the flow is constant and in the turbulent sublayer gradually decreases as it approaches the buffer sublayer, where the turbulence gradually attenuates, passing into laminar mode. Usually, the diffusive boundary layer are taking into account: a) the nature of fluid flow and convective transport of substance; b) molecular and convective diffusion in the transverse and tangential direction; c) the absence of a pronounced border between sublayers. The process of dissolution of asphaltene-resin substances in aromatic hydrocarbons is characterized by nonstationary behavior, in connection with which the models of mass transfer differ significantly according to the nature of the hydrodynamic flow around the particle. In particular, at laminar flow around the particle, the diffusion flux of dissolved substance per unit time from the surface of asphaltene particles can be defined by the following expression (23).

$$J = \sqrt{\frac{3}{\pi} \left(\frac{3D_M U}{2R} \right)^{-1/2}} \Delta C, _0 \lambda \geq \lambda_0 \tag{17}$$

In isotropic turbulent viscous streamline, with some assumptions, the same flow is defined as.

$$J = \sqrt{\frac{3}{\pi}} \left(\frac{\varepsilon_R}{\nu_C} \right)^{1/4} (UR)^{1/2} \Delta C, \lambda < \lambda_0 \tag{18}$$

Here $\Delta C = C_0 - C_1$ - driving force of the dissolution process, C_0, C_1- concentration of dissolved substance far from the particle surface and on the surface, D_M- coefficient of molecular diffusion, U- flow velocity, ε_R - dissipation of turbulent flow energy in the unit of mass, R - particle radius, ν_c- viscosity of the medium, α - coefficient. Both solutions were obtained at boundary conditions:

$$r \to \infty, C = C_0; r = R, C = C_1$$

As follows from equation (Eq. (16)), the flow of asphaltene mass from the particle surface is directly proportional to the dissipation of turbulent flow energy and inversely proportional to toluene viscosity, $(\varepsilon_R/\nu_c) = \left(k_0 \dot{\gamma}^{n+1}/\nu_C \right)$ i.e., at high viscosity values corresponding to low temperatures, the dissolution rate decreases and, similarly, at high rotation frequencies corresponding to high energy dissipation values, the dissolution rate increases.

It should be noted that the main requirement for dissolution of particles is the condition of equality of diffusive and convective fluxes on the surface of the boundary layer

$$-D_{eff} \frac{\partial C}{\partial r} \bigg| r = \delta = \beta (C - C^*) \tag{19}$$

where r- is the coordinate in the thickness of the interfacial layer, C- is the concentration of the absorbed substance, C^*- is the equilibrium concentration, δ- is the thickness of the interfacial layer, which is proportional to $\delta \sim (\alpha D_M / V_0)^{1/2}$ where V_0- is the streamline velocity at the particle surface, β- is the mass transfer coefficient, D_E- is the effective diffusion coefficient.

The effective coefficient of diffusion is defined as the sum of molecular D_M and turbulent D_E diffusion D_E, with molecular diffusion dominating in the viscous sublayer as it approaches the surface of the dissolving particle, and with sufficient distance from the surface, turbulent diffusion $D_T \gg D_M$.

The coefficient of turbulent diffusion is a function not only of the physical characteristics of the medium, but also the turbulent characteristics of the flow and the transverse coordinate. According to the statistical theory of turbulent diffusion, the value of mean square displacement of particles of dissolved medium from initial position in radial direction, is defined by dependence

$$r^{-2} = 2D_M \Delta t \tag{20}$$

The dissolution of asphalt-resin substances in toluene depends on the nature and properties of the particle and the solvent.

It can be assumed that the dissolution process is determined by softening of the upper layers of asphaltene particles as a result of diffusion penetration of the solvent into the near-surface layers. In this aspect, expression (Eq. (19)) is the basic condition for dissolution of asphaltene particles. It should be mentioned that in mixing devices the level of turbulence of flow in the apparatus is determined by the rotation frequency of the stirrer. Usually, at sufficiently high rotational speeds, achieved isotropic turbulence, which dampens as it approaches the surface of the particle. The mass flow from the unit surface of asphaltene particles is determined by Eq. (E18), where the dissipation specific energy is proportional to $\varepsilon_R \sim n_0^3 d^2$, n_0, d- rotation frequency and diameter of the mixer. Hence, the dissolution process intensifies with increasing frequency of rotation. In isotropic turbulent flow, the turbulent diffusion coefficient included in equation (Eq. (19)) depends on the dissipation energy and viscosity of the medium

$$D_T \sim \left(\frac{\varepsilon_R}{\nu_0}\right)^{1/2} \sim \left(n_0^{3/2}d\right)/v_c \tag{21}$$

In order to study the kinetics of asphalt-resin substances dissolution process in toluene experimental studies were carried out in stirrers in the temperature range 20–60^{0}C for 60 min at stirrer speed 1000–1200 min^{-1}. Asphalt-resinous substances were extracted by additional evaporation of bituminous tar fraction with density 1280kq/m^3, with asphaltene content of 14.28% and paraffin content of 7.15% from Azerbaijan oils mixture.

The dissolution mechanism of asphalt-resin substances in aromatic solvents is based on three stages: a) diffusive transfer of solvent to the surface of asphaltene particles; b) physical dissolution of surface layers at certain temperatures; and b) diffusive transfer of dissolution products to the volume through the boundary layer. Experimental studies on dissolution of asphalt-resin substances in toluene are presented in **Table 1**. It should be noted that with the increase of stirrer rotation frequency, the process of asphalt-resin substances dissolution in toluene is intensified

t, min	$T = 25^0 C$	$T = 40^0 C$	$T = 60^0 C$
	C, mass of unit	C, mass of unit	C, mass of unit
2.5	0.048	0.085	0.125
10	0.32	0.38	0.47
15	0.48	0.60	0.70
20	0.65	0.73	0.83
25	0.67	0.75	0.91
30	0.71	0.80	0.96
35	0.72	0.83	0.97
40	0.72	0.84	0.96
50	0.72	0.84	0.99
60	0.72	0.84	0.98

Table 1.
Dependence of asphalt-resin solubility on time at different temperatures.

to a certain limit and is improved by increasing the intensity of turbulent flow, turbulent diffusion and mass transfer coefficients. On the other hand, an increase in turbulence intensity leads to the growth of a number of collisions and turbulent diffusion of asphaltene particles, which may contribute to the development of their coagulation processes. At low turbulence intensity, the disperse system tends to structure formation, with visco-elastic rheological properties manifested.

Experimental studies show that at low temperatures the dissolution of asphalt-resinous substances is incomplete $T = 25^0 C$- at -72% and at $T = 40^0 C$-84%.

Using condition (Eq. (19)) and replacing the variables in expression (E20) $dr = \left(2D_{eff}\right)^{1/2} t^{-1/2} dt$, write the expression as follows

$$\frac{\partial C}{\partial t} = -K(C - C^*)$$

$$C(t)|t = 0 = C_0$$

(22)

here $K = \left(\frac{2}{D_{eff} t}\right)^{1/2} \beta$ - coefficient of mass transfer for the dissolution process, C_0 - initial concentration of asphaltenes in toluene, $C(t)$- concentration of dissolved asphaltene-resin substances in toluene. Expression (E22) is an equation for the kinetics of the mass-exchange diffusion process of dissolution. For the non-stationary process of dissolution, at the value of the coefficient K, depending on time, the solution of equation (Eq. (22)) will be presented as

$$C(t) = C^* - (C^* - C_0) \exp\left(-\int_0^t K(t)dt\right)$$

(23)

For pure solvent, this equation is simplified to

$$C(t) = C^* \left(1 - \exp\left(\int_0^t K(t)dt \right) \right) \qquad (24)$$

Using experimental studies and expression (E23), can be placed.

$K(t) = K_0 t^{-1/2}, K_0 = \left(\frac{2}{D_{tff}} \right)^{1/2} \beta$ and $C^*(T) = 57.33 + 0.7T^0C.$

Using the above-mentioned expressions for the dissolution of a single particle from expression (Eq. (24)) we will obtain dependencies, and ultimately the equation for the dissolution of asphalt-resinous substances in toluene will be given as

$$C(t) = C^* \left(1 - \exp\left(-\alpha t^{1/2} \right) \right) \qquad (25)$$

where $\alpha = 2K_0$.

For the "collective" dissolution of more particles, should be used the effective time-averaged value of the mass transfer coefficient

$$\beta_E = \frac{\int_0^{T_P} \beta dt}{T_P} = \beta \frac{t}{T_P} \qquad (26)$$

where T_P - is the complete dissolution time of the particles. As follow from the **Table 1** the time for the complete dissolution of asphaltene particles is about 50–60 min. Considering this expression, the solution (Eq. (26)) will be

$$C(t) = C^* \left(1 - \exp\left(-\alpha t^{3/2} \right) \right) \qquad (27)$$

where $\alpha = \frac{2K_0}{3T_P}$. The results of experimental studies and calculated values according to formula (Eq. (27)) for different temperatures are shown in **Figure 2**.
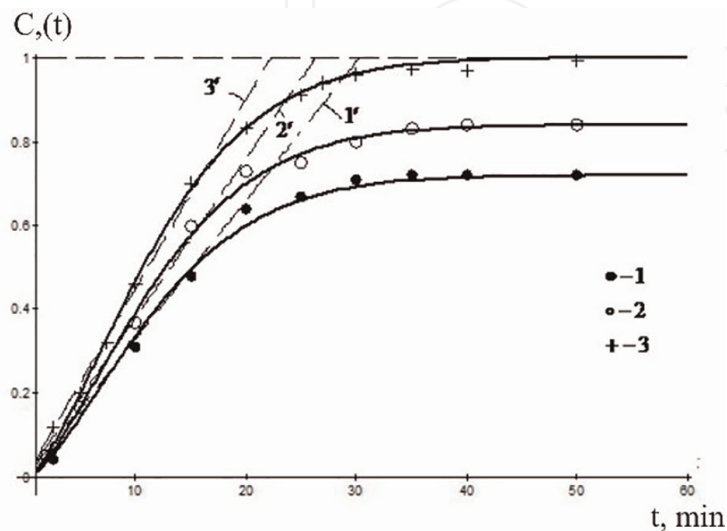
Figure 4.
Kinetic curves of dissolution of asphaltene-resin substances in toluene at temperatures equal to: 1-20°C; 2- 40°C; 3- 60°C. The straight lines correspond to the linear kinetics of asphaltene dissolution in toluene at temperatures equal to:1′ –20°C; 2′–40°C; 3′–60°C.

Practical calculations show that in the indicated range the temperature changes insignificantly $\alpha = 0.025 - 0.030$. Eq. 23 adequately describes experimental data on the dissolution of asphalt-resinous substances in toluene. **Figure 4** shows the areas corresponding to the approximate direct dependence of asphalt-resin dissolution kinetics in the form of the equation $C(t) = K(T)t$.

For a straight line dependence, the dissolution constant can be approximated by an expression depending on the temperature

$$\ln K = -\frac{752.087}{T} - 0.8507 \tag{28}$$

with a correlation coefficient equal to $r^2 = 0.992$. In this dependence T- is the absolute temperature. As **Figure 4** shows, the linear correlation of the direct solubility is only observed at $t \leq 20$ min. and within the viscous sublayer of the entire boundary layer volume.

4. Rheology of structured non-Newtonian oils in gas lift method extraction

The gas lift method is based on the principle of displacing oil from a reservoir by applying compressed gas (air) to its surface. Different schemes for the gas lift method are shown in **Figure 5**. High-pressure gas is injected into the annulus, which will lower the liquid level in the annulus and raise the level in the main pipe where the oil-gas mixture rises. When the liquid level drops to the lower end of the main pipe, the
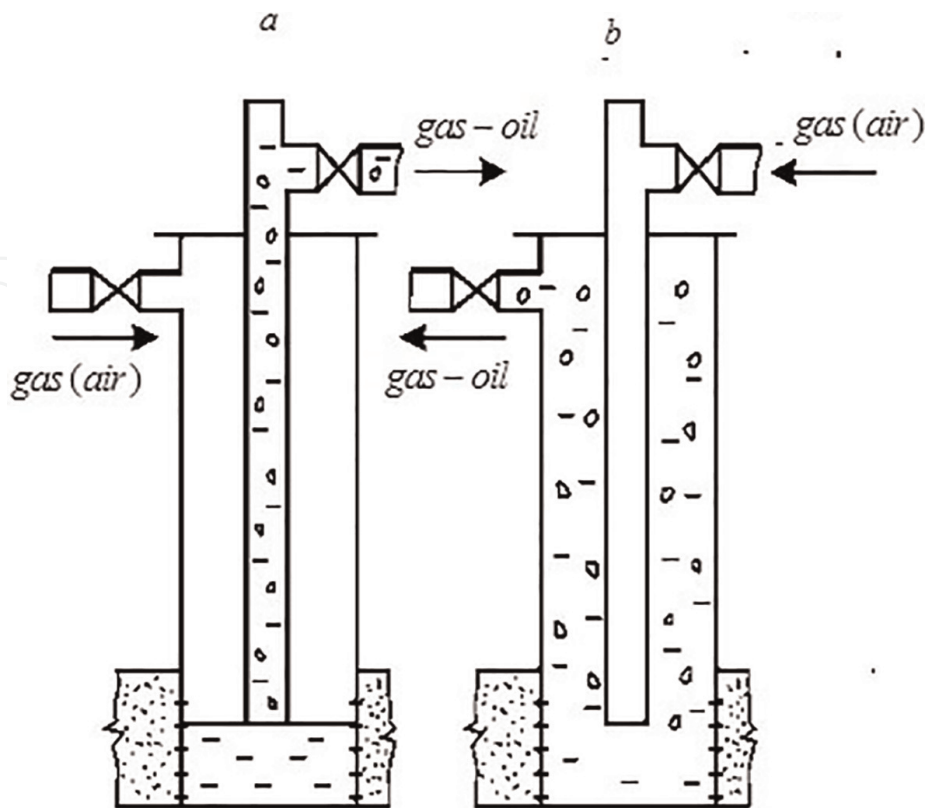
Figure 5.
Principle installations of gas lift extraction. (–)-oil, (o)-gas bubbles, (…)- sand.

compressed gas will begin to mix with the oil, resulting in a gas–liquid mixture of lower density and viscosity than the liquid coming from the reservoir, while the level in this liquid in the main pipe rises. The more gas is applied, the less dense the mixture will be and it will rise to a higher altitude. In a continuous gas injection into the well, the liquid (mixture) rises to the wellhead and pours to the surface, while a new portion of the liquid is constantly flowing from the reservoir into the well.

Despite the great advantages and disadvantages of the gas-lift method of oil extraction, significant attention is paid to improving the efficiency of this method of extraction of heavy oils, because the formation of gas–liquid mixture significantly affects the rheological properties, which is associated with a decrease in density and viscosity. In the case of a high-pressure gas supply at the gas–liquid interface, the gas is crushed into bubbles of a large range of sizes due to turbulization of the flow [24].

Research has shown that non-Newtonian oils have anomalous properties that are expressed primarily in the variability of their viscosity, their dependence on the effective shear stress, and the content of various dispersed particles (water droplets, gas bubbles, solid particles, asphalt-resinous substances). An increase in the content of high molecular weight compounds such as paraffin, resins, and asphaltenes in hydrocarbon liquids leads to the formation of spatial structural meshes and stable oil emulsions.

The composition of the oil and its various heavy fractions determine the structure of the rheological model. Currently, there are many rheological models, whose main tasks are to determine the dependence of shear stress on shear rate and identify a formula for calculating the effective viscosity of the medium. The rheological model does not reflect the essence of the process, but it is an important characteristic for evaluating the state of a complex medium.

Rheological properties of oil disperse systems are determined both by a high content of disperse particles (asphalt-resin compounds, water droplets, solids) in oil and by their ability to form different structures due to the physical interaction of particles between each other. Coagulation structures are formed by intermolecular links between particles, and if liquid interlayers remain between particles, the thickness of these interlayers significantly affects the strength of the coagulation structure. Aggregate-unstable oil systems are characterized by an unstable state of the medium due to continuous structuring and changes in the physical properties of the particles, i.e. changes in the volume and size of asphaltene particles due to their interaction, collision, coagulation and crushing at a certain concentration in a confined volume. The connection between the structure and viscosity of petroleum disperse systems, as well as the features of their non-Newtonian flow, is explained by a change in structure resulting from the emergence and collapse of aggregates of asphaltene particles. The successive coagulation or agglomeration of individual asphaltene nanoparticles into nanoaggregates and into clusters of nanoaggregates eventually form a viscoelastic framework that gives oils certain rheological properties characteristic of non-Newtonian fluids.

The structural and mechanical stability of emulsion systems is related to the formation of adsorption layers at the oil–water and oil-gas interface on the surface of water droplets and gas bubbles, whose composition consists of asphaltenes, resins, paraffins, mineral salts and solids. Analysis of the composition of adsorption films on the surface of water droplets and gas bubbles in crude oil from various fields shows that the main stabilizers are asphaltenes and resins, which are composed of high-melting paraffins and inorganic mechanical impurities. The creation and formation of an adsorption layer on the surface of water droplets and gas bubbles with elastic and

viscous properties contribute to the stabilization of oil emulsions, which significantly affects the structure formation and rheological properties of the whole oil. The mechanism for the formation of adsorption layers on the surface is determined by the diffusive transfer of mass of substance (asphaltenes) from the volume of oil to the surface of water droplets and gas bubbles. The viscosity of oil with different water content increases up to a certain value of the water volume fraction, after which it decreases. Using experimental data, the effective viscosity of an oil emulsion can be determined using the formula

$$\frac{\eta - \eta_W}{\eta - \eta_W} = \left(1 + 2.5\phi + a_0\phi^2\right)\exp\left(-a_1\phi^2\right) \tag{29}$$

here η_0, η_W - is the viscosity of oil and water. As follows from expression (E28) at high water content we have $\phi \to 1, \eta \to \eta_W$. **Figure 6** shows dependence of effective oil viscosity on water content calculated by formula (Eq. (29)) with coefficients $a_0 = 150, a_1 = 4.88$.

At low water concentrations in oil $\varphi \ll 0.1$, expression (Eq. (29) can be simplified to the form: $\eta = \eta_0(1 + 2.5\phi)$. It should be noted that the coefficients of expression (Eq. (29)) α_0, α_1 will depend on the nature and deposit of the oil.

The viscosity of oil is also significantly affected by its gas content. **Figure 7** shows a graph of the viscosity of oil. The viscosity of oil is plotted as a function of its gas content, with the viscosity of the oil/gas mixture $\varphi \ll 0.1$ calculated according to the formula: $\eta = \eta_0(1 + 2.5\phi)$. At large gas concentrations in the oil, should be used an expression such as E_{29}

$$\frac{\eta - \eta_g}{\eta_0 - \eta_g} = \left(1 + 2.5\phi + b_0\phi^2\right)\exp\left(-b_1\phi^2\right) \tag{30}$$

Here η_g- viscosity of the gas contained in the oil, b_0, b_1- coefficients determined on the basis of experimental data.

Although many advantages and disadvantages of the gas lift method of oil extraction, considerable attention has been paid to improving the technical and economic performance of gas lift wells producing non-Newtonian oil.
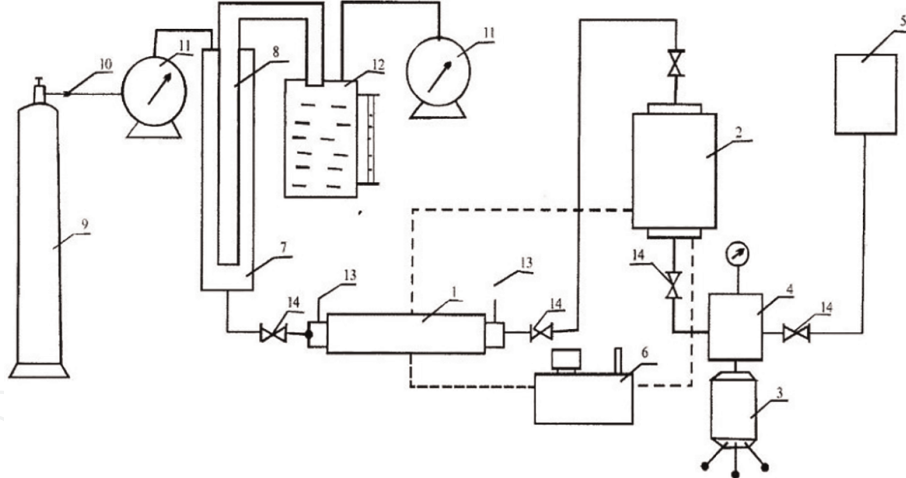
In order to develop a rheological model in non-Newtonian oil extraction by gas lift method, laboratory research was carried out on an experimental setup, the scheme of which is shown in **Figure 8**.
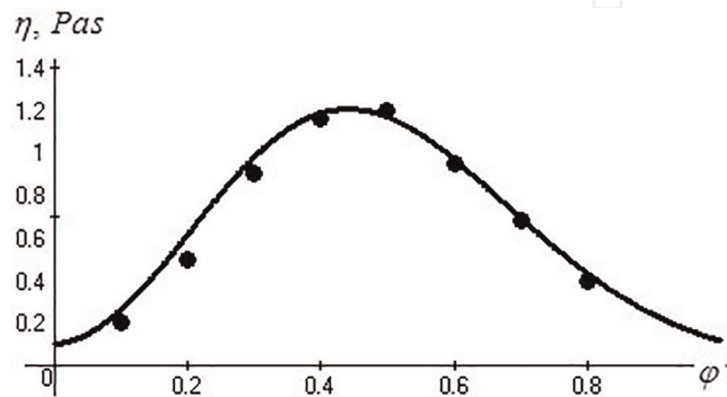
Figure 6.
The viscosity of oil emulsion at different water concentrations.
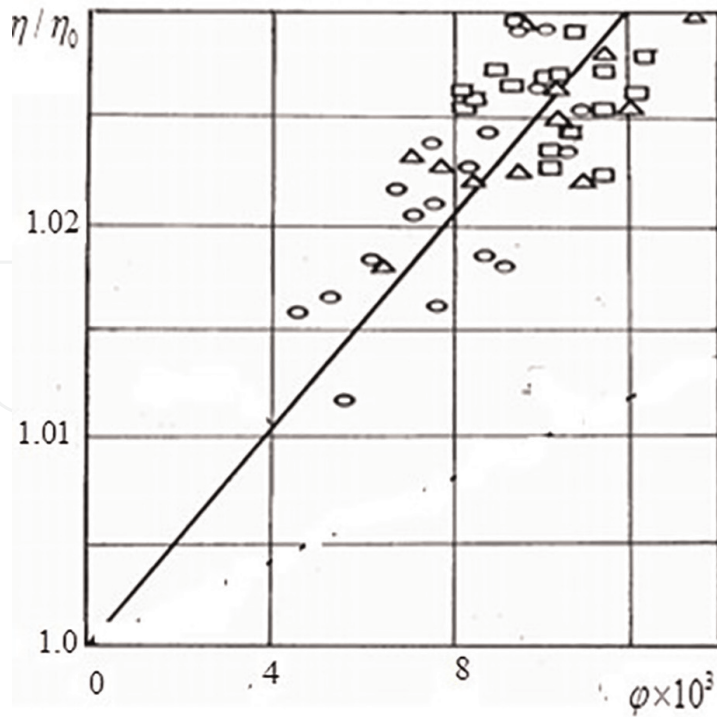
Figure 7.
Dependence of oil viscosity on low gas content for different oil.

Figure 8.
Scheme of experimental setup: 1- model of stratum (column with porous medium); 2- high-pressure capacity; 3-hand-operated press; 4-manifold; 5- fluid capacity; 6-thermostat; 7-model of gas–liquid-lift consisting of pipe $d = 8.\ 10^{-3}$ m length 3,4 m, which is located inside the pipe $d = 20.10^{-3}$ m length 3,5 m - 8; 9-gas cylinder; 10-pressure controller; 11-gas meter; 12- metering capacity; 13- model pressure gauge; 14- control valves.

As tested liquids are used a model fluid consisting of transformer oil with a density of 820 kg/m^3 and viscosity of 3.8 mPa.s and tar oil with different percentages. It is known, that viscous liquid at the addition of high-molecular compounds of vacuum residue is transformed into a non-Newtonian system. After setting the lift operating mode, both the lift capacity and the gas meter readings are measured.

Analysis of the results of the experiments shows that at the same rate of pressure change in the porous medium and lifter, degassing of viscoelastic oil in the stratum is difficult due to the non-equilibrium of this process and the oil coming into the well

containing an increased amount of formation gas, which performs useful work when lifting the liquid. Lifting of viscoelastic oils excludes such phenomena as breakthrough and slippage of compressed gas, whose work is close to the moment of piston oil squeezing. The analysis of the experimental results shows that the viscoelastic properties of the lifted fluids can be used to improve the technological processes of oil production.

On a rotary viscometer, are investigated the rheological properties of non-Newtonian oils. As a model of non-Newtonian oil, a solution of vacuum residue in transformer oil was chosen (at different percentages) with shear rates in the range $\dot{\gamma} = 100 - 1300$ s-1 and their corresponding values of shear tangential stress τ (**Table 2**).

It should be noted that the shear creeps deformation of oil media is many times greater than the elastic deformation. Rheological properties of dispersed oil media are determined by the movement of particles during volumetric change and, in particular, during structure formation, which is associated with different manifestations of rheological properties of non-Newtonian oils during shear and volumetric deformation. Shear deformation can progress over time at a decaying, constant, and progressing rate, while bulk deformation is obviously always at a decaying rate.

a) The Bingham model. Non-Newtonian fluids are primarily characterized by the fact that their character and flow patterns are predetermined by the particular

System	Density ρ, kq/м^3	Viscosity μ, mPa s	Measurement interval		η_{tf} Pa s	$1/\eta_{\text{эф}}^2$, Pa^{-2} s^{-2}	τ^2, Pa^2
			τ, Pa	γ, s^{-1}			
Transformer oil	878	10.0	-	-	-	-	-
Transformer oil +5% tar	890	12.6	9.5	364,5	0.0261	1470	90
			11.1	437,4	0.0255	1538	123
			16.3	656	0.0248	1639	266
			17.5	729	0.0240	1736	306
			31.2	1312	0.0238	1770	973
Transformer oil+10% tar	900	17.2	12.5	364.5	0.0342	855	156
			13.7	437.4	0.0313	1021	188
			17.7	656	0.0270	1371	313
			18.9	729	0.0259	1492	357
			32.5	1312	0.0248	1639	1056
Transformer oil +15% tar	915	20.5	12.7	218.7	0.0580	294	161
			13.2	243	0.0543	344	174
			14.5	364.5	0.0398	633	210
			15.2	437.4	0.0347	833	231
			20.3	656	0.0309	1048	412
			21.2	729	0.0291	1182	449
			35.4	1312	0.0270	1371	1253

Table 2.
Rheological fluid properties in different percentages (mixture of transformer oil and tar).

influence of the velocity gradient on the shear resistance. The general equation is described by which the rheological curve for a non-Newtonian viscoplastic oil:

$$\tau = \tau_0 + k \left(\frac{dV}{dy} \right)^n = \tau_0 + k \bullet \gamma^n \qquad (31)$$

here τ_0- limit of fluidity, $\dot{\gamma} = {dV}/{dy}$ - gradient of velocity, n - index of degree. Let us consider characteristic features of non-Newtonian fluids: a) viscoplastic fluids, for which $n < 1, \tau_0 = 0$. Examples of such fluids are polymer solutions, suspensions, and emulsions, including oil emulsions, oil sludge, and many oil products; b) dilatant fluids, for which $n > 1, \tau_0 = 0$.

Concentrated suspensions, pastes, etc. are examples of such fluids; c) Bingham fluids, which $n = 1, \tau_0 > 0$ and characterized by the fact that they can only flow at shear stress greater than the limit of fluidity τ_0. In most cases, crude oil exhibits Bingham's rheological properties.

Using the experimental data in **Table 2**, the rheological model can be represented as

$$\tau = \tau_0(\phi) + \eta_{eff} \dot{\gamma} \qquad (32)$$

where ϕ- is the tar content of the transformer oil.

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$$\tau = \tau_0(\phi) + \eta_{eff} \dot{\gamma} \qquad (33)$$

where ϕ- is the tar content of the transformer oil.

Figure 9 have been shown the experimental data and the calculated curve for model (Eq. (33)), using the expression $\tau_0 = 0.06\phi^{1.8}$.

b) Maxwell's model of a viscoelastic body. By connecting elastic and viscous elements in the sequence, we reach the Maxwell model characterizing the equation of state:

$$\dot{\gamma} = \frac{\tau}{\eta} + \frac{\tau}{G} \qquad (34)$$

where G- is the modulus of shear elasticity of the studied liquid; η_t- is true viscosity.

To estimate the relaxation properties, the data were processed according to the method proposed in [25] and developed in [26, 27], the essence of which is to relate the shear tangential stress (τ) to the effective viscosity (η_{eff}), in the form of a relation:

$$\frac{1}{\eta_{eff}^2} = \frac{1}{\eta_t^2} + \frac{\tau^2}{4G^2\eta_t^2} \qquad (35)$$

It should be emphasized that Eq. (E28) will hold if η_t and G take constant values, i.e. provided that the system is linear. The dependence curves $\tau = f(\dot{\gamma})$ have been recalculated in coordinates $1/\eta_{eff}^2 = f(\tau^2)$ (**Table 2**).

Based on the results of the calculation using the above formula, the dependence shown in **Figure 10** is shown. From the character of the curve, it is possible to estimate the interval of shear rates and also to identify the area of non-linearity.
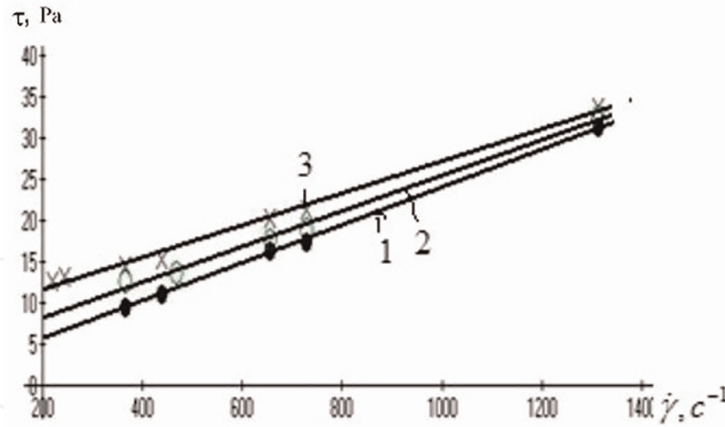
Figure 9.
Dependence of shear stress on shear rate at values φ equal to: 1–5%; 2–10%; 3–15%.
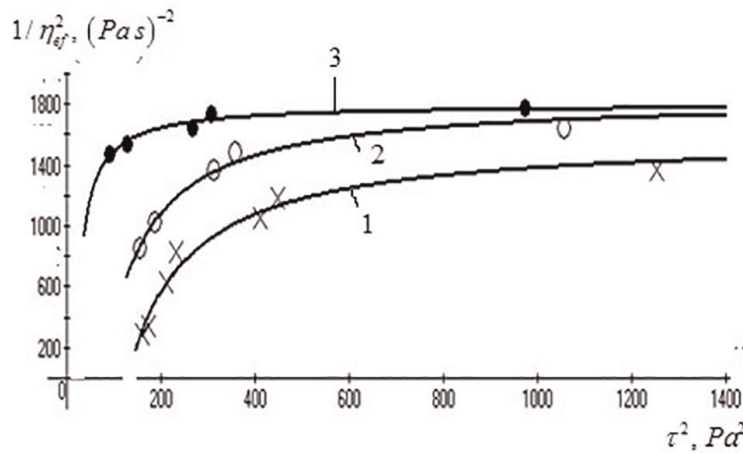
Figure 10.
Dependence $1/\eta_{eff}^2$ on τ^2 the solution transformer oil + tar at different temperatures oil + tar at different temperatures: 1-30°C; 2- 40°C; 3- 50°C.

The results of rotor viscometric tests for the solution (transformer oil + tar) at different temperatures show that the dependence sections which are parallel to the τ^2, axis correspond to viscous fluid flow and are linear in nature.

The other part of the dependencies, which correspond to non-linear sections, expresses the manifestation of the viscoelastic properties of the fluid being tested.

As the oil moves towards the bottom of the well, the pressure in its volume drops and resulting in the gas release. Furthermore, in theoretical terms, have been determined changes in pressure over time and along the radius of the stratum.

Studies have shown that fluid movement in porous media takes the greatest velocity in the bottom hole zone, although the fluid gathers maximum velocity at the walls of the lifter. Calculations of pressure reduction rate, given when non-Newtonian fluid is lifted along the wellbore, showed that the rate of pressure change during lifting decreases. As follows from expression (Eq. (25)), the manifestation of inertial forces takes place in the bottom hole zone due to a change of velocity, mainly in direction, which leads to the increasing influence of abnormal oil properties [28] (**Figure 10**).

Analysis of the results of conducted experiments shows that at the same rate of pressure change in the porous medium and lifter, degassing of viscoelastic oil in the reservoir is difficult because of the non-equilibrium of this process. The oil flowing

into the well contains increased amounts of formation gas, which performs useful work in lifting the fluid. Therefore, the specific agent flow rate in wells producing viscoelastic oil is lower than in wells with conventional viscous oil.

5. Problems of improving rheological properties in Refining heavy crude oil

Heavy oils, due to their high content of heavy components (65–70% of fractions >350 boiling point) exhibit the visco-plastic properties of non-Newtonian oils. Currently, there are no special technologies for processing heavy oils, except for some works [29, 30], because their processing is limited by problems of oil transportation in pipes, pumps, heat exchangers, and other equipment.

The viscosity of heavy oils, which determines their fluidity, depends on the content of asphalt-resinous substances, paraffin, water in the oil, and other factors. **Figure 11** shows experimental data on changes in viscosity of West Siberian oil depending on asphaltene content [31].

The existence of a spike in oil viscosity $\phi = 0.3$ when confirmed by flow-structuring phenomena that degrade the rheological properties of the oils. When refining heavy oils in operating plants at relatively low temperatures, the effective viscosity of the oil increases, which contributes to a decrease in its flowability and, in general, the productivity of the process. The reduction in viscosity of heavy oils, which ensures their transportability, depends on two factors: increasing their temperature and diluting them with lighter components.

In this regard, the main factor for improving the rheological properties of oil and its refining conditions in solving practical problems is to increase the temperature of the oil feedstock at the inlet to the unit and partially dilute it by creating recirculation of the lighter component to mix with the crude oil (**Figure 12**). This intensification of the refining process solves both problems simultaneously. Process of primary processing of oil is carried out by its preliminary heating to the certain temperature with its intermediate purification from water and salts, containing in oil, in the dehydrator 4 and the further separation in a mass-exchange rectification column 5.

A Kazakhstan heavy paraffinic oil with high viscosity was used in the installation. In order to improve the rheological properties and viscosity of the crude oil, a portion of the fraction >240°C. From column 5, the oil is returned back to mix with the crude oil after pre-cooling to pump 2. Creating a recirculation system allows for reducing the kinematic viscosity of the crude oil by increasing its temperature to 20–25°C and diluting it with lighter fluid. Experimental studies have shown a significant effect of the amount and temperature of fraction >240°C on the effective viscosity of the crude oil (**Figure 13**).

As **Figure 13** shows, the viscosity of the oil decreases as the temperature and the amount of fraction added increase. An expression describing the change in kinematic viscosity of oil with temperature at different fractional contents >240°C, can be represented as

$$v = 108.95(1 - 2.17\phi)\exp{(-0.028T)} \times 10^{-6} \qquad (36)$$

here $\phi = m_R/(m_0 + m_R)$ - is the mass fraction of fraction > 2400C in crude oil, m_0, m_R - mass flows of crude oil and fraction.
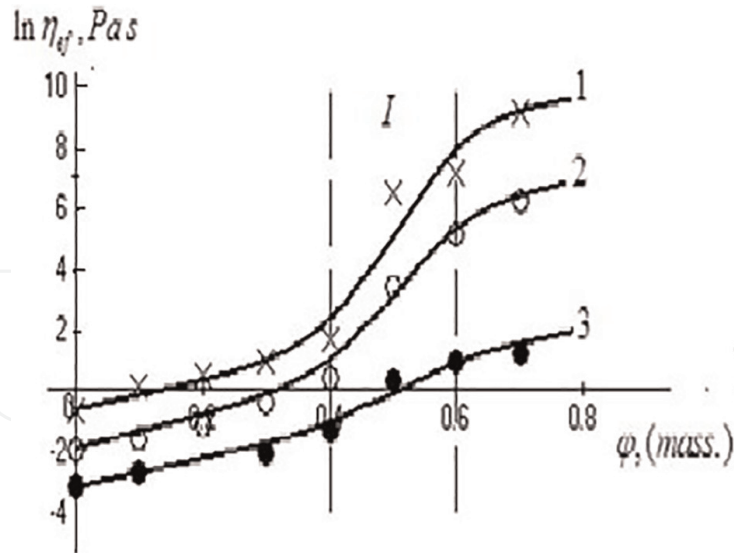
Figure 11.
Dependence of effective viscosity on asphalt-resin content at temperatures: 1–84°C; 2–112°C; 3–144°C (– area of discontinuous structure formation).
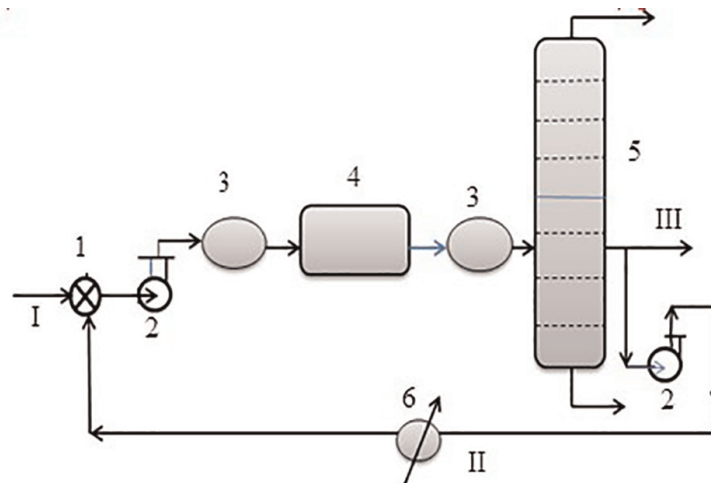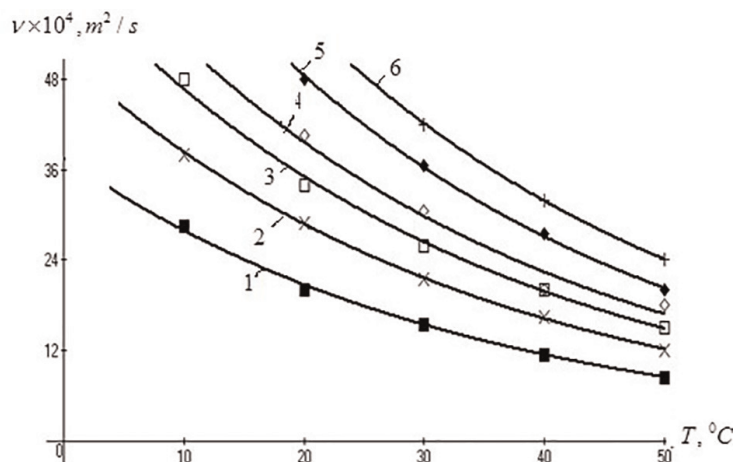

Figure 12.
Scheme of primary oil refining with recirculation: 1-mixer; 2-pump; 3-heat exchanger system; 4-dehydrator; 5-distillation column; 6-cooler; I- crude oil; II-recirculation line; III-fraction >240°C.

Recirculating some of the lighter refining fraction with higher temperature and lower viscosity to mix with the crude oil reduces the viscosity of the oil by increasing its temperature and partially diluting it with the lighter fraction (**Figure 13**).

6. Discussion of results

The problems of the rheology of non-Newtonian oil, accompanied by physical phenomena of formation and destruction of coagulation structures and aggregates, significantly affecting the flow have been considered and analyzed. Have been considered the issues of hydrodynamic interaction of particles leading to the formation of disordered structures that significantly change the rheological properties of non-Newtonian oil.

Figure 13.
Dependence of kinematic viscosity of oil on temperature at its content of fraction >240°C φ (mass fraction) equal to: 1–0.3; 2–0.25; 3–0.20; 4–0.15; 5–0.10; 6–0.05. $\nu \times 10^4, m^2/s$.

It is noted that the use of aromatic hydrocarbons to dissolve asphalt-resinous substances leads to improved rheological properties of heavy oil.

This section has considered some of the challenges in improving the rheological properties of non-Newtonian oils, both in gas lift production and transportation, and refining. Obviously, there are other ways to improve the rheological properties of oils containing high concentrations of asphalt-resinous and paraffinic substances, solid-phase particles, and dispersed water.

Nomenclature

α	particle diameter
C	concentration
c_p	specific heat capacity at constant pressure
D_T	coefficient of turbulent diffusion of a liquid
D_{eff}	coefficient of effective diffusion
m	mass of substance
R	radius of the particle
t	time
T	temperature
U	the average flow velocity
Greek letters	
β_L	coefficient of mass transfer
ε_R	specific energy dissipation per unit mass
ε	porosity
η_{eff}	effective viscosity of suspensions
η_c, η_d	dynamic viscosity of the medium and particles
λ	scale of turbulent pulsations
λ_0	Kolmogorov scale of turbulence
ν_c, ν_d	kinematic viscosity of the medium and particles
ρ_c, ρ_d	density of the medium and particles
τ_p	relaxation time
τ	shear stress

τ_0	yield strength
ϕ	the volume fraction of particles
ω	frequency of turbulent coalescence
d	a solid particle
g	is gas bubble
k	is a drop

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