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## Chapter

# Rheology of Heavy Oils

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## Abstract

The problems of heavy oil rheology, accompanied by physical phenomena of the formation and destruction of coagulation disordered structures and aggregates as a result of the hydrodynamic interaction of particles (asphaltenes, paraffins, resins, and solid-phase particles) contained in the oil, which significantly affect its properties and flow, are considered and analyzed. Rheological models of viscous-plastic heavy oils are considered and developed, consistent with a variety of experimental data. New rheological models for viscous-plastic heavy oils are proposed, which make it possible to generalize many existing models. It is noted that the variety of rheological models for heavy oils is determined by the conditions for the formation of disordered structures in the bulk of the oil flow. For heavy oils, a nonlinear equation for filtration in porous media is proposed, depending on the shear stress, pressure gradient, effective viscosity of the oil, and a number of other parameters. An analytical solution to this equation is proposed, which is consistent with the experimental data. Models for the settling rate and drag coefficient of particles in heavy oils are proposed. Applied problems of rheology aimed at improving the rheological properties of heavy oil during their processing as a result of creating a recirculation scheme at an operating oil refining unit are considered.

**Keywords:** rheology, heavy oils, effective viscosity, filtration, structure formation, asphaltenes, rheological models, production, processing, settling, drag coefficient, recycling

## 1. Introduction

Crude oil, due to the content of various particles of different nature and properties, shapes, and sizes, is an oil dispersed system, with their inherent physical and chemical phenomena of physical interaction between particles, structure formation, particle settling, and stratification of the entire system, affecting the phenomena of all types of substance transfer [1–3]. The rheology of oil and oil products is a field of science that studies and quantitatively predicts the formation and transformation of the state of oil dispersed systems over time. Heavy oils with a high content of various impurities (water, solid-phase), asphalt-resin compounds, and various paraffin are prominent representatives of media with rheological properties.

In the practice of oil production, transportation, refining, and the use of petroleum products, it is necessary to solve various, sometimes opposite problems of regulating the structural and mechanical properties of oil dispersed systems. The rheological

properties of oil dispersed systems are defined as a high content of dispersed particles (water drops, gas bubbles, and solid particles), as well as asphalt-resin and paraffin particles dissolved in heavy oil, and their ability to form various structures as a result of the physical interaction of particles with each other. An important role in structure formation and the construction of rheological models of non-Newtonian fluids is played by such factors as the size and shape of particles, their concentration, and properties. In works [1–6], a lot of rheological models are given that describe the flow of oils from various fields, characterized by different properties. As is known, the nature of the relationship between shear stress  $\tau$ , shear strain  $\dot{\gamma}$  and their changes in time  $d\tau/dt$  and  $d\dot{\gamma}/dt$  is the essence of rheological research [2–5]. Rheological parameters are used as a criterion in the classification of high-viscosity viscous-plastic oils and heavy products of its processing. The mechanism of plastic flow consists of a set of acts of rupture and restoration of contacts in aggregates and coagulation structures, between dispersed particles after overcoming the limiting shear stress  $\tau_0$  of the system under study. A special class is the problems associated with the rheology of non-Newtonian oils or oil dispersed systems containing, in addition to particles of the solid phase, liquid drops of water and gas bubbles, and asphaltenes, resins, and at low temperatures paraffin particles dissolved in oil. The presence of such a spectrum of particles of various sorts and nature leads to the formation of very complex coagulation structures, which later turn into aggregates, clusters of aggregates, up to the formation of a viscoelastic framework, at which the flow velocity is zeroed. In this case, the viscosity of the oil system increases sharply with an increase in the content of particles, and accordingly the shear stress increases to a limiting value. At values of shear stress greater than the elastic limit stress  $\tau > \tau_0$ , the structure is destroyed and the viscosity drops sharply. Rheological models of oil dispersed systems allow one to judge the fundamental properties of the systems themselves. These models, due to the lack of a common mechanism of flow and deformation, are mainly of an empirical or semiempirical nature. Despite a large number of empirical rheological models of structured disperse systems, there are currently no qualitative and quantitative theories linking the rheological properties of the system with the structure parameters. It is possible that in each particular case this is due to the nature of the flow, deformation, and formation of a disordered structure. Therefore, the results show that the same experimental study can be described by different empirical or semiempirical equations with the same model accuracy.

Heavy oils with a high content of asphalt-resinous substances are viscous-plastic liquids and are mainly described by the following rheological models [4–8]: a) Bingham model  $\tau = \tau_0 + \eta\dot{\gamma}$ , b) Ostwald-de Ville model  $\tau = k_0\dot{\gamma}^n = k_0\dot{\gamma}^{n-1}\dot{\gamma}$ , c) Hershel-Bulkley model  $\tau = \tau_0 + k_0\dot{\gamma}^n$ , and other modified type equations, where  $k_0$  is the consistency coefficient and  $\dot{\gamma}$  is the shear rate.

The purpose of this work is to build a rheological model of heavy oils, accompanied by structure formation by particles contained in the volume, free settling of particles, and oil filtration through anisotropic porous media.

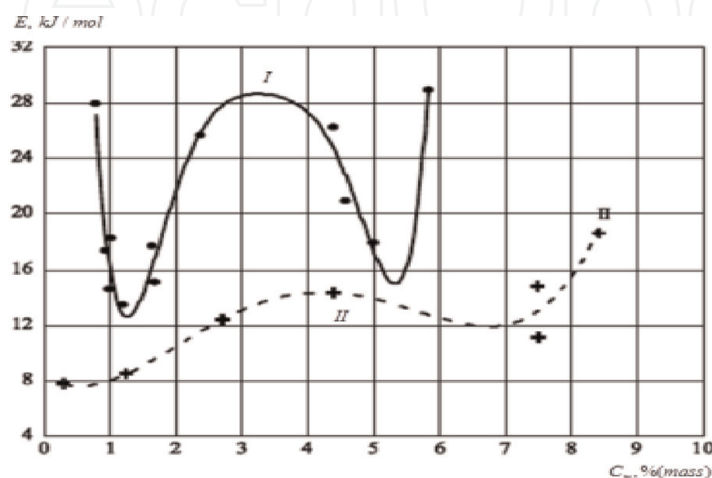
## 2. Structure formation in oil dispersed media

Coagulation structures are formed due to intermolecular bonds between particles, and if liquid interlayers remain between the particles, then the thickness of this interlayer significantly affects the strength of the coagulation structure. Aggregative unstable oil systems are characterized by the variability of the state of the

environment, due to continuous structure formation and changes in the physical properties of particles, that is, a change in the volume and size of asphaltene particles as a result of their interaction, collision, coagulation, and crushing at a certain concentration in a closed volume. The relationship between the structure and viscosity of petroleum dispersed systems, as well as the features of their non-Newtonian flow, are explained by a change in the structure as a result of the formation and destruction of aggregates from asphaltene particles in the presence of resins. Oil structured systems containing crystals of high molecular weight paraffin, resins, and asphaltene particles, and at very low laminar flow rates or in the absence of flow form a chain or, in the limiting case, a continuous grid (framework). Sequential coagulation or agglomeration of individual asphaltene nanoparticles into nanoaggregates and clusters of nanoaggregates eventually leads to the formation of a viscoelastic framework that imparts certain rheological properties to heavy oils. The paper notes that real oil dispersed systems are classified according to activation energies into two structural groups that differ in the nature of the intermolecular interaction of particles in an oil disperse medium. These groups differ from each other in the content of asphaltenes and resins, and they can be classified into immobile with a low content of asphaltenes and interacting with a high content of asphaltenes. **Figure 1** shows the characteristic changes in activation energies for the two indicated groups.

The rheological model of the flow of oil dispersed systems can be based on the following assumptions [2, 7]:

- In structured oil system, there are nanoaggregates that have arisen as a result of collision, coagulation, and aggregation of asphaltene particles due to diffusion in laminar and turbulent shear flow and sedimentation (gravitational coagulation), the formed asphaltene aggregates can be deposited on the surface, forming a rather thick layer of deposits on the walls of the porous oil reservoir. Moreover, pressure drops depending on temperature can lead to redissolution or detachment of particles of precipitated asphaltenes during intensive mixing or turbulent flow;
- Nanoaggregates move as independent flow units until they collide with other similar aggregates or asphaltene particles;



**Figure 1.**  
The dependence of the activation energy on the content of asphaltenes for the immobile group (I) and interacting group (II) [2].

- Nanoaggregates, when colliding with each other, unite into clusters of nanoaggregates and then create a viscoelastic frame of a disordered structure with the highest possible viscosity and a loose coagulation structure. The maximum size of the framework of nanoaggregates is determined by the dimensions of the channels (pores and pipes) through which the flow flows. It is important to note that the formation of disordered structures in the volume of oil is the reason for the diversity of rheological models;
- Nanoaggregates can rotate in a gradient field and break under the action of tensile hydrodynamic forces depending on the pressure gradient or flow rate;
- The linear dimensions of nanoaggregates are in the range of the size of an individual asphaltene particle up to the maximum size of a cluster or framework of a disordered structure;
- In the limiting case of infinite velocity, all aggregates, under the condition  $\lim_{\tau \rightarrow \infty} (\tau_0/\tau) \rightarrow 0$ , are destroyed to individual particles as a result of which the flow of a dispersed system approaches Newtonian;
- In the presence of aromatic hydrocarbons, asphaltenes dissolve well, thereby preventing structure formation, i.e. formation of clusters and viscoelastic framework. The solubility of asphaltenes is affected by the presence of other compounds contained in the oil, such as resins.

The rheological equation of Maxwell's viscoelastic fluid in substantial derivatives is written as:

$$\lambda \left( \frac{\partial \tau}{\partial t} + U \frac{\partial \tau}{\partial y} \right) + \tau = \eta_c \dot{\gamma} \quad (1)$$

$$t = 0, \quad \tau = \tau_0, \quad \dot{\gamma} = 0$$

where  $\lambda$  is the relaxation time.

The Eq. (1), presented in the form :

$$\lambda \left( \frac{\partial \tau}{\partial t} + U \frac{\partial \tau}{\partial y} \right) + \tau = 0 \quad (2)$$

The solution of Eq. (2) can be represented as:

$$\tau = C_1 f(y - Ut) \exp(-t/\lambda) \quad (3)$$

Substituting this solution into Eq. (2), we obtain an identity. Here,  $\lambda = \eta_c/G$  is the Maxwell relaxation time,  $U$  is the strain front movement velocity,  $f(y - Ut)$  is a function that determines the strain front in the frame,  $y$  is the coordinate,  $G$  is shear modulus,  $\dot{\gamma} = d\gamma/dt$  is shear rate,  $\gamma$  is the shear gradient,  $\tau_0$  is the ultimate shear stress or yield strength. Moreover, if  $\tau \leq \tau_0$ , then  $\dot{\gamma} = 0$ . The complete solution of Eq. (3) takes the form:

$$\tau = C_1 f(y - Ut) \exp(-t/\lambda) \tau_0 \quad (4)$$



or this equation can be written in logarithmic form:

$$\ln \tau = \ln \tau_0 - t/\lambda + \ln(C_1 f(\dot{\gamma} - Ut)), \quad \tau_0 = \eta_c \dot{\gamma} \quad (5)$$

It is obvious that the value  $t/\lambda$  in Eq. (5) characterizes the deformation of the viscoelastic frame in time and, with the flow of heavy oils, depends on the velocity or pressure gradient, which in the approximation can be represented as:  $t/\lambda = t\dot{\gamma}/Wi \sim f[(\text{grad}P/(\text{grad}P)_0)^n]$  (where  $Wi = \lambda\dot{\gamma}$  is the Weissenberg number).

### 3. Rheology of viscoplastic heavy oils

Heavy oils with constant differential viscosity are characterized by viscous-plastic properties. Bingham viscous-plastic fluids include petroleum dispersed systems, polymeric fluids, many types of food materials, cement mortars, oil paints, and others that exhibit viscous, plastic, and highly elastic properties. They differ from conventional liquids in that some finite stress is required to initiate flow. The rheology of viscoplastic fluids is described by the Bingham equation.

$$\tau = \tau_0 + \eta\dot{\gamma}, \quad \dot{\gamma} > 0 \quad (6)$$

Moreover, if  $\dot{\gamma} = 0$ , then  $\tau \leq \tau_0$ . In this equation, the shear rate can be represented as the flow velocity gradient:

$$\dot{\gamma} = \frac{d(dx/dy)}{dt} = \frac{d(dx/dt)}{dy} = \frac{dV_x}{dy} \quad (7)$$

Based on expression (6), the viscosity of viscous-plastic oil can be determined as:

$$\eta = \frac{\tau - \tau_0}{\dot{\gamma}} \quad (8)$$

However, some viscous-plastic fluids exhibit properties that do not obey Eq. (6). Such fluids are usually described by other nonlinear rheological equations of the Ostwald-de Ville or Herschel-Bulkley type, which are widely used to describe the flow of plastic fluids, heavy petroleum oils, and petroleum dispersions. This is explained by the presence of various particles of the dispersed phase in the liquid, and therefore, such systems are prone to the formation of coagulation structures up to the formation of a cluster of aggregates and a framework under the condition  $\tau < \tau_0$ .

In addition, the coefficients included in the rheological equation will depend on the concentration, size, and properties of particles, as well as on temperature and many other parameters.

Currently, there is no consensus on the mechanism of non-Newtonian flow of oil dispersed systems, and therefore, the set of flow equations  $\tau(\dot{\gamma}^*)$  or rheological viscosity equations  $\eta(\dot{\gamma}^*)$  or  $\eta(\tau)$  used in practice are mainly empirical or semiempirical. However, despite a large number of works and a variety of approaches in the field of rheology of structured disperse systems, including oil disperse systems, there is still no satisfactory quantitative theory linking the rheological properties of bodies with their structural parameters. It is possible that this is due to the formation of various

disordered structures that affect the type of rheological model and the nature of the dependence of the effective viscosity of the disperse system on stress and shear rate.

Let us assume that the change in relative viscosity is proportional to linear deformation in a nonlinear form:

$$\frac{\Delta\eta}{\eta} \sim m_0 \left( \frac{\Delta x}{\Delta y} \right)^p \quad (9)$$

where  $\Delta x$  is the increment of linear deformation with a change in  $\Delta y$  and  $m_0$  is the coefficient of proportionality.

In the limiting case  $\Delta y \rightarrow 0$ , passing to the differential form, we take the nonlinear form of the expression (9)

$$\frac{d\eta}{\eta} = m_0 \left( \frac{dx}{dy} \right)^{p-1} d \left( \frac{dx}{dy} \right) \quad (10)$$

Here  $p$  is the exponent characterizing the degree of nonlinearity. Assuming that  $dx/dy = \lambda dV_x/dy = \lambda \dot{\gamma}$ , we have:

$$\frac{d\eta}{\eta} = m_0 (\lambda \dot{\gamma})^{p-1} d(\lambda \dot{\gamma}) \quad (11)$$

Taking into account the initial and infinite viscosity, the solution, Eq. (11) can be written as:

$$\eta - C = (\eta_0 - C) \exp \left( \frac{m_0}{p} (\lambda \dot{\gamma})^p \right) \quad (12)$$

where  $\lambda$  is relaxation time. Moreover, if  $\dot{\gamma} \rightarrow \infty$ , then  $C = \eta_\infty$ . As a result, the dependence of the viscosity of fluid on the shear stress in the general case for  $m_0 = 1 - n$ , we obtain the dependences of the viscosity on the shear rate for a dilatant ( $n < 1$ ), viscous-plastic fluid ( $n > 1$ ), and a Newtonian fluid ( $n = 1$ ) in the form.

$$\begin{aligned} \frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} &= \exp \left( \frac{m_0}{p} (\lambda \dot{\gamma})^p \right), \quad n > 1, \quad m_0 > 0 \\ \frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} &= \exp \left( -\frac{m_0}{p} (\lambda \dot{\gamma})^p \right), \quad n < 1, \quad m_0 < 0 \\ \eta &= \eta_0, \quad n = 1, \quad m = 0 \end{aligned} \quad (13)$$

We represent the last expression in the form:

$$\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = \exp \left( -\frac{m_0}{p} (\lambda \dot{\gamma})^p \right) = \frac{1}{[\exp((\lambda \dot{\gamma})^p)]^{m_0/p}} \quad (14)$$

Expanding the exponent in a series  $\exp((\lambda \dot{\gamma})^p) = 1 + (\lambda \dot{\gamma})^p + \frac{1}{2}(\lambda \dot{\gamma})^{2p} + \dots$ , we get

$$\frac{\eta - \eta_\infty}{\eta_0 - \eta_\infty} = \frac{1}{\left[ 1 + (\lambda \dot{\gamma})^p + \frac{1}{2}(\lambda \dot{\gamma})^{2p} \right]^{m_0/p}} \quad (15)$$

Most of the formulas for determining the viscosity of a viscous-plastic fluid, taking into account the linear expansion of the exponential, are based on Ref. [12]. In particular, for describing the rheology of viscous-plastic polymeric fluids, the most effective is the Carreau-Yasuda rheological model [9, 10] presented in the form:

$$\frac{\eta - \eta_{\infty}}{\eta_0 - \eta_{\infty}} = [1 + (\lambda\dot{\gamma})^p]^{(n-1)/p} \quad (16)$$

Here,  $\eta_0, \eta_{\infty}$  are the value of the fluid viscosity at the beginning and at infinity,  $\lambda$  is the relaxation time, and  $p$  is a dimensionless coefficient characterizing the transition from the region with initial viscosity to the region with final viscosity. As noted in Ref. [9], Eq. (16) describes the rheology of polymeric liquids at various concentrations and temperatures. In Ref. [9], a simpler form is also considered for describing viscous-plastic fluids at  $\eta_{\infty} = 0$

$$\eta = \frac{\eta_0}{1 + \lambda\dot{\gamma}} \quad (17)$$

In Ref. [11] for viscous-plastic fluids, Bingham or Herschel–Bulkley proposed the following rheological models.

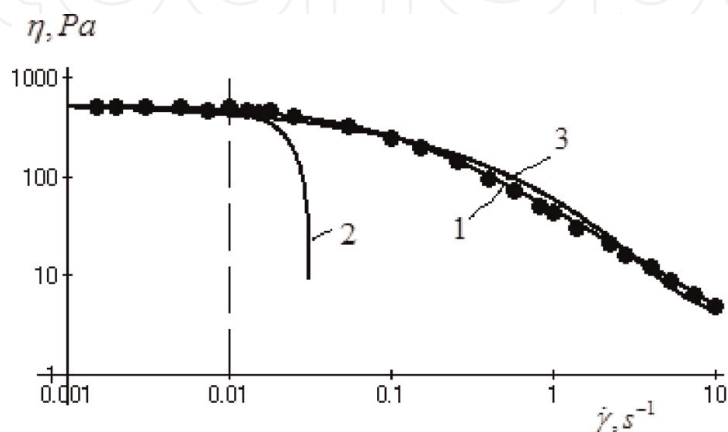
$$\frac{\eta - \eta_{\infty}}{\eta_0 - \eta_{\infty}} = \frac{1}{1 + \left(\frac{|\dot{\gamma}|}{\dot{\gamma}^m}\right)^m} \quad (18)$$

$$\eta = \begin{cases} \tau_0 + k_0|\dot{\gamma}|^m, & \tau > \tau_0 \\ \infty & \tau \leq \tau_0 \end{cases} \quad (19)$$

It is also important to note the dependence of viscosity on shear stress in the following empirical formula (13)

$$(\eta - \eta_{\infty})/(\eta_0 - \eta_{\infty}) = \exp(-(\lambda\dot{\gamma})^n) \quad (20)$$

Figure 2 shows the dependence of viscosity on the shear rate for some viscous-plastic fluid.



**Figure 2.** Dependence of fluid viscosity on shear rate for various equations: 1-(17),  $\eta_0 = 500 \text{ Pa}\cdot\text{s}$ ,  $\lambda = 10 \text{ s}$ ; 2- $\eta = \eta_0 - b\dot{\gamma}^2$  [1]; 3- $(\eta - \eta_{\infty})/(\eta_0 - \eta_{\infty}) = \exp(-2.3\dot{\gamma}^{0.45})$ .



In [13], the viscosity of a non-Newtonian polymer fluid containing particles of various concentrations is determined depending on the shear stress in the form (Figure 3).

$$\eta = \frac{\eta_0}{1 + (m \tau)^n} \quad (21)$$

It should be noted that the coefficients  $\eta_0, m, n$  included in Eq. (12) depend on the concentration (volume fraction), particle material, and temperature.

Figure 4 shows the curves describing the experimental values [11] of changes in the viscosity of polymeric liquids by Eqs. (15) and (16).

The above calculations and comparison with experimental data allow us to confirm the correctness of the accepted hypothesis about the proportionality of the relative viscosity of deformation (9).

In principle, rheological models for various flows of a non-Newtonian fluid do not obey physical laws but are empirical and semiempirical approximations and formulas that describe flow curves in a certain range of shear rates. Experimental measurements are approximated by certain approximate empirical or semiempirical equations, and the choice of the most convenient of them is largely determined by the

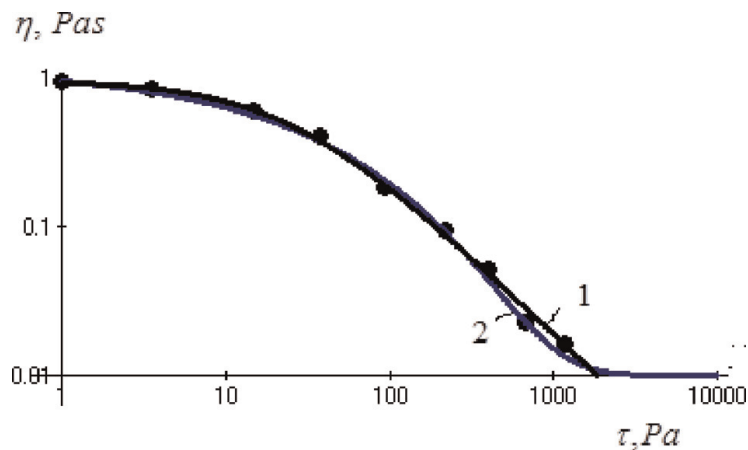


Figure 3.

Dependence of viscosity on shear stress for various equations: 1-  $\eta = 0.95 / (1 + (0.046\tau)^{1.055})$ ; 2-  $\eta = 1.15 \exp(-0.21\tau^{0.47}) + 0.01$ .

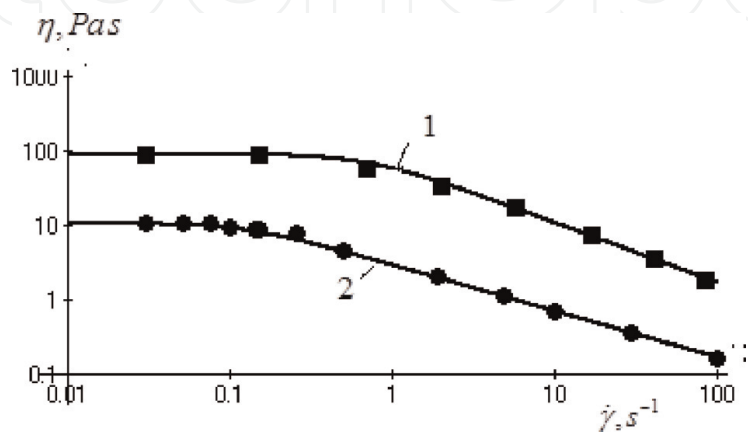


Figure 4.

Dependence of viscosity of polymeric fluids on shear rate: 1-0.75% polyacrylamide in 95/5 mixture by weight of water and glycerin ( $p = 2, m_o/p = 0.4$ ); 2-7% aluminum soap in decalin and *m*-cresol ( $p = 2, m_o/p = 0.3$ ).

degree of maximum approximation of calculated and experimental measurements or by the simplicity of the formulas used when solving applied problems. tasks.

#### 4. Dependence of the effective viscosity of oil on the content of asphaltenes

An experimental study of the influence of the content of asphaltenes and resins in oil on its rheological properties and viscosity was proposed in the works [13–15].

Using the results of these studies, it can be noted that the presence of asphaltenes, resins, and paraffins in oil, which change the properties of the oil, significantly affects their movement and transport. First of all, this is reflected in the stress and shear rate and the increase in the viscosity of non-Newtonian oil. **Figure 5** suggests the dependence of the effective viscosity of Iranian oil on the shear rate by various rheological models [16].

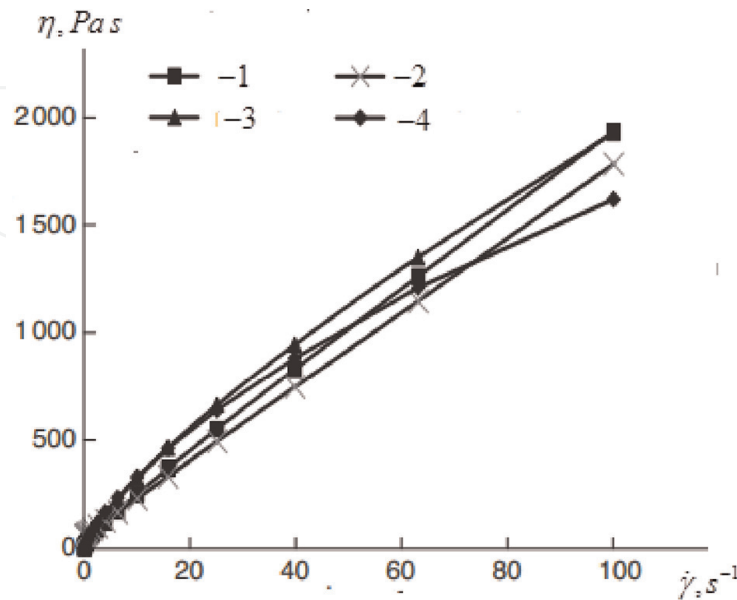
The **Table 1** shows the values of the main coefficients included in these rheological models at various temperatures [16].

Of all the models, a satisfactory approximation to the experimental data is given by the expression.

$$\eta = 45.86\dot{\gamma}^{0.75} \quad (22)$$

Given this expression, the rheological dependence that satisfies the experimental data can be represented as:

$$\tau = \tau_0 + 45.76\dot{\gamma}^{1.75} \quad (23)$$



**Figure 5.** Approximation of the dependence of viscosity on shear rate by various rheological models: 1 - Casson model –  $\tau = \left(\tau_0^{1/2} + |k|^{1/2}\dot{\gamma}^{1/2}\right)^2$ ; 2 - Bingham model –  $\tau = \tau_0 + \eta\dot{\gamma}$ ; 3 - exponential function –  $\tau = k\dot{\gamma}^n$ ; 4 - experiment.

Model	Temperature								
	25°C			45°C			60°C		
	$\tau_0, Pa$	$k, Pas$	$n$	$\tau_0, Pa$	$k, Pas$	$n$	$\tau_p, Pa$	$k, Pas$	$n$
Casson	8.13	4.11	–	0.57	2.2	–	0.15	1.53	–
Power law	–	54.65	0.77	–	9.26	0.88	–	3.86	0.9
Bingham	61	17.23	–	8.66	4.81	–	3.13	2.33	–

**Table 1.** Coefficients of rheological models at temperatures  $T = 25^\circ C, 45^\circ C$  and  $60^\circ C$ .

The dependence of the consistency coefficient on temperature can be expressed by the following equation.

$$k = 399.2 \exp(-0.081T) \tag{24}$$

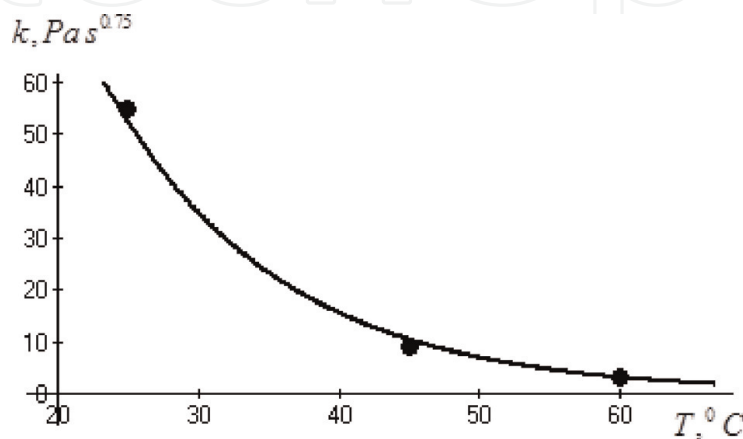
**Figure 6** shows the dependence of the consistency coefficient on temperature.

The dependence of oil viscosity on the content of asphaltenes (% wt.) in oil using experimental data is expressed by the formula (**Figure 7**).

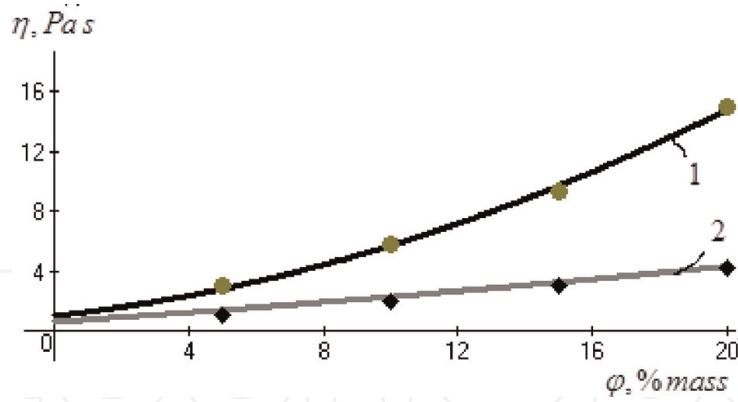
$$\eta_{eff} = \eta_0(1 + 0.25\varphi + k_0\varphi^2) \tag{25}$$

where  $\varphi$  is the volume fraction of asphaltenes in oil.  $\eta_0$  is viscosity of oil in the absence of asphaltenes.  $\eta_{eff}$  is effective viscosity of oil. Provided that  $\varphi < 10\%$  this expression coincides with the Einstein formula.

In Ref. [17], similar studies were carried out for West Siberian oils for the concentration of asphaltenes in oil from 4 to 72% (wt.). This paper presents experimental studies of the effective viscosity of non-Newtonian oil on the content of asphaltenes at various temperatures. As follows from **Figures 7–9**, the region transitions from Newtonian to non-Newtonian properties of oil as the content of asphalt-resinous substances  $I$  increases are limited by a stepwise change in oil viscosity for all temperatures. Obviously, this is due to the fact that with an asphaltene content of 38–46% (wt.) in West Siberian oil, there is a stepwise change in the effective viscosity of oil, structural and mechanical strength, transition temperature to the state of a non-Newtonian liquid and molecular weight, which is due to the formation of coagulation structures, aggregates



**Figure 6.** The dependence of the consistency coefficient on temperature.



**Figure 7.** The dependence of the viscosity of oil on the content of asphaltenes at temperatures: 1 – 25°C, ( $k_o = 0.022$ ); 2 – 45°C, ( $k_o = 0.003$ ).

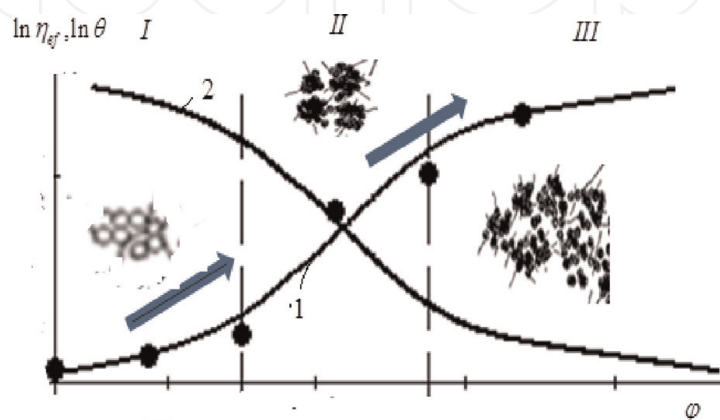
up to the frame throughout the volume. A stepwise change in viscosity during periods of structure formation and destruction of the structure is a characteristic feature of non-Newtonian oils, which complicates the nature of the description of the entire viscosity and mobility curve of the oil system. The process of formation of coagulation structures is associated with an increase in the probability of interaction and collision of particles with an increase in their concentration in the volume. In the following section, the problems of coagulation, coalescence of droplets and bubbles, and many issues related to the solution of this problem will be discussed in detail.

As follows from **Figure 8**, when structure formation and asphaltene concentration increase, the mobility or fluidity of the oil system decreases, and the fluidity of the system is defined as:

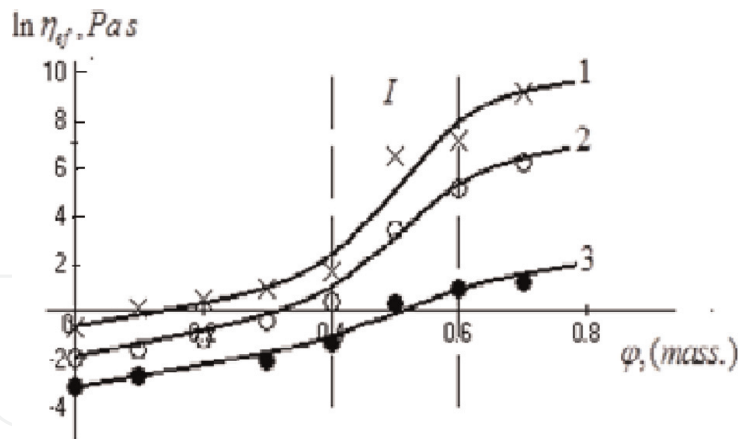
$$\ln \theta = \frac{\ln \eta_{eff\infty}}{\ln \eta_{eff}} \quad (26)$$

where  $\theta$  is the fluidity of the medium. **Figure 9** shows experimental data on the change in the viscosity of West Siberian oil depending on the content of asphaltenes [19].

The equation describing the experimental data on the viscosity of oil in large intervals of asphaltene content change is presented in the form:



**Figure 8.** Characteristic stages of structure formation in oil depending on the content of asphaltenes: I - oil dispersed system; II - area of formation of structures; III - structured oil system, 1 - viscosity; 2 - fluidity.



**Figure 9.** Dependence of effective viscosity on the content of the dispersed phase of tar-asphaltenes at temperatures: 1 – 84°C; 2 – 112°C; 3 – 144°C.. (I-is the region of stepwise change structure formation).

$$\ln \eta_{eff} = \ln \eta_{eff 0} + b_0 \varphi + b_1 \delta(\varphi) + b_2 (1 - \exp(-b_3 \varphi^6)) \quad (27)$$

Here,  $\varphi$  is the mass fraction of asphaltenes in oil,  $b_0 - b_3$  is coefficients determined experimentally and depending on temperature,  $b_1 = 0.2$  is the maximum value of the delta function,  $\delta(\varphi)$  is delta function, determined in the form:

$$\delta(\varphi) = \frac{1}{\exp(72.5(\varphi - 0.45)) + \exp(-72.5(\varphi - 0.45))} \quad (28)$$

$\mu_0$  - initial viscosity,

$$\eta_{eff 0} = 2.05 \times 10^{-8} \exp\left(\frac{6075}{T + 273}\right) \quad (29)$$

The value of the delta function characterizes the viscosity jump in the region of structure formation. In particular, the main property of the delta function is the following:

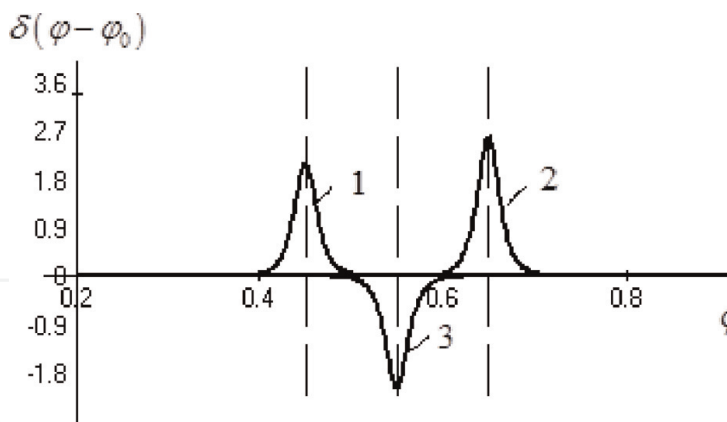
$$\delta(\varphi - \varphi_0) = \begin{cases} 0, & \varphi \neq \varphi_0 \\ \infty, & \varphi = \varphi_0 \end{cases} \quad (30)$$

The partial approximation expression of the delta function can be represented as:

$$\delta(\varphi) = \frac{1}{\exp(b_1(\varphi - \varphi_0)) + \exp(-b_2(\varphi - \varphi_0))} \quad (31)$$

Here,  $b_1, b_2$  are the coefficients that determine the width of the base of the delta function,  $\varphi_0$  is the coordinate of the jump center. **Figure 10** shows different kinds of delta functions.

Thus, the use of the delta function makes it possible to describe all the stepwise phenomena occurring during the formation and destruction of structures in non-Newtonian oil. At the same time, satisfactory results are obtained by using a higher-order exponential function, which makes it possible to obtain a soothing effect in the region of the jump.



**Figure 10.**  
 Delta functions: 1,2 - positive values of the function  $\delta(\varphi) > 0$  with centers  $\varphi_0 = 0.45, 0.65$ ; 3 - negative values of the function  $\delta(\varphi) < 0$  with the center  $\varphi_0 = 0.55$ .

The use of aromatic and other solvents partially dissolves asphaltenes, thereby reducing or eliminating the formation of coagulation structures, which improves the rheological properties of oil dispersed media. As follows from **Figure 9**, for this oil, provided that the asphaltene content is less than  $\varphi < 0.4$ , the formation of coagulation structures is excluded, although for different oil fields there may be other conditions. Analysis of various studies on the effect of asphalt-resin substances on the rheology of non-Newtonian oil of various fields leads to conflicting results, although in all cases an increase in viscosity is observed as a result of structure formation. It should be noted that, in addition to asphaltenes, the rheological properties of oil dispersed system are affected by the content of water and solid-phase in it.

## 5. Nonlinear equation of filtration of heavy oils in porous media

Oil structured systems containing coagulation structures of crystals of high molecular weight paraffin and asphaltene particles and forming a chain or, in the limiting case, a continuous network (framework), acquire the ability to flow only after the destruction of this network at  $\tau > \tau_0$  (where  $\tau_0$  is the yield strength), and small external stresses produce elastic deformation mesh or frame. The interaction of asphaltene particles is accompanied by the creation of sufficiently strong aggregates of a coagulation nature, and above all, doublets and triplets, due to the Brownian diffusion motion of individual particles.

These structures disintegrate into individual particles as a result of the fragmentation of aggregates under the action of shear flow, and the equilibrium shifts towards the formation of individual particles as the shear rate increases. The frequency of collisions of two asphaltene particles in the volume as a result of Brownian diffusion is determined by the following expression [3, 18].

$$\omega = 4\pi(D_1 + D_2)(R_1 + R_2)N_0 \quad (32)$$

The formation of aggregates from asphaltene particles also occurs in pipelines with an intense turbulent flow of oil. The frequency of particle collisions in an isotropic turbulent flow of oil in pipes is determined by the turbulence parameters, the capture



coefficient, and the physicochemical properties of oil and asphaltenes. In refs. [2, 3, 18], an expression for the frequency of coagulation and fragmentation of particles in an isotropic turbulent flow due to turbulent diffusion of particles is given in the form:

$$\omega(a) = C_{01}N_0a^3 \left(\frac{\varepsilon_R}{\nu_c}\right)^{\frac{1}{2}} \exp \left[ -C_{02} \frac{\sigma}{(\nu_c\varepsilon_R)^{\frac{1}{2}}a\rho_c} \right] \quad (33)$$

It follows from this expression that the higher the oil viscosity, the lower the frequency of collisions and the lower the probability of formation of coagulation aggregates. Asphaltene particles become larger as a result of coagulation, reach the maximum size of an unstable aggregate, after which their crushing begins. It can be assumed that under the action of hydrodynamic forces, all bonds between particles in an aggregate are stretched to a critical value, as a result of which this aggregate primarily breaks up into smaller aggregates, and then secondary, tertiary etc. decomposition occurs up to a single particle. The destruction of coagulation structures formed by paraffins and asphaltenes is characterized by the fact that after applying a certain load for oil injection, no immediate destruction is observed. In the limiting case of infinite shear rate  $\tau > \tau_0$ , aggregates can be completely destroyed up to a single particle, and the flow of such oils or oil emulsions can be considered as the flow of Newtonian fluids. Since the effective viscosity  $\eta_{eff}(T, \tau)$  depends on the temperature and on the shear rate (pressure gradient), additional energy will be required to destroy the structure. It should be noted that after the load is removed, the strength of the structure spontaneously recovers completely and the viscosity reaches its maximum value, that is, a certain thixotropy is observed. Thixotropic properties are most typical for structured nonstationary dispersed petroleum systems characterized by aggregative instability and accompanied by aggregation and coagulation of dispersed asphaltene particles. It should be noted that oils and oil products with a high content of asphalt-resinous substances are characterized by thixotropic properties, which are usually described by the Herschel-Bulkley equation and the Ostwald-de Ville power-law, and at high shear rates by the Bingham equation, which is the main equation for describing heavy oils. The equations for the thixotropic behavior of such systems can be represented by various dependencies of the shear stress on time. At the same time, it should be noted that despite the large number of publications offering various approaches to the rheology of structured oils, there is still no satisfactory theory. Linking the rheological coefficients with the structural rheological properties of oil, namely, the forces of interaction between asphaltene particles, the structure, and size of aggregates.

Anomalous viscous-plastic oils differ in their properties from ordinary oils and their rheological description obeys the laws of flow of non-Newtonian Bingham fluids.

$$\tau = \tau_0 + \eta_{eff}\dot{\gamma} \quad (34)$$

From the Newtonian equation ( $\eta_j \tau_\gamma$ ) and taking into account Eq. (34), we obtain:

$$\eta = \eta_{eff} + \frac{\tau_0}{\dot{\gamma}} = \eta_{eff} \left( \frac{\tau_0}{\eta_{eff}\dot{\gamma}} + 1 \right), \quad (35)$$

Determining from expression (34)

$$\eta_{eff}\dot{\gamma} = \tau - \tau_0 \quad (36)$$

we finally obtain an expression for the effective viscosity in the form:

$$\eta = \eta_{eff} + \frac{\tau_0}{\dot{\gamma}} = \eta_{eff} \frac{\tau}{\tau - \tau_0}, \quad (37)$$

from which it follows that with increasing  $\tau$  the value of  $\mu$  decreases and in the limit at  $\tau \gg \tau_0$ , the value of  $\eta \rightarrow \eta_{eff}$ , corresponding to a system with a completely destroyed structure  $\tau_0/\tau \rightarrow 0$ . Thus, the viscosity of a structured system in the course of flow under the action of increasing shear stress changes from  $\eta_0$ , corresponding to an undamaged structure, to  $\eta_\infty$ , characteristic of a completely destroyed structure. Substituting Eq. (37) into expression (34), we obtain a nonlinear filtration equation for a structured oil system [3, 4].

$$V = -\frac{k_p}{\eta_{eff}} \left(1 - \frac{\tau_0}{\tau}\right) \frac{\partial P}{\partial x} \quad (38)$$

when  $\tau \gg \tau_0$  this expression goes into the usual Darcy equation for unstructured oil. An analysis of experimental data on the filtration of non-Newtonian oils made it possible to approximate the ratio  $\tau_0/\tau$  in the form:

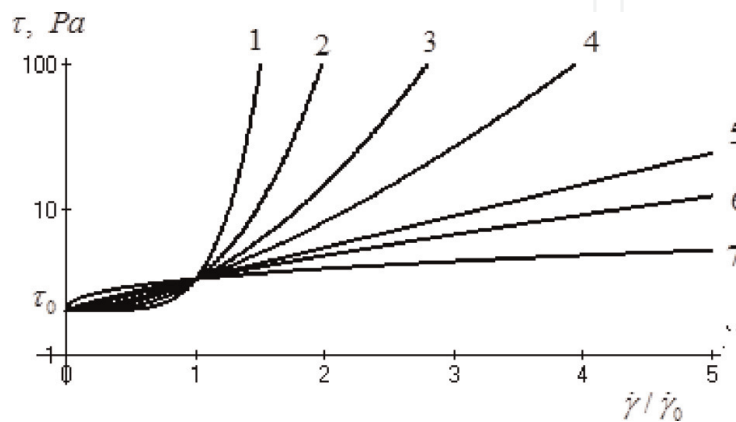
$$\ln \frac{\tau}{\tau_0} = \alpha \left( \frac{\text{grad} P}{(\text{grad} P)_0} \right)^n \quad (39)$$

Here,  $\alpha$ — is the coefficient determined on the basis of experimental data,  $n$  is the exponent. The rheological equation can be written in the form (**Figure 11**)

$$\tau = \tau_0 \exp(\alpha(\dot{\gamma}/\dot{\gamma}_0)^n) \quad (40)$$

Expression (40) can be considered as a new rheological equation describing the viscoplastic flow of non-Newtonian oils.

Obviously, the index  $n$ , depending on the temperature and properties of the porous layer, characterizes the complete destruction of the structure. To date, many concepts



**Figure 11.** Dependence of shear stress on shear rate at  $n$ , equal to: 1–5.0; 2–3.0; 3–2.0; 4–1.5; 5–1.0; 6–0.8; 7–0.4.

$\alpha_2$	$K_2$	$T^{\circ}\text{C}$	$(\text{grad}P)_0$
0.08	0.000033	24	0.0140
0.04	0.0000895	50	0.00625
0.02	0.000245	80	0.00470

The dependence of the initial pressure gradient on temperature is given as:  $(\text{grad}P)_0 = 2.197 \times 10^{-4} + \frac{0.3275}{T}$ .

**Table 2.**  
Coefficients of rheological models included in Eq. (41) depending on the temperature.

and models have been put forward to describe the shear flow of oil dispersed systems, resulting in a wide variety of rheological dependences of effective viscosity on shear stress  $\tau$  and shear rate  $\dot{\gamma}$ .

## 6. Rheological models of oil filtration

Using the experimental data of Ref. [19] and Eqs. (38) and (40), we represent the filtration rate in the following form:

$$V = K_2(T) \left( 1 - \exp \left( -\alpha_2(T) (z/z_0)^6 \right) \right) z \quad (41)$$

where  $\alpha_2 = 0.1422 \exp(-0.0247T)$ ,  $K_2(T) = 1.4 \times 10^{-5} \exp(0.0364T)$ ,  $z = \text{grad}P$ ,  $z_0 = (\text{grad}P)_0$ ,  $K_2 = k/\eta^*$  are oil mobility. The high value of the exponent is explained by a sharp drop in viscosity during the destruction of the formed structure. **Figure 12** below shows a comparison of the calculated Eq. (41) and experimental values of the filtration rate of heavy oils at different temperatures for various West Siberian fields [19].

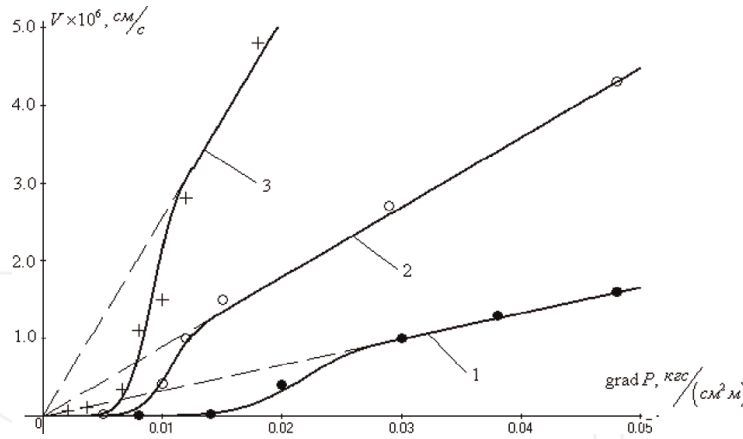
The **Table 2** shows the values of the coefficients included in Eq. (41) depending on the temperature.

## 7. Drag coefficient of particles in a non-Newtonian fluid

Heavy oils differ from ordinary liquids in that their viscosity changes with increasing shear rate, and the shear rate index  $n$  characterizes the degree of non-Newtonian behavior of the material. At  $n < 1$ , liquids exhibit viscous-plastic properties, and at  $n > 1$ , dilatant properties. These conditions impose special conditions on the description of the hydrodynamics of the flow of non-Newtonian fluids and are reflected in the description of the drag coefficients of particles in such fluids.

Theoretical and experimental studies of hydrodynamics and the drag coefficient of solid particles, drops, and bubbles in a Newtonian fluid are given in Ref. [3], in a power-law non-Newtonian fluid are given in refs. [20–26]. Using asymptotic methods, a formula was obtained in [27, 28] for calculating the drag coefficient of a bubble in a power-law non-Newtonian liquid ( $n < 1$ ) for small numbers  $\text{Re}_t < 1$ .

$$C_D = \frac{16}{2^n \text{Re}_t} X(n), \quad X(n) = 2^{n-1} 3^{\frac{n-3}{2}} \frac{13 + 4n - 8n^2}{(2n+1)(n+2)} \quad (42)$$



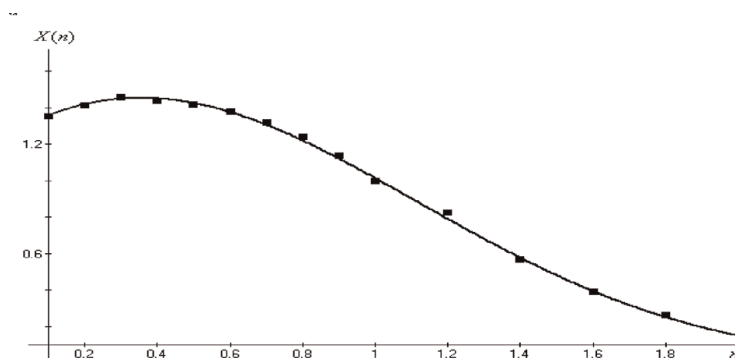
**Figure 12.**  
 Change in filtration rate of abnormal oils at different temperatures: 1 -  $T = 24$  C, 2 -  $T = 50$  C; and 3 -  $T = 80$  C.

where  $X(n)$ —is a parameter that characterizes the rheological properties of the flow and depends on the exponent  $n$ ,  $Re_t = \frac{d^n \rho_c V^{2-n}}{k_0}$ . Calculations using formula (42) show that for pseudo-plastic fluids the drag coefficient is higher, and for dilatant fluids, it is lower than the corresponding drag coefficients, when a Newtonian fluid flows around a bubble. In addition to expression (42), the authors of refs. [27, 28] present the experimental values and various dependencies  $X(n)$  on  $n$ .

$$\begin{aligned}
 X(n) &= 2^n 3^{\frac{n-3}{2}} [1 - 3.83(n - 1)], \quad 0.7 \leq n \leq 1; \\
 X(n) &= \frac{2}{3} (3\gamma^2)^{\frac{n-3}{2}} \frac{13 + 4n - 8n^2}{(2n + 1)(n + 2)}, \quad \gamma > 10; \\
 X(n) &= 3^{-\frac{n+3}{2}} \left[ \frac{2(2n + 1)(2 - n)}{n^2} \right]^2; \quad X(n) = 2^n 3^{\frac{n-3}{2}} \frac{1 + 7n - 5n^2}{n(n + 2)}, \quad n < 1.
 \end{aligned}
 \tag{43}$$

The second formula (43) characterizes the behavior of drops in non-Newtonian fluid. The above expressions, obtained theoretically, do not allow solving this problem for the general case.

A satisfactory dependence of  $X(n)$  on  $n$  for solid particles in a non-Newtonian liquid for a sufficiently large region of variation  $0.1 \leq n \leq 1.8$  using experimental data [27, 28] can be represented as:



**Figure 13.**  
 Dependence  $X(n)$  on  $n$ .

$$X(n) = \frac{7}{450} (5n + 9)^2 \exp \left[ -n^{2.5} \left( 1 + \frac{n^2}{10} \right) \right] \quad (44)$$

It follows from formula (44) that at  $n = 1$ , the value of  $X(n) \approx 1$ . **Figure 13** shows a comparison of the calculated values with the experimental values.

In refs. [27, 28], various formulas are also given that characterize the deformation of bubbles in a non-Newtonian fluid.

$$\begin{aligned} Y_K &= \frac{b_0}{a_0} = 0.0628\delta^{0.46}, & 20 \leq \delta \leq 100, \\ Y_K &= 1.4, & \delta \leq 4, \\ Y_K &= 6.17\delta^{-1.07}, & 4 \leq \delta \leq 20, \\ \delta &= \text{Re}_t \text{Mo}_t^{0.078}, & \text{Mo}_n = \text{We}^{n+2} \text{Fr}^{2-3n} \text{Re}_t^{-4}, & 0.64 \leq n \leq 0.9 \end{aligned} \quad (45)$$

where  $a_0, b_0$  are the major and minor axes of the ellipsoid.

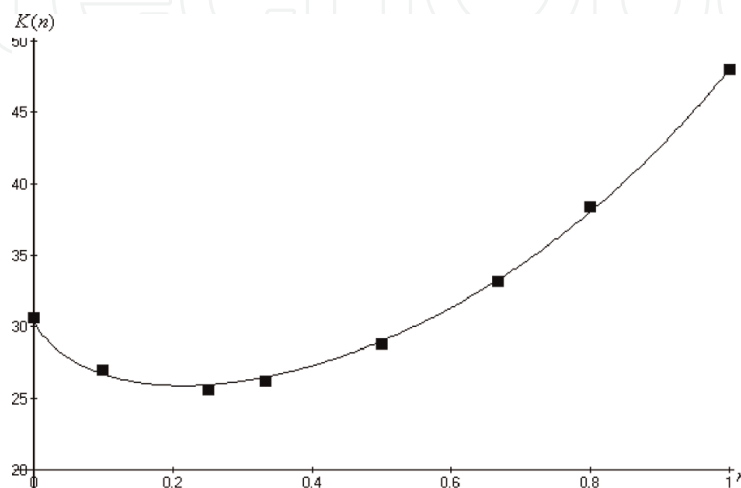
For small values of the number  $5 \leq \text{Re}_d \leq 25$ , the drag coefficient of a bubble in a non-Newtonian liquid, according to the works in refs. [2, 27, 29], can be determined in the form:

$$C_{DG} = \frac{K(n)}{\text{Re}_t} \quad (46)$$

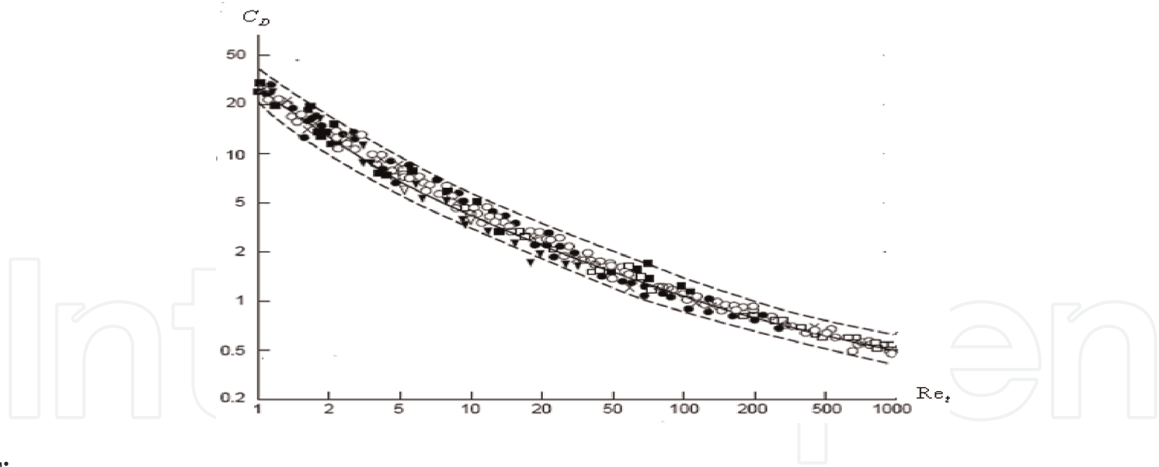
Here  $K(n)$  – is a coefficient depending on  $n$ . Using the experimental data given in refs. [27, 28], the following formula is proposed in this work:

$$K(n) = 30.6 \frac{\exp(2.53n)}{\left(n^{\frac{6}{7}} + 1\right)^3} \quad (47)$$

As follows from **Figure 14**, the coefficient  $K(n)$  varies significantly depending on  $0 \leq n \leq 1$  and, passing through a minimum at  $n = 1$ , tends to  $K(n) \approx 48$ , which characterizes the drag coefficient of a bubble in a Newtonian fluid, proposed in Ref. [29] for small numbers  $\text{Re}_d$ .



**Figure 14.**  
Dependence of  $K(n)$  on  $n$ .



**Figure 15.** Drag coefficient of solid particles in non-Newtonian liquid (solid line - drag coefficient in Newtonian fluid and dotted lines correspond to 30% of the experimental data scatter threshold) [30].

A lot of experimental data for the drag coefficient of particles in a non-Newtonian fluid collected from the literature for  $Re_d < 1000$ , is given in refs. [30, 31] and in **Figure 15**.

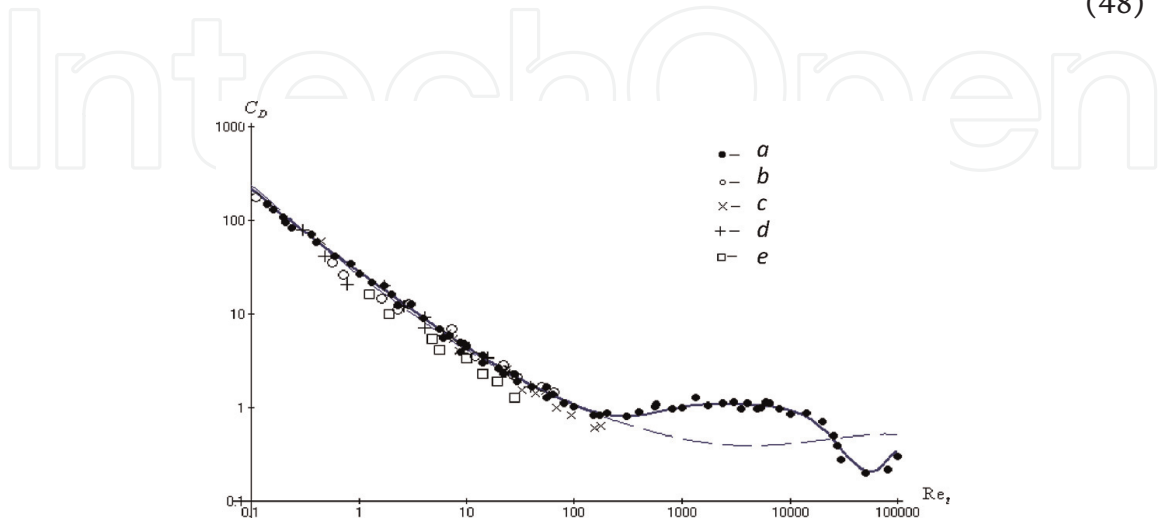
In this paper, using experimental data from Ref. [22], we propose an equation for determining the drag coefficient of solid particles in a power-law non-Newtonian fluid for a wide range of number variation  $0.1 < Re_t < 10^5$ .

$$C_D = \frac{24X(n, Re_t)}{Re_t} \left[ 1 + Re_t^{\frac{2}{3}} \xi(Re_t) \right] + 0.3(1 - \exp(-0.2 \times 10^{-19} Re_t^4))$$

$$X(n, Re_t) = \frac{5 \cdot 6^{n-1}}{1 + 4n} \left[ 1 + (1 - n)^{\frac{1}{3}} \frac{Re_t^{\frac{3}{2}}}{1 + (1 - n)^{\frac{1}{3}} Re_t^{\frac{3}{2}}} \right], \quad 0.8 < n \leq 1, \quad 0.1 \leq Re_t \leq 10^5$$

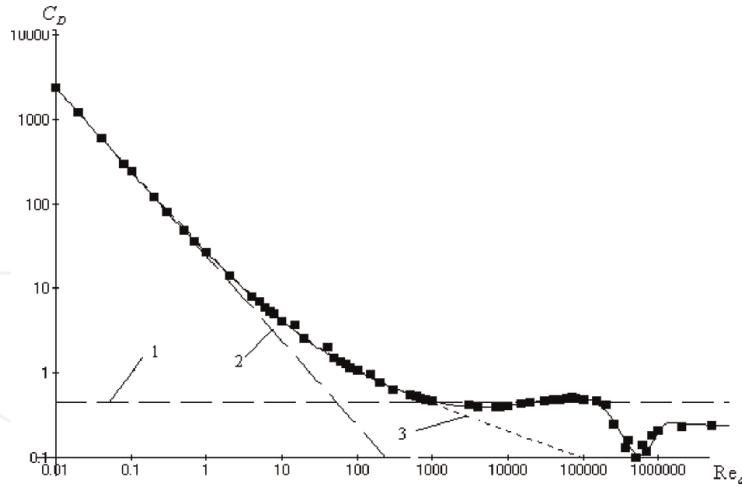
$$\xi(Re_t) = \frac{Re_t^2}{5 + 8 Re_t^2} + \frac{Re_t}{6.8 \times 10^6 Re_t^{-\frac{5}{4}} + 32 Re_t^{\frac{2}{3}} + 952.8 \times 10^{-13} Re_t^{\frac{10}{3}}}$$

(48)



**Figure 16.** The dependence of the drag coefficient of a solid particle in a non-Newtonian fluid on the number  $Re_d$  according to the data of various authors (dashed line is the drag coefficient of a particle in a Newtonian fluid):  $a - n = 0.84 - 0.86$  [32];  $b - n = 0.75 - 0.90$  [32];  $c - n = 0.75 - 0.92$  [33];  $d - n = 0.56 - 0.75$  [34];  $e - n = 0.73$  [35].





**Figure 17.**  
The drag coefficient for a solid spherical particle.

For values  $Re_t < 1000$ , this equation simplifies to the form:

$$C_D \approx \frac{24X(n, Re_t)}{Re_t} \left( 1 + 0.125 Re_t^{\frac{2}{3}} + 2.51 \times 10^{-5} Re_t^2 \right) \quad (49)$$

**Figure 16** compares the calculated values of the drag coefficient according to Eq. (49) with the experimental data given in refs. [31–35]. As follows from this figure and calculations using the formula in Eq. (49), decreasing the drag coefficient of solid particles in viscous-plastic liquids is less than in Newtonian ones.

The drag coefficient of solid spherical particles in a Newtonian fluid is shown in **Figure 17** [1, 3]. Comparing **Figures 16 and 17** in the region of the drag crisis, it should be noted that when a non-Newtonian flow flows around a solid particle, the drag crisis occurs much earlier than in a Newtonian fluid.

$$1 - C_D \approx 0.44; 2 - C_D = \frac{24}{Re_d}; 3 - C_D = \frac{24}{Re_d} (1 + 0.15 Re_d^{0.687}). \quad (50)$$

## 8. Free settling of particles in heavy oil

The motion of a single particle in a force field with a slow flow of the medium, taking into account the added mass, the weight force, corrected for the Archimedes force and the resistance force, is described by the Eq. (3)

$$\frac{d\mathbf{V}_P}{dt} = \frac{\Delta\rho}{2\rho_d + \rho_c} \mathbf{g} - \frac{3}{4} C_D \frac{\rho_c}{2\rho_d + \rho_c} |\mathbf{V}_P - \mathbf{U}| (\mathbf{V}_P - \mathbf{U}) \quad (51)$$

In the steady-state, this equation for a Newtonian fluid, as a result of the balance of forces acting on a particle and for small numbers  $Re$ , is transformed into a simpler form:

$$\frac{4}{3} \pi a^3 \Delta\rho g = \rho_c V_s^2 \pi a^2 C_D \quad (52)$$

From this equation provided that  $C_D = 24/Re$  we determine the deposition rate in small numbers  $Re_d \ll 1$  in the form of the Stokes model  $V_s = \frac{1}{18} \frac{\Delta\rho a^2 g}{\eta_c}$ .

**Figure 18** shows characteristic curves and experimental data on the settling rate. Given expressions for a non-Newtonian fluid are as follows:

$$C_D = \frac{24}{2^n \text{Re}} X(n), \quad X(n) = 2^{n-1} 3^{\frac{n-3}{2}} \frac{13 + 4n - 8n^2}{(2n + 1)(n + 2)}, \quad \text{and} \quad \text{Re}_t = \frac{a^n \rho_c V^{2-n}}{k_0}. \quad (53)$$

Let us rewrite Eq. (52) for a power-law non-Newtonian fluid, it will be represented as:

$$\frac{4}{3} \frac{a \Delta \rho g}{\rho_C} \frac{\text{Re}}{24 X_n} = V_s^2 \quad (54)$$

$$X_n = 2^{-1} 3^{\frac{n-3}{2}} \frac{13 + 4n - 8n^2}{(2n + 1)(n + 2)}$$

Transforming Eq. (54), we finally obtain an expression for the settling rate of a single solid particle in a non-Newtonian fluid flow.

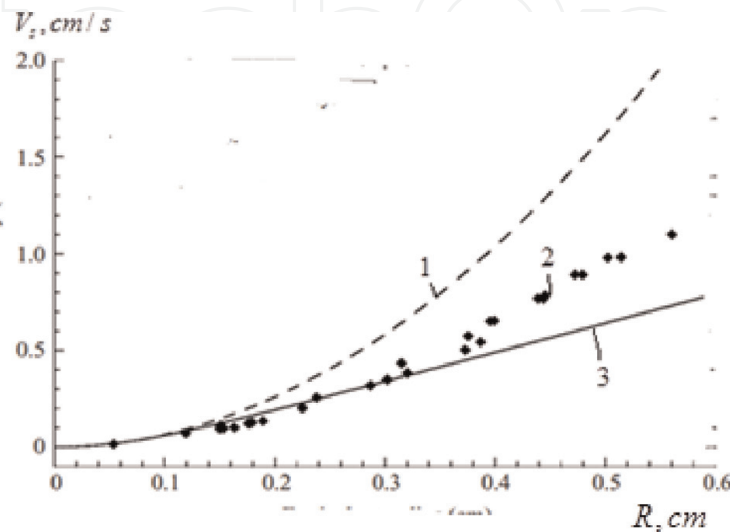
$$V_s = \left( \frac{1}{18} \frac{\Delta \rho g}{k_0 X_n} \right)^{1/n} a^{(n+1)/n} \quad (55)$$

It should be noted that if  $n = 1$ , then  $X_n = 1$  and expression (55) also turns into the Stokes equation for the deposition of a solid particle in a Newtonian fluid at small numbers  $\text{Re}$ .

$$V_s = \frac{1}{18} \frac{\Delta \rho g a^2}{\eta_C} \quad (56)$$

Obviously, an important role in the migration and sedimentation of particles belongs to the resistance forces, depending on the number  $\text{Re}_d = Ua/\nu_c$ , shape and size of particles, on the physicochemical properties of particles, and the medium.

Numerical calculations using the above formulas show that for fine-dispersed particles of asphaltenes and paraffins, with increasing particle size, the degree of



**Figure 18.** Settling velocity profiles depending on particle radius: 1 - Hadamard-Rybczynski equation, 2 - experiment, and 3 - Eq. (52).

entrainment of particles by a pulsating medium decreases, and fine-dispersed particles react to turbulent pulsations of the medium, perform under their influence a pulsating motion relative to the moles of the carrier phase and random motion in all directions due to turbulent diffusion. These parameters are very important factors that determine the degree of migration and settling of particles in a turbulent flow. It should be noted that for Stokes spherical solid particles at  $Re_d < 1$ , the rate of their settling from the volume is determined as:

$$V_s = \tau_p g \tag{57}$$

For a viscous-plastic fluid obeying the Ostwald–de Ville model  $\tau = \eta_c \dot{\gamma}^n$ , Eq. (56) can be written as:

$$V_p = \frac{1}{18} \frac{\Delta \rho g a^2}{\tau / \dot{\gamma}^n} \tag{58}$$

with a relaxation time equal to

$$\tau_p = \frac{\rho_a a^2 \dot{\gamma}^n}{18 \tau} \tag{59}$$

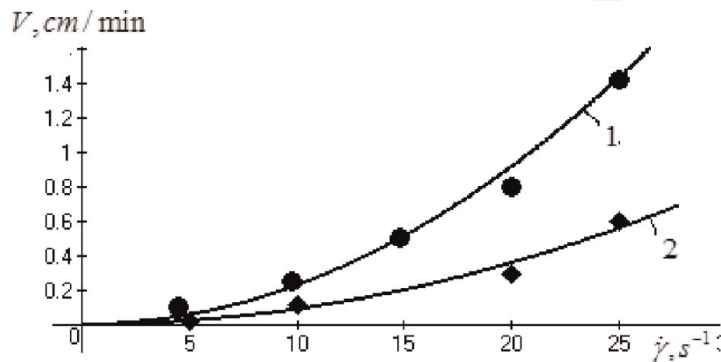
For a viscous flow of a power-law fluid, we obtain the expression for the relaxation time in the form:

$$\tau_p = \left( \frac{1}{18 k_0 X_n} \frac{\Delta \rho g}{\tau} \right)^{1/n} a^{(n+1)/n} \tag{60}$$

Using the experimental data for the deposition of glass particles in a non-Newtonian liquid [36], it can be noted that the deposition rate dependence is linear only at low shear rates.

A somewhat different formula for the deposition of single particles in a non-Newtonian liquid was proposed in Ref. [37], which can be obtained from (58) at  $X_n = 2 - n$ ,  $0.4 < n < 1.2$

$$V_p = \left( \frac{\Delta \rho g}{18 k_0 (2 - n)} \right)^{1/n} a^{\frac{n+1}{n}} \tag{61}$$



**Figure 19.** Dependence of the particle settling rate in a non-Newtonian liquid on stress and shear rate for particles of size  $a = 275 \mu m$ , 1-  $\tau = 8.5 Pa$ ; 2 -  $\tau = 15 Pa$ .

Using the experimental data on the settling of solid particles in a non-Newtonian fluid [38] in **Figure 19** shows a comparative characteristic according to the model (58).

For large numbers of Re, expression (52) can be written as:

$$\text{Re}^2 C_D^{2-n}(n, \text{Re}) = \left(\frac{8}{3} \Delta \rho g\right)^{2-n} \rho_C^n \frac{a^{n+2}}{k^2} \quad (62)$$

Expression (62) does not allow an analytical determination of the settling rate and is solved only numerically.

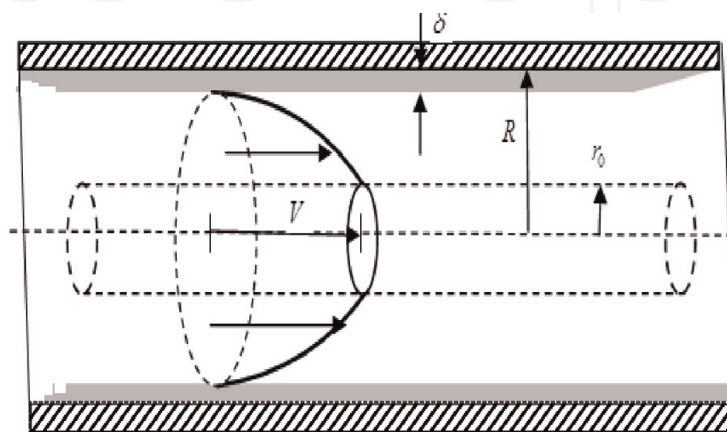
## 9. Flow of heavy oil in pipes with settling

During the flow of heavy oil, asphalt-resinous substances and particles of the solid phase are deposited on the inner surface of the pipes. As a result of the deposition of particles on the surface of the pipe, a layer of deposits is formed, which continuously grows and is compacted by the normal component of pressure. The quantitative content of asphaltenes and resins, due to their high energy of adhesion and cohesion, significantly affect the rheological properties of heavy oils and the formation of a dense layer on the surface. The high ability to form coagulation structures and aggregates contributes to the deterioration of the hydrodynamic characteristics of the flow of non-Newtonian oils up to zero flow velocity due to an increase in the effective viscosity of the medium. The mechanism of sedimentation of particles from heavy oil can be carried out by diffusion (for a vertical surface), gravitational (for a horizontal surface), and diffusion-gravitational way. The formation of a dense layer on the inner surface of the pipe has a significant effect on the transfer of mass and heat, and on the hydrodynamics and rheological structure of the flow.

The hydrodynamic equation for the flow of viscous-plastic oil in a pipe, described by the Bingham rheological equation  $\tau = \tau_0 + \eta_C \dot{\gamma}$  for the stationary case in cylindrical coordinates, will be represented as:

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial V}{\partial r} \right) = \frac{1}{\eta_C} \frac{\partial P}{\partial x} \quad (63)$$

The boundary conditions for the flow of viscous-plastic oil in the pipe are (**Figure 20**)



**Figure 20.**  
 Velocity distribution of a viscous-plastic fluid over a pipe section.

$$r = R, \tau < \tau_0, \tau = \tau_0 + \eta_C \frac{\partial V}{\partial r} = 0, V = 0 \quad (64)$$

$$r = R, \tau > \tau_0, \tau = \tau_0 + \eta_C \frac{\partial V}{\partial r} \neq 0, V = 0 \quad (65)$$

Integrating condition (64), we have:

$$V = -\frac{\tau_0 R}{\eta_C} \quad (66)$$

Integrating condition (65), we obtain:

$$V = \frac{\tau_0 R}{\eta_C} \left(1 - \frac{\tau}{\tau_0}\right) \quad (67)$$

Integrating expression (63) twice, we obtain:

$$V = -\frac{1}{4\eta} \frac{\partial P}{\partial x} r^2 + A \quad (68)$$

Here,  $A$  is the integration coefficient equal to for the first boundary condition (64)

$$A = \frac{1}{4\eta_C} \frac{\partial P}{\partial x} R^2 - \frac{\tau_0 R}{\eta_C} \quad (69)$$

and for the second boundary condition (3.43)

$$A = \frac{1}{4\eta} \frac{\partial P}{\partial x} R^2 - \frac{\tau_0 R}{\eta} \left(1 - \frac{\tau}{\tau_0}\right) \quad (70)$$

As a result of solving the hydrodynamic Eq. (63) for a viscous-plastic fluid described by the expression in a pipe with boundary conditions (64) and (68) and assuming that, taking into account (68)–(70), in the simplest case, we obtain the following distribution for the flow velocity (**Figure 20**)

$$\begin{aligned} V(r) &= \frac{\Delta P R^2}{4\eta_C l} \left(1 - \frac{2l}{R} \frac{\tau_0}{\Delta P}\right), \quad r < r_0 \\ V(r) &= \frac{\Delta P R^2}{4\eta_C l} \left[1 - \frac{r^2}{R^2} - \frac{4l}{R} \frac{\tau_0}{\Delta P} \left(1 - \frac{r}{r_0}\right)\right], \quad r_0 \leq r < R \end{aligned} \quad (71)$$

where  $l$  is the pipe length and  $r_0$  is the plug radius. For the case of the formation of a dense layer of particles on the inner surface of the pipe with a thickness of  $\delta$ , which is typical for the flow of viscous oil and, assuming that  $R = R_0\beta$ ,  $\beta = 1 - \delta/R_0$  ( $\delta$  is the thickness of the sediment layer), then these expressions (71) for the quasi-stationary case will be presented as:

$$\begin{aligned} V(r) &= \frac{\Delta P R_0^2 \beta^2}{4\eta_C l} \left(1 - \frac{2l}{R_0\beta} \frac{\tau_0}{\Delta P}\right), \quad r < r_0 \\ V(r) &= \frac{\Delta P R_0^2 \beta^2}{4\eta_C l} \left[1 - \frac{r^2}{R_0^2 \beta^2} - \frac{4l}{R_0\beta} \frac{\tau_0}{\Delta P} \left(1 - \frac{r}{r_0}\right)\right], \quad r_0 \leq r < R_0\beta \end{aligned} \quad (72)$$

It should be noted that if  $\beta = 1$ , then the thickness of the deposits is absent, and if  $\beta = 0$ , then  $\delta = R_0$ ; there is a complete blockage of the pipe and the flow velocity  $V(r) \rightarrow 0$ , that is, carrying capacity is reduced to almost zero.

This distribution of velocities in hydrodynamics is called the “structural regime of motion.” The volumetric flow rate of viscous-plastic oil over the pipe section is determined as:

$$Q = \frac{\pi R^4 \Delta P}{8l\eta_c} \left( 1 - \frac{4}{3} \frac{r_0}{R} + \frac{1}{3} \frac{r_0^4}{R^4} \right) \quad (73)$$

As a result of the formation of a dense layer on the inner surface of the pipe, assuming that  $R = R_0\beta$  you can write:

$$Q = \frac{\pi R_0^3 \beta^3 \Delta P}{8l\eta_c} \left( 1 - \frac{4}{3} \frac{r_0}{R_0\beta} + \frac{1}{3} \frac{r_0^4}{R_0^4 \beta^4} \right) \quad (74)$$

As follows from this equation, with an increase in the thickness of deposits on the inner surface of the pipe, which corresponds to a decrease in  $\beta$ , the volume flow of oil decreases. Thus, with a complete blockage of the pipe section, the fluid flow also tends to zero. For a Bingham fluid, the conditions for the transition from a structural flow to a turbulent regime are defined as:

$$\text{Re}_{cr} = \frac{1 - 4\alpha + \alpha^4}{24\alpha} \text{He}, \quad \text{He} = 16800 \frac{\alpha}{(1 - \alpha)^2} \quad (75)$$

or you can write

$$\text{Re}_{cr} = 700 \frac{2 + (1 + \alpha)^2}{1 - \alpha} \quad (76)$$

Here,  $\text{Re}_{cr} = V_{cr}d/\nu_c$  is the critical Reynolds number,  $\text{He} = \tau_0 d^2/\nu_c$  is the Hedstrom number, and  $\alpha = \tau_0/\tau_R$ ,  $\tau_R$  is the shear stress on the pipe wall with a diameter equal to  $d$ . In the presence of a dense layer on the inner surface of the pipe, one can write  $\text{He} = \text{He}_0\beta^2$ , that is, as the thickness of the deposits grows, the intensity of turbulence decreases. For some oils, power-law of flow is observed  $\tau = k\dot{\gamma}^n$ , for which one can write:

$$\text{Re} = \frac{8}{k_0} \left( \frac{n}{6n + 2} \right)^n \rho_c d^n V^{2-n} = \text{Re}_0 \beta^{3n-4} \quad (77)$$

Thus, the transition from a structural flow regime to a turbulent one occurs at certain values of stress  $\tau$  and dimensionless deposit thickness  $\beta$ , and, if  $n > 1$  the turbulence intensity decreases as the thickness increases and, conversely, at  $n \leq 1$ – the turbulence intensity increases.

## 10. Practical ways to improve rheological properties at heavy oil refining

Heavy oils, due to the high content of heavy components (65–70% fraction  $>350^\circ\text{C}$  boiling) exhibit viscous-plastic properties of non-Newtonian oils. Currently, there are



no special technologies for the processing of heavy oils, with the exception of some works [39, 40], since their processing is limited by the problems of oil transport 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, paraffins, water-in-oil, and other factors [41–44]. When processing heavy oils at operating plants at relatively low temperatures, the effective viscosity of the oil increases, which helps to reduce its fluidity and, in general, the productivity of the process. The decrease in the viscosity of heavy oils, which ensures their transportability, depends on two factors: an increase in its temperature and dilution with light components.

In this regard, the main factor for improving the rheological properties of oil and the conditions for its processing when solving practical problems is to increase the temperature of the oil stock at the inlet to the plant and its partial dilution by creating a recirculation of a lighter component for mixing with crude oil (Figure 21). This intensification of the processing process allows us to solve both problems simultaneously. The process of primary oil refining is carried out by its preliminary heating to a certain temperature with its intermediate purification from water and salts in the dehydrator 4, contained in the oil, and further separation in the mass-exchange distillation column 5 (Figure 20).

The unit used Kazakhstan heavy paraffinic oil with high viscosity (Figure 22).

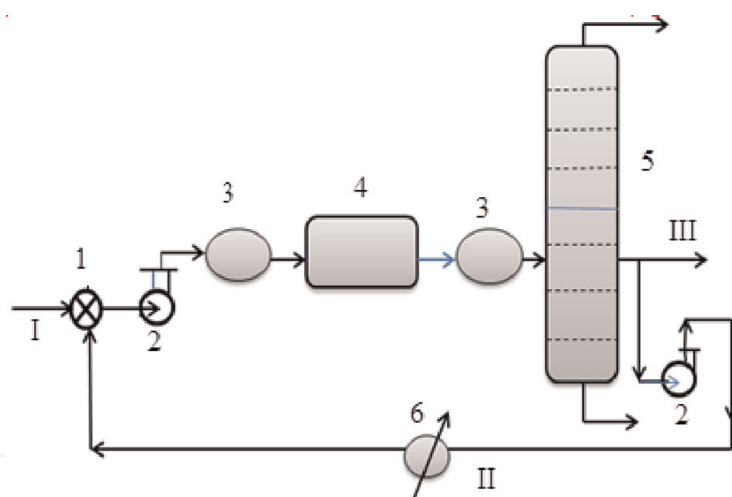


Figure 21. Scheme of primary oil refining with recirculation: 1 - mixer, 2 - pump, 3 - system of heat exchangers, 4 - dehydrator, 5 - distillation column, and 6 -refrigerator; I - crude oil, II -recirculation line, and III - fraction >240.

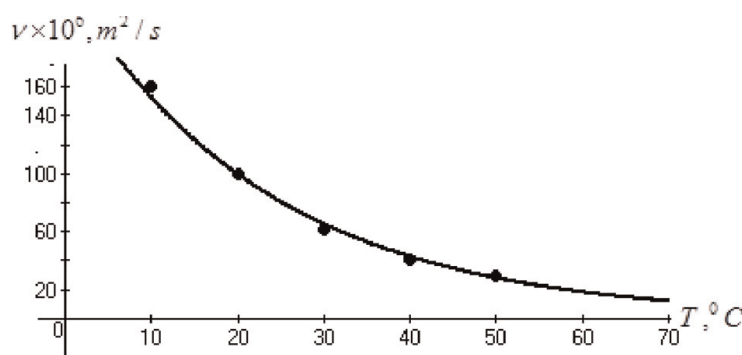


Figure 22. Temperature dependence of crude oil viscosity.

The dependence of the kinematic viscosity of crude oil on temperature is represented by the expression

$$\nu = 234.05 \exp[-0.048T] \times 10^{-6}, m^2/s \quad (78)$$

To improve rheological properties and viscosity in order to ensure its transportability, it is proposed to return part of the  $>240^{\circ}C$  fraction from column 5 back for mixing with crude oil after preliminary cooling to pump 2. The creation of a recirculation system makes it possible to reduce the kinematic viscosity of crude oil by increasing its temperature to  $20-25^{\circ}C$  and diluting it with a lighter liquid. Experimental studies have shown a significant effect of the amount and temperature of the  $>240^{\circ}C$  fraction on the effective viscosity of crude oil (**Figure 23**).

As follows from **Figure 23** with an increase in temperature and the amount of added fraction, the viscosity of the oil decreases. The expression describing the change in the kinematic viscosity of oil from the temperature at various fraction contents  $>240^{\circ}C$  can be represented as:

$$\nu = 108.95(1 - 2.17\varphi) \exp(-0.028T) \times 10^{-6} \quad (79)$$

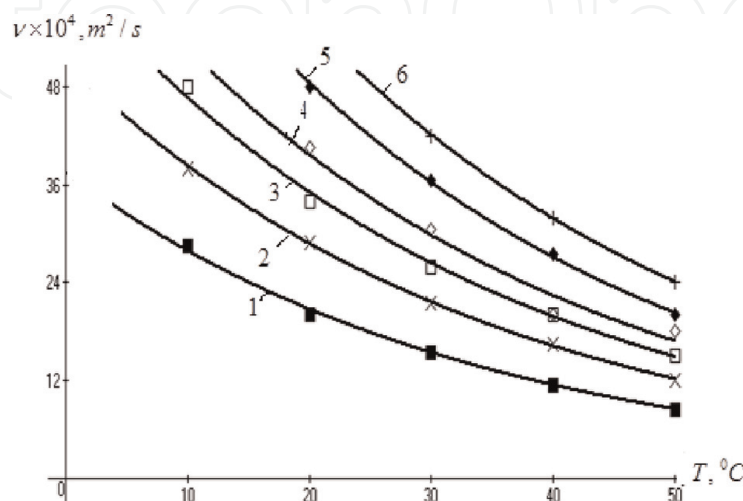
Here,  $\varphi = m_R/(m_0 + m_R)$ —is the mass fraction of the  $>240^{\circ}C$  fraction in crude oil, and  $m_0, m_R$  are the mass flow rates of crude oil and fractions. The viscosity of heavy oil due to dilution with a light fraction is defined as:

$$\nu = \frac{\nu_0(T)}{1 + m\varphi^2} \quad (80)$$

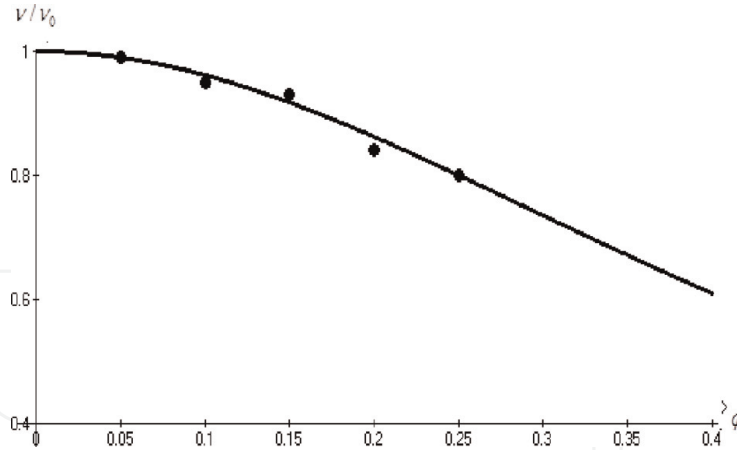
Data of **Figures 23** and **24** show the dependence of the viscosity of oil when it is diluted with a light fraction, calculated by Eq. (80).

As follows from **Figure 23**, the relative fluidity of heavy oil increases, as a result of its dilution with a light fraction, which improves the rheological properties of the oil.

The temperature of a mixture of crude oil diluted with a light fraction  $>240^{\circ}C$  is defined as:



**Figure 23.** The dependence of the kinematic viscosity of oil on temperature when it contains fractions  $>240^{\circ}C$   $\varphi$  (mass fractions), is equal to: 1–0.3; 2–0.25; 3–0.20; 4–0.15; 5–0.10; 6–0.05,  $\nu \times 10^4, m^2/s$ .



**Figure 24.**

The dependence of the viscosity of heavy oil on the proportion of the light fraction at a temperature  $T = 30^\circ\text{C}$ ,  $m = 3.5$ .

$$T = (1 - \phi)c_{p0}T_0 + \phi c_{pR}T_R \quad (81)$$

Here  $T$  is the temperature of the mixture at the inlet to column 5;  $c_p$  is the heat capacity of crude oil and fraction; and  $T_0, T_R$  are temperatures of crude oil and fraction.

Recirculation of some lighter, higher temperature, lower viscosity refining fraction for blending with crude oil reduces the viscosity of the oil by raising its temperature and partially diluting it with the lighter fraction (Figure 24).

## 11. Analysis and discussion of results

The main problem of the rheology of heavy oils is the study of the regularity of their behavior under the action of external deforming stresses, taking into account their structure. At the same time, the processes associated with irreversible residual deformations and the flow of various viscous and plastic materials, as well as the phenomena of relaxation of heavy oils, allow us to study the fundamental properties of oil disperse systems characterized by shear stress, effective viscosity, corresponding to a certain disordered structure of the system. The effective viscosity of non-Newtonian oils depends on temperature and pressure, as well as on the shear strain rate, the properties of the disordered structure, the content of asphalt-resin substances and the concentration of the dispersed phase, and determines the degree of oil transportability during its extraction and processing.

Concluding this study, we note the main stages and problems of the rheology of heavy oils considered in this paper:

- A high content of asphalt-resinous, paraffinic substances and solid phases of various types and nature in heavy oils creates a special physical structure that gives them viscous-plastic properties, the description of which is based on the rheological models of Bingham, Hershel-Bulkley, and Ostwald-de Ville model. Such high-viscosity oil systems are formed as a result of the combination of oil emulsions with the presence of water droplets, oil suspensions with the presence

of a solid phase in the composition of crude oil and gas suspensions, including gas bubbles, as well as asphalt-resinous and waxy substances dissolved in oil. The complex physical structure of heavy oil predetermines the possibility of the formation of more complex coagulation structures that adversely affect the rheology.

- For the filtration of heavy oils in an anisotropic porous medium, a new nonlinear filtration equation is proposed that takes into account shear stress, elastic limit, and effective viscosity. Analysis and solutions of this equation show a high degree of dependence on the filtration rate and effective viscosity on the pressure gradient. It is noted that the nonlinearity of the rheological model of filtration in porous media is determined by the nature of the hydrodynamic flow and the high content of asphalt-resinous and paraffinic substances in the composition of heavy oil. Solutions of the rheological filtration model make it possible to determine the filtration rate depending on the shear stress or the pressure drop and on the effective viscosity.
- Expressions were proposed for determining the flow rate of heavy oils and the rate of free settling of particles, particulate matter and particle drag coefficients in heavy oils.
- Proposed effective ways to improve the rheological properties of oil during its processing at existing plants, ways to create a recirculation scheme.

The problems associated with the rheology of heavy oils considered in this paper show the complexity, both in theoretical and practical aspects, of solving these problems, which is determined by the course of many interconnected physical phenomena and the high randomness of these processes. The relationship between determinism and randomness in these phenomena gives rise to some problems that lead to the use of empirical models that do not take into account the true physical mechanisms and phenomena. In particular, there is no way to take into account the nature and properties of the formed disordered structures, and the conditions for their formation and destruction in rheological models. All these phenomena are taken into account in rheological models indirectly in the form of introducing the volume fraction of particles into the model in the form of various functions. As a result of this, the same experimental data can be simultaneously described by many empirical models within a given accuracy. Obviously, taking into account these factors will lead to a more complex structure of the rheological model, and therefore, for practical calculations, it is possible to use various empirical models for specific rheological fluids.

## Nomenclature

$a$	is particle diameter
$C_D$	is drag coefficient of particle
$D_M$	is coefficient of molecular diffusion
$D_{eff}$	is coefficient of effective diffusion
$d$	is diameter of the pipe
$k_0$	is consistency factor'

$k_p$	is permeability coefficient
$m$	is mass of substance
$N$	is total number of particles per unit volume
$n$	is an indicator of the degree of non-Newtonian fluids
$P$	is total pressure
$R$	is radius of the particle
$t$	is time
$T$	is temperature
$U$	is the average flow velocity
$V_p$	is total particle deposition rate

### Greek letters

$\beta$	is dimensionless thickness of particle deposits
$\varepsilon_R$	is specific energy dissipation per unit mass
$\varepsilon$	is porosity
$\eta_{eff}$	is effective viscosity of suspensions
$\eta_c, \eta_d$	are dynamic viscosity of the medium and particles
$\nu_c, \nu_d$	are kinematic viscosity of the medium and particles
$\rho_c, \rho_d$	are density of the medium and particles
$\rho_i$	is concentration of particles
$\sigma_D$	is coefficient of surface tension
$\tau$	is shear stress
$\tau_0$	is yield strength
$\varphi$	is the volume fraction of particles
$\omega$	is frequency of turbulent coalescence
He	is the Hedstrom number
Re	is the Reynolds number
Wi	is the Weissenberg number

### Indices

$d$	is a solid particle
$c$	is medium
0	is the initial value

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