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*G.I. Kelbaliyev^a, S.R. Rasulov^b, M.R. Manafov^a***A GENERAL RHEOLOGICAL EQUATION FOR HIGHLY VISCOUS NON-NEWTONIAN FLUIDS AND ITS APPLICATIONS**^a Institute of Catalysis and Inorganic Chemistry of the National Academy of Sciences of Azerbaijan, Baku, Azerbaijan^b Azerbaijan State University of Oil and Industry, Baku, Azerbaijan

A new generalized rheological equation for highly viscous fluids is proposed, which captures the relationship between shear stress, relaxation time, and shear rate. Various analytical solutions with boundary conditions are presented to evaluate shear stress and effective viscosity in highly viscous dispersed systems. A general rheological equation for power-law fluids is also provided. Practical applications include estimating the effective viscosity of heavy oils and dispersed systems, considering the formation and the breakdown of coagulation structures. The new rheological model has been validated against experimental data, showing satisfactory agreement.

Keywords: rheology, petroleum dispersed systems, highly viscous non-Newtonian fluids, generalized rheological equation, coagulation structures, effective viscosity, shear stress, shear rate, relaxation time.

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Introduction

The rheology of highly viscous non-Newtonian fluids has traditionally relied on numerous empirical and semi-empirical models relating shear stress to shear rate, often derived from Newton's classical equation. These models treat the fluid as a homogeneous medium whose mechanical behavior matches observed experimental results, a common approach in continuum mechanics. This allows for simplified analysis while avoiding the complexity of interphase interactions [1–8].

Rheological models of non-Newtonian fluids relate shear stress to shear rate through the medium's viscosity or consistency. It is worth noting several semi-empirical formulas: the Bingham equation ($\tau = \tau_0 + \eta\dot{\gamma}$) (for viscoplastic fluids), Casson ($\tau^{1/2} = \tau_0^{1/2} + k^{1/2}\dot{\gamma}^{1/2}$), Herschel–Bulkley ($\tau = \tau_0 + k\dot{\gamma}^n$), Ostwald–de Waele ($\tau = k_0\dot{\gamma}^n$), Prandtl (for pseudoplastic fluids), Maxwell (for

viscoelastic fluids), and many other empirical equations that do not reflect the actual mechanisms of shear flow in such fluids. Models describing the behavior of structured dispersed systems often do not fully comply with physical laws, except in the case of individual particles, and represent empirical or semi-empirical approximations to the true behavior [9–15].

The aim of this study is to derive the general equation of shear flow of highly viscous fluids and to apply it in practice to the construction of rheological models for various systems.

Results and discussion*Equation of shear flow of highly viscous fluid*

Shear vortex-free flow of highly viscous fluids is accompanied by shear deformation and displacement of individual fluid layers with time relative to each other along the axis direction (Fig. 1). From the selected triangle (Fig. 1), the length of the deformed edge is defined as

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A general rheological equation for highly viscous non-Newtonian fluids and its applications

$$l = l_0 \sqrt{1 + (\dot{\gamma}t)^2} \approx l_0 \dot{\gamma}t. \quad (1)$$

In this equation, the shear rate, $\dot{\gamma}$, can be represented as the change in relative strain over time in the form of a flow velocity gradient:

$$\dot{\gamma} = \frac{d(dx/dy)}{dt} = \frac{d(dx/dt)}{dy} = \frac{dV_x}{dy}. \quad (2)$$

The value of relative deformation of the liquid layer is defined as

$$dx/dy = \lambda dV_x/dy = \lambda \dot{\gamma}. \quad (3)$$

Here λ is the relaxation time, $\dot{\gamma}$ is the shear rate, and t is the flow time.

The relaxation time and flow time are related through the Deborah number equal to $De = \lambda/t$. Let us make the following assumptions: a) for highly viscous fluids with Reynolds number $Re = Vx/\nu \ll 1$, the convective terms are much less viscous; b) the pressure distribution along the transverse axis is negligible $grad_x P \rightarrow 0$; c) in the equation of vortex-free flow of a viscous fluid along the direction of the axis x , let us assume that $\partial^2 V_x / \partial x^2 \gg \partial^2 V_x / \partial y^2$. Multiplying both parts of this expression by η , and differentiating by y , $\tau_{xy} = -\eta \partial V_x / \partial y$, we finally obtain $\partial^2 \tau_x / \partial x^2 \gg \partial^2 \tau_x / \partial y^2$.

Then, having omitted the lower indices, the equation of viscous fluid flow directed along the axis x , let us represent it in the following form:

$$\rho \frac{\partial V}{\partial t} = -\frac{\partial P}{\partial x} + \eta \left(\frac{\partial^2 V}{\partial x^2} + \frac{\partial^2 V}{\partial y^2} \right) \quad (4)$$

Multiplying both parts of equation (4) by η and differentiating the obtained expression by y , changing places of derivatives, we finally obtain:

$$\rho \frac{\partial}{\partial t} \left[\eta \frac{\partial V}{\partial y} \right] = -\frac{\partial}{\partial x} \left(\eta \frac{\partial P}{\partial y} \right) + \eta \frac{\partial^2}{\partial x^2} \left(\eta \frac{\partial V}{\partial y} \right) + \eta \frac{\partial^2}{\partial y^2} \left(\eta \frac{\partial V}{\partial y} \right) \quad (5)$$

Taking into account the insignificance of the transverse pressure gradient $\partial P / \partial y \ll 1$ and condition c), from expression (5) for vortex-free flow characteristic for non-Newtonian fluids, we obtain:

$$\frac{\partial \tau}{\partial t} = \nu \frac{\partial^2 \tau}{\partial x^2}. \quad (6)$$

The nonlinear expression (6) is a general second-order rheological equation for shear flow of highly viscous fluids. In particular, to solve equation (6), consider the following boundary conditions:

$$x = 0, t > 0, \tau = \tau_0; \quad x \rightarrow \infty, \tau \rightarrow \tau_\infty. \quad (7)$$

By introducing a dimensionless variable $\xi = x/\sqrt{4\nu t}$, equation (7) is represented in the form:

$$\frac{d^2 \tau}{d\xi^2} + 2\xi \frac{d\tau}{d\xi} = 0.$$

Considering the given boundary conditions (7), the analytical solution of this equation will be represented as

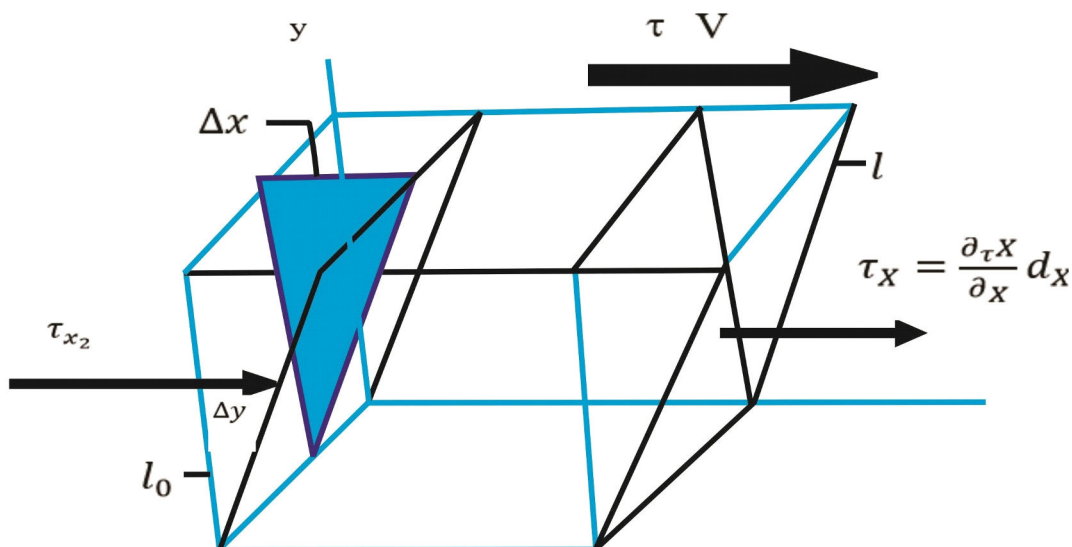


Fig. 1. Shear deformation of a cube of highly viscous fluid

$$\tau = \tau_0 + (\tau_\infty - \tau_0) \operatorname{erf}(\xi). \quad (8)$$

If $\tau_\infty \rightarrow 0$, then the solution is represented as

$$\tau = \tau_0 (1 - \operatorname{erf}(\xi)). \quad (9)$$

The error function can be defined by its own formula [1,3]:

$$\operatorname{erf}(\xi) = 1 - \exp\left(-\xi - \frac{\sqrt{\pi}}{2} \xi^2\right). \quad (10)$$

It should be noted that, depending on the nature of the boundary conditions, various analytical solutions of equation (6) can be obtained.

The variable $\xi = x/\sqrt{4vt}$ is a complex function of time and spatial coordinates.

Assuming $x = Vt$ and $y = \sqrt{vt}$, and also taking into account expressions (2) and (3), for large flow times, one can write $\xi = \lambda\dot{\gamma}$, where $\lambda = tDe/2$ is the relaxation time.

Taking these expressions into account, solution (8) takes the form:

$$\tau = \tau_0 + (\tau_\infty - \tau_0) \operatorname{erf}(\lambda\dot{\gamma}). \quad (11)$$

For power-law fluids, one can write $\xi^n = (x/\sqrt{4vt})^n$, substituting this expression into equation (6), we obtain:

$$\frac{\partial^2 \tau}{\partial \xi^{2n}} + 2\xi^n \frac{\partial \tau}{\partial \xi^n} = 0 \quad (12)$$

Introducing the auxiliary variable $z = \partial \tau / \partial \xi^n$, we rewrite equation (12) in the following form:

$$\frac{\partial z}{\partial \xi^n} = -2\xi^n z. \quad (13)$$

Solution (13) is expressed as $z = \exp(-\xi^{2n})$ or

$$\frac{\partial \tau}{\partial \xi^n} = \exp[-\xi^{2n}]. \quad (14)$$

The solution of equation (14) under condition (7) is given as

$$\tau = \tau_0 + (\tau_\infty - \tau_0) \operatorname{erf}((\lambda\dot{\gamma})^n) \quad (15)$$

Let us consider special cases of expression (15).

If we assume that $De \ll 1$, corresponding to a large duration of flow time, we can write $\operatorname{erf}((\lambda\dot{\gamma})^n) \approx (\lambda\dot{\gamma})^n$ either

$$\tau = \tau_0 + (\tau_\infty - \tau_0)(\lambda\dot{\gamma})^n = \tau_0 + k\dot{\gamma}^n. \quad (16)$$

where $k = (\tau_\infty - \tau_0)\lambda^n$ is the consistency coefficient.

Equation (16) coincides with the Hershel-Bulkley

rheological model and $n=1$ with the Bingham model.

Having expressed the specific energy dissipation per unit mass during the flow of non-Newtonian fluids as $\varepsilon_D = v\dot{\gamma}^2$ [1,3], we define the dependence of shear stress on energy dissipation considering (11) in the following form:

$$\frac{\tau - \tau_0}{\tau_\infty - \tau_0} = \operatorname{erf}\left[\left(\frac{\lambda^2 \varepsilon_D}{v}\right)^{1/2}\right]. \quad (17)$$

Using the property of the function $\operatorname{erf}(x) \rightarrow 1$ at $x \geq 3$, we can obtain the value of specific energy dissipation as $\varepsilon_D = (3/\lambda)^2 v$, at which the shear stress takes the maximum value $\tau \rightarrow \tau_\infty$ corresponding to the formation of viscoelastic disordered structures.

Rheological models of oil disperse systems

Oil suspensions and emulsions belong to multiphase systems characterised by all varieties of phenomena inherent in disperse systems. Classical features of disperse systems are: aggregate state of phases, dispersity and size of particles, concentration of dispersed phase and the nature of interaction at the interface. A special class of these systems are oil disperse systems, combining simultaneously in one volume suspensions (solid phase–oil), liquid emulsions (water–oil), gas suspensions (gas–oil) and many other forms of their existence. In these systems, the first phase is distributed in oil in the form of solid particles, water droplets, dissolved gas bubbles and their various combinations. In heavy oil, a special place is occupied by the presence of dissolved asphalt-resinous and paraffinic substances that play an important role in structure formation with inherent rheological properties. Oil disordered structured systems containing coagulation structures from crystals of high molecular weight paraffin and particles of asphaltenes-resins and forming a chain or in extreme case a continuous grid (framework), acquire the ability to flow only after destruction of this grid at $\tau \gg \tau_0$ (where τ_0 is the yield strength), and small external stresses produce elastic deformation of the grid or framework. In principle, heavy oils belong to aggregatively unstable systems where there is continuous formation and destruction of disordered structures through coagulation, coalescence, aggregation, fragmentation and fracture. Interaction of asphaltene particles is accompanied by creation of rather strong aggregates of coagulation nature and, first of all, doublets, triplets, due to Brownian diffusion motion of separate particles [9–14].

These disordered structures disintegrate into individual particles as a result of aggregate breakdown under the action of shear flow, with the equilibrium shifting towards the formation of individual particles

as the shear rate increases. It can be assumed that under the action of hydrodynamic forces there is a stretching of all bonds between particles in the aggregate up to a critical value, as a result of which this aggregate initially disintegrates into aggregates of smaller sizes, and then there is a secondary, tertiary, etc. disintegration down to a single particle. In the limiting case of infinite shear rate $\tau \gg \tau_0$ the complete breakdown of aggregates down to a single particle is possible and the flow of such oils or oil emulsions can be considered as the flow of dispersed liquids. Destruction of such structures is carried out at imposition of an external load, which corresponds to the following solution:

$$\tau = \tau_\infty + (\tau_0 - \tau_\infty) \text{erf}(\lambda \dot{\gamma}). \quad (18)$$

Equations (8) and (18) form hysteresis lines in the formation ($\tau < \tau_\infty$) and collapse ($\tau > \tau_\infty$) of disordered structures.

At the same time, it should be noted that despite the large number of publications offering various approaches in the field of rheology and filtration of structured oils, there is still no satisfactory theory linking rheological coefficients with structural and rheological properties of oil, namely, interaction forces between particles of asphalt-tar and paraffinic substances, shear stress, structure of disordered structures and size of aggregates.

Consider the dependence of shear stress on shear rate for heavy oils using formulae (10) and (11) and experimental data [6]:

$$\tau = \tau_0 + (\tau_\infty - \tau_0) \text{erf}(\lambda \dot{\gamma}), \lambda = 0.015 \text{ s}, \quad (19)$$

$$\tau = 30 + 1500 \cdot$$

$$\cdot (1 - \exp(-0.015 \dot{\gamma} - 0.886 \times 0.000225 \dot{\gamma}^2)). \quad (20)$$

Figure 2 shows the comparison of experimental values of shear stress with calculated values (20).

Effective viscosity of non-Newtonian dispersive systems

At present, there is no consensus on the mechanism of non-Newtonian flow of dispersed systems and therefore the many flow equations $\tau(\dot{\gamma})$ or rheological viscosity equations or $\eta(\tau)$ used in practice are mostly empirical or semi-empirical. For dispersed systems, using formula (11) under the condition $\tau = \tau_0 + \eta \dot{\gamma}_0 \varphi$ and $\dot{\gamma} \cong \dot{\gamma}_0 \varphi$, $\varphi > 0$, the effective viscosity coefficient taking into account the volume fraction of particles, φ , can be defined as

$$\eta = \eta_0 + (\eta_\infty - \eta_0) \text{erf}(\lambda \dot{\gamma}_0 \varphi), \quad (21)$$

where η_∞ is the maximum effective or shear viscosity of the dispersed system, corresponding to the shear viscosity of a dense packing of particles with a completed disordered structure at $\varphi_\infty = 0.74$, φ is the volume fraction of particles.

Expression (21), using (10), will be represented in the following form:

$$\eta = \eta_0 + (\eta_\infty - \eta_0) \left[1 - \exp\left(-k\varphi - \frac{\sqrt{\pi}}{2} k^2 \varphi^2\right) \right], \quad (22)$$

where $k = \lambda \dot{\gamma}_0$ is the empirical coefficient.

In the work [1], experimental studies of the

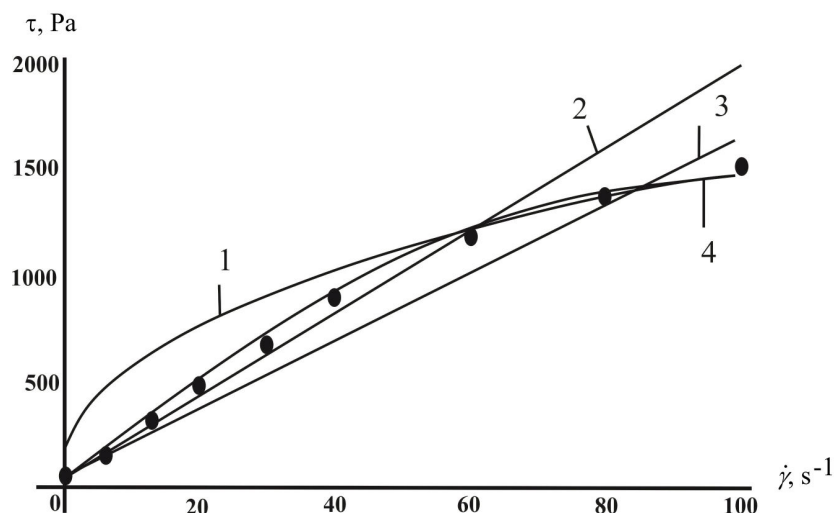


Fig. 2. Approximation of the dependence of shear stress on shear rate by different rheological models: 1 – the Hershel–Bulkley model $\tau = \tau_0 + k_0 \dot{\gamma}^n$, $\tau = 30 + 193.8 \dot{\gamma}^{0.44}$; 2 – the Ostwald-de-Ville model $\tau = k_0 \dot{\gamma}^n$, $\tau = 23.9 \dot{\gamma}^{0.92}$; 3 – the Bingham model $\tau = \tau_0 + \eta \dot{\gamma}$, $\tau = 61 + 15.75 \dot{\gamma}$; 4 – model (19), points – experiment [14]

effective viscosity of non-Newtonian oil from the content of asphaltenes at different temperatures were carried out (Fig. 3). For West Siberian oils for concentration of asphaltenes in oil in the range from 4 to 72 wt.%. Using equation (22), Figure 3 shows experimental data and calculated values of viscosity change of West Siberian oil depending on asphaltenes content.

Here

$$\ln \eta_0 = 15.77 - \frac{5360}{T + 273},$$

$$\ln \eta_\infty = 28.23 - \frac{13548}{T + 273},$$

$$\ln k = 27.3 - \frac{2258}{T},$$

T is the temperature (in °C). The calculated curves are very sensitive to the choice of initial and final values of η_0 and η_∞ , small changes in which can lead to a strong discrepancy between the experimental data and the calculated values.

When using expression (22) to describe the experimental data without value η_∞ , the problem arises of estimating the two coefficients η and k :

$$\eta = \eta_0 (1 + k_0 \varphi + k_1 \varphi^2), \quad (23) \quad \eta = \eta_0 \exp \left(k \varphi + \frac{\sqrt{\pi}}{2} k^2 \varphi^2 \right). \quad (25)$$

where k_0 and k_1 are two temperature-dependent coefficients.

The value η_∞ corresponds to the value of shear viscosity of disordered coagulation structure of dispersed medium at sufficiently high content of particles in a unit volume.

Experimental studies have shown that at low concentrations of dispersed phase the dependence of shear stress on shear rate is linear.

Using experimental data, the rheological model (8) or (13) can be represented as the Bingham equation:

$$\tau = \tau_0(\varphi) + \eta \dot{\gamma}, \quad (24)$$

where $\tau_0 = 0.06 \varphi^{1.8}$.

It should be noted that as the concentration of the dispersed phase increases, the velocity of the dispersed system decreases $V = V_0(1 - \varphi)$, resulting

$$\text{in a negative velocity gradient } \gamma = \frac{\partial V}{\partial \varphi} \frac{\partial \varphi}{\partial x} < 0.$$

Due to the difficulty in determining the value of $\eta_\infty(\varphi_\infty)$, corresponding to dense packing of particles, in the case of neglecting the term associated with η_∞ , a more practical formula for estimating the effective viscosity of the suspension at $\varphi \ll \varphi_\infty$, is the following semi-empirical expression:

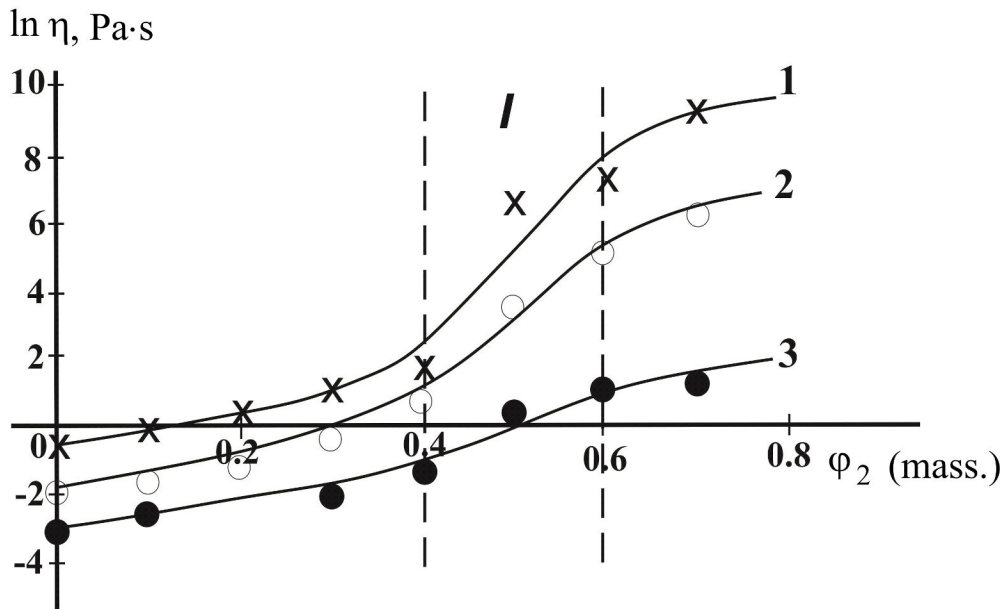


Fig. 3. Dependence of effective viscosity on the content of dispersed phase of asphaltenes at different temperatures (°C): 1 – 84; 2 – 112; 3 – 144. (I is the region of jump-like structure formation)

For highly dilute dispersed systems ($\varphi < 0.1$), the second term in (25) is insignificant, then this equation can be simplified to the following form:

$$\eta = \eta_0 \exp(k\varphi) \cong \eta_0(1 + k\varphi), \quad (26)$$

which is a modification of the Einstein equation, where $k=2.5$.

The analysis of many experimental data has shown that the k coefficient depends on the particle size, and the value of the coefficient increases with their growth.

As follows from the experimental data and from formula (24), the effective viscosity of the disperse system depends significantly on the volume fraction, size and shape of the particles. Moreover, with increasing particle size, the effective viscosity also increases. In all probability, in the limiting case, depending on the particle properties, both coagulation structures and aggregates and simple dense packing of particles with limiting porosity and shear viscosity can be formed. The effective viscosity of the dispersed system grows up to a critical value corresponding to their dense packing, which affects the velocity and character of flow. Coagulation structures are formed due to intermolecular bonds between particles, and if liquid interlayers remain between the particles, the thickness of this interlayer significantly affects the strength of the coagulation structure. The viscosity of free-dispersed systems increases as the concentration of dispersed phase increases. The presence of dispersed phase particles leads to distortion of the character of fluid flow in the vicinity of these particles, which affects the viscosity of the dispersed system [1,15]. The nature and properties of coagulation structures significantly affect the basic properties of the dispersed medium.

Heavy oil suspensions and emulsions belong to multiphase systems characterized by all the diverse phenomena inherent in disperse systems. Classical features of dispersed systems are: aggregate state of phases, dispersity and particle size, concentration of dispersed phase and the nature of interaction at the interface, which has a significant impact on the main rheological parameters (stress and shear rate) of such systems.

Conclusions

The empirical models for the rheology of high-viscosity fluids used in the literature for specific applications are formulae for adequate approximation of experimental data. Nevertheless, we note that attempts to find a general rheological equation for different systems are considered an impossible task in advance. In this study, based on flow hydrodynamics, a generalized second-order nonlinear differential

equation (6) is proposed for the rheology of highly viscous non-Newtonian fluids, reflecting the dependence of shear stress on relaxation time and shear rate. Using different boundary conditions, different analytical solutions of this equation (8)–(10) are proposed. General rheology equations for stepped fluids (12)–(15) are derived and proposed. Practical applications of these solutions for estimating shear stress as a function of shear rate and relaxation time and effective viscosity of heavy oils and dispersed systems as a function of particle volume fraction are proposed. Many specific applications of the new rheological model to solve practical problems using experimental data (20), (21), (22) and (26) are presented. Comparison of the proposed rheology models with available experimental data gave satisfactory results.

This, the proposed physically meaningful rheological models can be used in various fields related to the flow of highly viscous non-Newtonian media, such as the flow of polymer solutions or food products, etc.

Symbols

l – cube face size; P – pressure; T – temperature; t – time; V – flow velocity; x and y – spatial coordinates; ε_D – specific energy dissipation; φ – volume fraction of particles; ν – kinematic viscosity of the medium; $\dot{\gamma}$ – shear rate; λ – relaxation time; ρ – density of the medium; η – effective viscosity of the medium; η_∞ – shear viscosity corresponding to dense packing of particles; τ – shear stress; τ_0 – initial shear stress or yield stress.

Functions and indices

$\text{erf}(x)$ – error function; D – dispersed phase; 0 – initial values; ∞ – limit values; n – degree index.

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ЗАГАЛЬНЕ РЕОЛОГІЧНЕ РІВНЯННЯ ДЛЯ ВИСОКОВ'ЯЗКИХ НЕНЬЮТОНІВСЬКИХ РІДИН ТА ЙОГО ЗАСТОСУВАННЯ

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Запропоновано нове узагальнене реологічне рівняння для високов'язких рідин, яке описує взаємозв'язок між напруженням зсуву, часом релаксації та швидкістю зсуву. Наведено різні аналітичні розв'язки з граничними умовами для оцінювання напруження зсуву та ефективної в'язкості у високов'язких дисперсних системах. Також наведено загальне реологічне рівняння для рідин, що описуються степеневим законом. Практичне застосування включає оцінювання ефективної в'язкості важких нафт і дисперсних систем з урахуванням утворення та руйнування коагуляційних структур. Нова реологічна модель перевірена на експериментальних даних і показала задовільну узгодженість.

Ключові слова: реологія; нафтові дисперсні системи; високов'язкі неньютонівські рідини; узагальнене реологічне рівняння; коагуляційні структури; ефективна в'язкість; напруження зсуву; швидкість зсуву; час релаксації.

A GENERAL RHEOLOGICAL EQUATION FOR HIGHLY VISCOUS NON-NEWTONIAN FLUIDS AND ITS APPLICATIONS

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A new generalized rheological equation for highly viscous fluids is proposed, which captures the relationship between shear stress, relaxation time, and shear rate. Various analytical solutions with boundary conditions are presented to evaluate shear stress and effective viscosity in highly viscous dispersed systems. A general rheological equation for power-law fluids is also provided. Practical applications include estimating the effective viscosity of heavy oils and dispersed systems, considering the formation and the breakdown of coagulation structures. The new rheological model has been validated against experimental data, showing satisfactory agreement.

Keywords: rheology; petroleum dispersed systems; highly viscous non-Newtonian fluids; generalized rheological equation; coagulation structures; effective viscosity; shear stress; shear rate; relaxation time.

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