

## Chapter 2

### THE RELATIONSHIP BETWEEN THE PROPERTIES OF FUELS AND LUBRICANTS, CHEMMOTOLOGICAL RELIABILITY, OPERATIONAL TECHNOLOGY AND ECONOMICS OF ROAD AND AIR TRANSPORT

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#### 2.1 CHEMMOTOLOGICAL, TRIBOTECHNICAL AND RHEOLOGICAL PROPERTIES OF BOUNDARY LUBRICATING LAYERS USED IN EVALUATION OF OPERATION RELIABILITY OF TRIBOSYSTEMS

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One of the directions in increasing the reliability of modern machines and mechanisms is the development and creation of such modes of their operation that provide a dominant manifestation of external friction conditions. A comprehensive approach to the solution of these problems should include an accurate system analysis with taking into account many factors whose kinetics of change affects the anti-friction and anti-wear properties of contact. External friction is primarily determined by the structure and properties of the surface layers of metal [1], stress-strain state of contact surfaces [2, 3], structure of boundary lubricating layers [4], modes of tribocoupling operation, *etc.* An integral component of a tribotechnical system is a lubricant, the self-organization of which at various load-speed modes is an important condition for the manifestation of external friction in contact and improvement of its operation reliability.

The development of tools and methods for control of oil lubricity directly during the operation process is an urgent trend in the study of processes of tribosystem self-organization, which makes it possible to predict the operation reliability. Under a boundary lubrication mode, the interaction between two surfaces occurs in areas of meeting surface irregularities in the friction contact. Such contacts manifest themselves *via* a wide range of thermal-mechanical-chemical transformations, from elastic deformation to plastic one up to the destruction of material; increase in the friction and heat release intensity in contact, and growth of the rate of chemical reactions. As a result of these processes, organic and inorganic films are formed on friction surfaces.

Mechanisms of formation and structure of boundary lubricating layers have been investigated by U. B. Hardy, A.S. Akhmatov, F. Bowden, D. Teybor, G.V. Vinogradov, B.V. Deryagin, R.M. Matviyevskiy, I.A. Buyanovskiy, V.A. Godlevskiy, M.V. Raiko, *et al.* [5, 6, 7].

The investigation of a wide range of tribological boundary layers has shown that some of them are characterized by high wear-resistant and protective properties and some exhibit a destruction action [8]. Although the processes of physical and chemical interaction of lubricant components with the surface metal layers activated under friction are manifold, difficult to describe and still incomprehensible, there have been many models that take into account processes of contact mechanics, lubricant chemistry, molecular dynamics and thus contribute to the understanding of mechanisms of forming boundary lubricating films in the tribological contact [9, 10].

Contamination, lubricant and doping elements diffuse to the surface, which leads to a higher concentration of secondary elements near the surface than that in the bulk material. As known, solid body atoms are interconnected by various forces such as covalent, ionic, metallic, and Van der Waals'. Accordingly, the existence of these forces between the components of the boundary lubricating layers and secondary structures formed in the process of friction causes the need for different quantities of energy to break them. For some crystalline solids, different crystalline planes often exhibit different physical, mechanical, and chemical properties. In particular, friction studies

have shown that different crystalline phases are characterized by different resistance to sliding friction under the same friction conditions, which indicates their anisotropic properties [11].

In [12], tribological properties of dry, liquid, and boundary modes of lubrication were analyzed. For dry friction, it was shown that with increase in the frequency of external periodic effects, the number of harmonics decreases up to the achievement of certain frequency, upon which the surfaces completely stick together and further behave as a single whole. For a hydrodynamic mode of lubrication, there were analyzed the characteristics of friction pair with a lubricant material having properties of the Newtonian fluid as well as of pseudoplastic and dilatant non-Newtonian fluids. It was established that pseudoplastic liquids and a boundary film of lubricant material in a wide range of parameters result in realizing a stick-slip friction, which is one of the main causes of fracture of parts.

Modern lubricants consist of mineral or synthetic basic fractions and a number of multifunctional additives. The petroleum basic components generally consist of molecules containing from 18 to 40 carbon atoms and belong to three main types of hydrocarbons, namely paraffins, aromatic hydrocarbons, and naphthenes (cycloparaffins) [13]. Most of the molecules are of mixed types and contain two or more main hydrocarbon structures. These basic components also contain a small percent of heteroatoms, such as sulfur, nitrogen, and oxygen, and may transform into various stereochemical hydrocarbon structures. This is one of the reasons for the complexity in identifying the structure of formed boundary films. Generally, in typical basic components, the mass fraction of aromatic hydrocarbons ranges from 5 to 40 % (average content 20 %), straight chain paraffins fall in the range of 10 to 20 %, and cycloparaffins form the remainder. Molecules containing heteroatoms (N, S, O) are generally found in the base in the range of 0.5 to 4 % depending on oil type, processing technology, and viscosity class.

There have been many studies of physical and chemical properties of boundary lubricating films. Their findings indicate that the chemical composition of the films (predominantly micron and submicron particles of iron and iron oxides) interlace with a high molecular weight of organometallic compounds from 3000 to 100,000 [14]. The film appearance and morphology can be heterogeneous, continuous or discrete and can have various colors, from green to brown or black.

Over the last decades, many researchers have tried to measure the film strength. In studying shear strength of thin films under various high pressures (MPa to GPa), it was found that the shear strength increases with increasing pressure in films of calcium stearate, copper, and polyethylene [15]. As a rule, the film strength increases with increasing loads for basic fluids, zinc dithiophosphates, and calcium sulphonates, but it decreases with loading in the case of using friction modifiers. An increase in temperature reduces the shear strength of lubricating layers.

As shown in papers [16, 17], there is an optimal range of high lubricating properties thanks to the formation of a lubricating film on contact surfaces, which depends on external factors and the strength of its adhesion to the surface. Investigation into the structuring of boundary films has shown that the surface of triboelements, which is an integral part of reacting system, significantly influences the rates of oxidation and polymerization processes.

Improvement of test instruments and rapid development of computer technology provide a real opportunity to investigate, for the first time, tribological processes of the formation of dissipative structures under friction at the atomic and molecular levels. For example, the SFA complex allows one to measure film thickness to 0.1 nm and to detect extremely small surface forces, which gives it significant advantage over other devices and allows one to use it as a main tool for studying rheological, lubricating, and antifriction properties of tribocontact at the nanoscale [18, 19].

In calculating friction forces, the dependence of the oil viscosity on the pressure is taken into account. A lubricant in the boundary layer is characterized by anisotropic properties: in the tangential direction toward the surface, the molecular layers easily bend and, at a certain layer

thickness, slide one on another, whereas in the normal direction, the film is characterized by high compression resistance. Herein the tensile stresses on the outlying surface under rolling-with-sliding conditions are determined by the formula [20]:

$$\tau = \mu_0 \exp^{\alpha p} \frac{U \Sigma}{h} \left( 3 \frac{h-h_0}{h} + \frac{V_s}{U \Sigma} \right), \quad (1)$$

where  $\mu_0$  is the lubricant dynamic viscosity at contact input,  $\alpha$  is the piezoviscosity coefficient,  $p$  is the contact pressure,  $U \Sigma$  is the total rolling speed,  $V_s$  is the sliding velocity,  $h$  is the thickness of lubricating film,  $h_0$  is the minimum film thickness in contact, ( $h_0 = 0,8h$ ).

However, at high pressure in contact, tangential stresses determined by this formula become overestimated. To eliminate this, the following assumptions should be used: the hypothesis of limit tangential stresses ( $\tau \leq \tau_{\text{пред}}$ ); the hypothesis of oil solidification for calculation of lubricant viscosity ( $\mu \leq \mu_0 \exp(\alpha p_{\text{затв}})$ ); models of non-Newtonian liquid behavior; and semiempirical dependences for calculation of the friction coefficient [20].

The properties of boundary lubricating layers markedly differ from their bulk characteristics because of supermolecular self-organization of boundary films. Such layers play a significant role in the processes of energy dissipation in friction pairs operating in mixed and limited modes of lubrication. In addition to the type and material of surface, which determine the nature and intensity of interaction on the solid-liquid boundary, the process of fluid structuring is facilitated by the introduction of additives that increase the wear-resistant properties of lubricant due to the formation of polymolecular mesomorphic epitropic liquid-crystal structures [21, 22].

The techniques for measuring viscosity of quasi-crystalline layers of motor oils using a rotary viscometer allowed simulation of the processes occurring in the friction pair “shaft – insert of diesel engine S6A2”, in particular the shear rate and thickness of the lubricating layer [23]. In such a way, the dependence of viscosity on the shear rate was established, which confirmed the “non-Newtonian” properties of the lubricant in the narrow gap of the friction pair shaft – bearing insert. In addition, the results confirmed the assertion that boundary lubricating layers adjacent to the metal surface of the friction pair are more viscous and characterized by the orientational ordering of molecules [24]. In order to describe a lubricant state, a parameter of excess volume (which arises due to the structure disordering in solids during melting) was introduced [23]. It increases with increasing the total internal energy during the boundary film melting.

As shown in [25], there is a correlation dependence between the rheological (in particular, viscosity  $\nu$ ) and liquid crystal characteristics (boundary layer thickness and degree of orientation ordering of molecules,  $d_s \Delta n_{cp}$ ) of motor oil, which is determined experimentally. The dependence  $\nu = f(d_s \Delta n_{cp})$  is linear in the range of shear rate  $\dot{\gamma}$  (100, 150  $s^{-1}$ ). At higher shear rates it becomes exponential. Increase in the shear rate leads to a decrease in the liquid crystal characteristics of lubricant and, consequently, to a decrease in the oil viscosity in the boundary lubricating layer. This is due to the “cutting” of structured molecular layers with increasing the crankshaft rotation speed, which leads to a decrease in the oil film bearing capacity.

Principal approaches to the creation of a structural and thermodynamic friction model for a limited mode of lubrication have been developed on the basis of physical model of friction and the basic thermodynamic laws for open tribological systems [26]. Processes of the formation and destruction of adsorbed and chemically modified films under the limited supply of lubricant should be considered as processes of self-organization in thermodynamic friction systems. Herein the equations for transition temperatures used in this paper for description of the processes occurring in tribosystems under the limited mode of lubrication are nonlinear. Transitions to new steady levels occur in the case of deviation of the critical system parameters from equilibrium with subsequent self-organization at a new level.

In [27], rheological and thermodynamic models are proposed which describe melting of an ultrathin lubricating film located between atomically-smooth solid surfaces. The film melting under friction is presented as a result of the action of spontaneously arising shear stresses caused by external supercritical heating. The critical temperature of the friction surfaces which leads to the lubricant melting was established to increase with increasing a certain characteristic value of the shear viscosity and to decrease with increasing the oil shear module in a linear way. A thermodynamic model of melting of a thin lubricating layer has been constructed. Melting and solidification of lubricant are considered as first-order phase transitions. Depending on the system parameters, three lubricant states are possible under friction: solid or liquid states, or periodically melting-solidification transitions occur, which lead to a discontinuous motion of friction pairs [28].

The obtained in [29] time dependences for the friction force, the relative speed of friction surfaces during shear, and the elastic component of shear stresses in the lubricant indicate that in the liquid state the oil shear module and elastic stresses are equal to zero. An experimentally observed stick-slip friction mode is described, for which it was established that with increasing oil temperature, the frequency of phase transitions between oil structural states and the amplitude of the total friction force and elastic stresses decrease. When temperature or elastic deformations overcome certain critical values, melting of oil occurs and a kinetic sliding mode sets, in which the elastic component of friction force equals zero. It was also shown [30] that in the course of movement, a lubricant tries to acquire a uniform slide plane structure, which leads to a periodic dependence of the main parameters on time in the stick-slip mode of operation.

Functional dependences for thermal oxidation of multipurpose mineral, partially synthetic, and fully synthetic oils were obtained at static and cyclic testing temperatures, which made it possible to quantify the influence of the base and additives on oxidation processes [31]. The intensity of oxidation product formation and the oxidation rate were suggested. as indicators of the potential resource of oils in their identification and classification according to the operation properties. The mechanism of thermal oxidation of lubricant, which characterizes redistribution of absorbed heat and acts as an indicator of oxidation products formation and their volatility was clarified. The possibility of using such indicators as criteria for evaluating thermal transformations in a lubricant and self-organization processes running during thermal oxidation was shown.

This review of studies in the field of structural and rheological properties of boundary lubricating layers demonstrates that there have not yet been defined criteria for evaluating boundary films formed on contact surfaces because of the complexity of processes of forming/destroying boundary films in the tribotechnical contact. Therefore the development of structural-dynamic models characterizing the influence of boundary lubricating layers on the tribotechnical properties of contact and the kinetics of formation of secondary structures in the process of structural adaptability of triboelements under friction will allow one to predict the kinetics of changes in tribotechnical properties of friction pairs and their durability.

General aspects for selection of techniques for tribological investigations and modeling of friction processes.

The structural adaptability and evolution of a tribological system during its operation can significantly change the parameters used in predicting the friction unit resource. A numerical simulation of friction mechanisms for limited and mixed modes of lubrication, dominant in a non-stationary operation of triboelements, is a promising trend in terms of estimating changes in the properties of structural materials during operation.

In order to obtain reliable results when studying tribocoupling and to reach their reproducibility and convergence in repeated experiments, a clear structure of tribological researches is required (Fig. 1). It should include such elements as experimental means for conducting experiments (scheme and design of plants), research objects (material, construction, accuracy of manufacturing), conditions for carrying out the experiment (loading, kinematic and

temperature factors), measuring and controlling means, and techniques for processing experimental results.

The development of computer technology allows widespread propagation of simulation techniques for many engineering tasks. Simulation of tribological processes on the basis of empirical data allows one to develop adequate measures for control of tribosystems [32].

The knowledge obtained in such a way combined with practical experience in the field of chemotology and tribology will be used to improve operation of tribosystems in industry; herein it is possible to define the following main tasks:

- to optimize technological processes of designing and manufacturing materials for tribotechnical purposes in order to provide high physical and mechanical-chemical properties of materials for specific operation conditions;

- to provide continuity in the production process taking into account optimization of tribosystem design in order to increase the wear resistance of friction pairs required for a high level of product reliability;

- to reach effective characteristics of compatibility of structural elements by tribotechnical parameters for specific operation conditions on the basis of instrumental modeling techniques based on the contact mechanics calculations taking into account dynamic models of crack propagation, the depth of thermal stress propagation, and intensity of running tribochemical reactions;

- to predict the durability of tribosystems taking into account large-scale changes in their tribotechnical properties when operating on the nano-, micro-, and macro-levels.

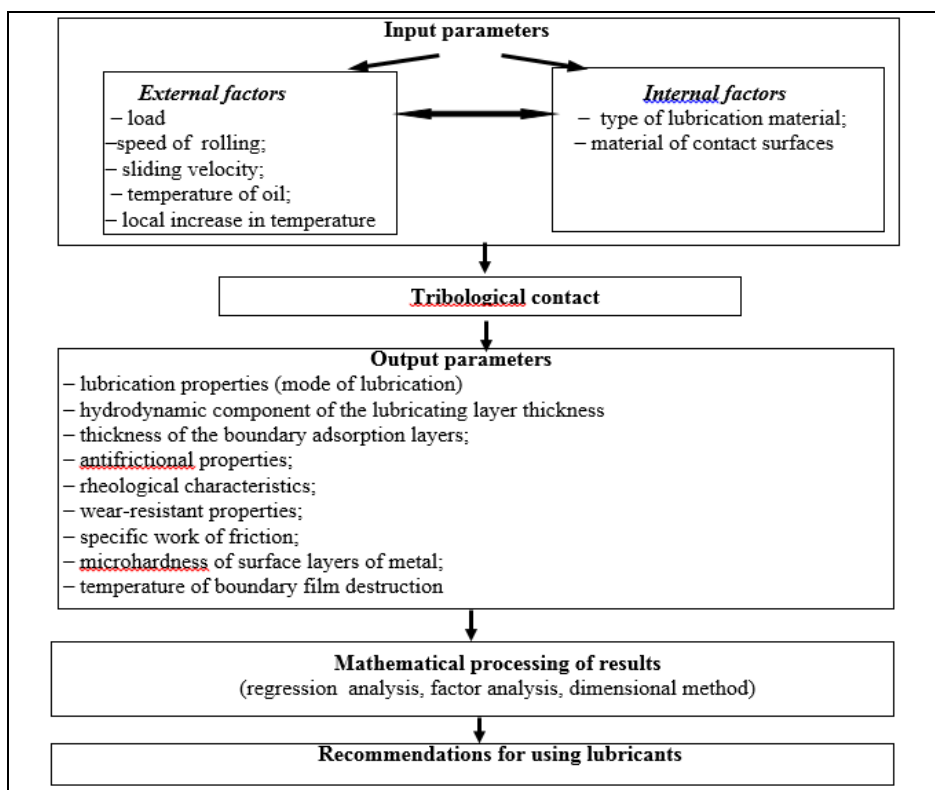


Fig. 1. Scheme of evaluation of tribological parameters in contact

The purpose of this research was to determine the dependence of the kinetics of change in the friction coefficient under the conditions of the stopping of lubricant supply on the lubrication and rheological properties of the boundary films formed on the friction-activated metal surface layers.

Investigation of the lubrication, antifriction and rheological properties of boundary films was carried out on an apparatus for testing tribological characteristics of triboelements described in [33]. The friction moment, speed of rollers, temperature of lubricant, stress drop in the lubricating layer in contact were recorded and processed using a PC (ProfiLab software) in real time with a graphical representation of their changes.

The investigated non-stationary conditions of friction implied carrying-out of cyclic experiments in a start – stationary operation – slowdown – stop regime (Fig. 2).

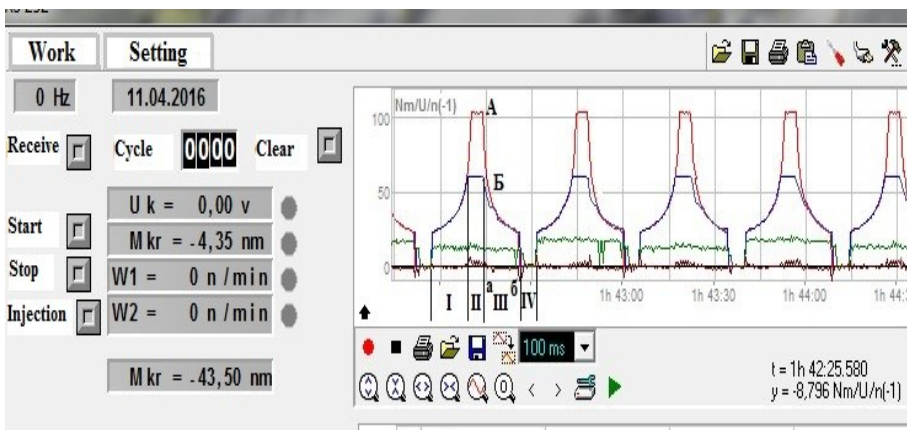


Fig. 2. Scheme of tribosystem operation in non-stationary conditions of friction: Region I – start; Region II – stationary operation; Region III– slowdown; Region IV – stop

Region I corresponds to the initial period of work of friction pairs and is characterized by a gradual increase in the rolling speed of rollers at  $V_{sl} = 0$ . In Region II, the specified maximum sliding of rollers is achieved at constant rolling speed of friction pairs. Region III corresponds to slowdown characterized by two periods: initial decrease in the speed of rollers with preservation of the specified sliding at point A and gradual decrease in the degree of sliding to zero at point B (IIIa). Further deceleration occurs in conditions of a simultaneous decrease in the speed of both rollers with maintaining  $V_{sl} = 0$  (IIIb). Region IV corresponds to a stop. If the selected cycle is projected onto an involute gearing, the pole gearing zone corresponds to Regions I and IIIb, the near-pole zone – to Region IIIa, and extreme points of gearing with maximum sliding –to Region II.

The maximum rotation speed for the outlying surface was 1000 rpm. The simulated sliding degrees were 3, 10, 20, 30, and 40 %. Maximum contact stress by Hertz was 250 MPa.

Rollers from steel 45 (HRC 38,  $R_a$  0.57  $\mu\text{m}$ ) were used as samples. Lubrication of contact surfaces was performed through immersing a low roller into a bath with oil. As a lubricant, mineral transmission oil for mechanical gearboxes and main gears of cars and trucks Okko GL-4 80w/90 was used. The oil volume temperature was 20 °C.

Adaptation of contact surfaces with a duration of 100 cycles and further work of friction pairs during 400 cycles were carried out in conditions of abundant lubrication, followed by the cut-off of lubricant supply. The total number of cycles in each experiment was as follows: 500 (abundant lubrication by immersing a low roller in an oil bath), 400 (simulation of oil starvation by

stopping of regular supply of lubricant from the bath to the contact area), 100 (forced removal of lubricant from the contact surfaces by wiping the rollers).

In conditions of the absence of lubricant supply, lubrication properties of contact is only determined by the stability of the boundary layers formed on the friction-activated contact surfaces during abundant lubrication. Up to 400 cycles after start-up, an increase in the lubricant layer thickness by 0.5–2  $\mu\text{m}$  was observed due to the availability of free lubricant material located in the contact area owing to the adhesion and cohesion forces of interaction. In case of abundant oil supply into the contact area, a hydrodynamic mode of lubrication is realized. The stopping of the lubricant supply creates preconditions for the tribosystem transition into harder operation conditions. In this case, different modes of lubrication, from semi-dry to hydrodynamic ones, take place (Table 1).

Table 1

Efficiency of lubrication at different sliding degrees of friction pairs		
Sliding degree, %	Thickness of lubricating layer, $\mu\text{m}$	Mode of lubrication by parameter $\lambda$
Below 3	0.2–5.1	0–10.86 (semi-dry – hydrodynamic)
10	0.35–4.0	0.5–7.2 (semi-dry – hydrodynamic)
20	0.2–3.8	0.5–6.7 (semi-dry – hydrodynamic)
30	0.15–2.1	0.5–5.4 (semi-dry – hydrodynamic )
40	0–1.9	0–2.7 (semi-dry – mixed)

The decrease in the hydrodynamic component of the lubricating layer thickness by on average 70 % affects the antifriction properties of contact in different ways. In Region I, corresponding to the start-up in conditions of a pure rolling process, the friction coefficient  $f$  does not change for surfaces with subsequent sliding of 10–20 %, whereas for surfaces with sliding of 30 and 40 %,  $f$  increases by 1.8 and 1.2 times, respectively. A more significant  $f$  increase (by 1.22–2.4 times) was established after 400 cycles of work, after the forced removal of free lubricant from the contact area, except for contact surfaces that further operated with a minimum sliding of 3 %, since reduction in the lubricating layer thickness does not affect the antifriction properties of contact and their friction coefficient is within 0.006–0.008, like in conditions of abundant lubrication.

In Region II, in the rolling-and-sliding conditions, a sharp  $f$  increase was fixed, by on average 3 times for sliding of 10–40 %, again except for contact surfaces characterized by a minimum sliding: their antifriction contact properties remained unchanged (Fig. 3).

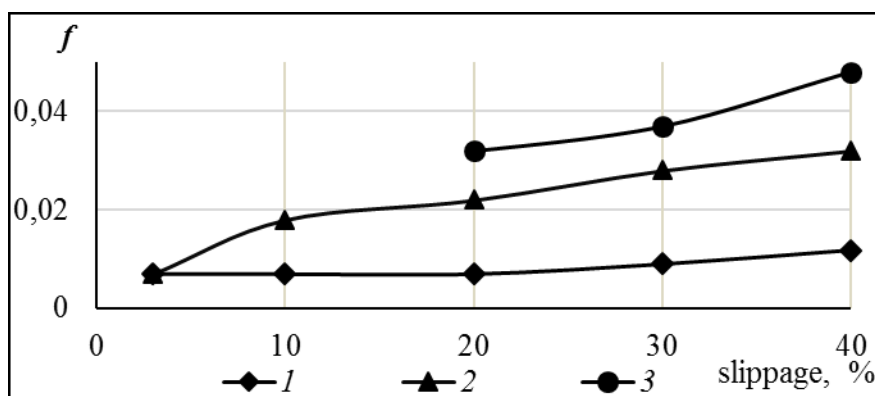
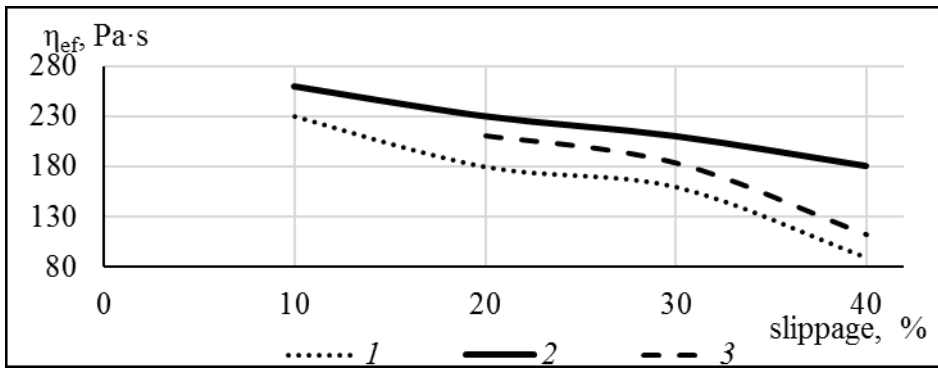
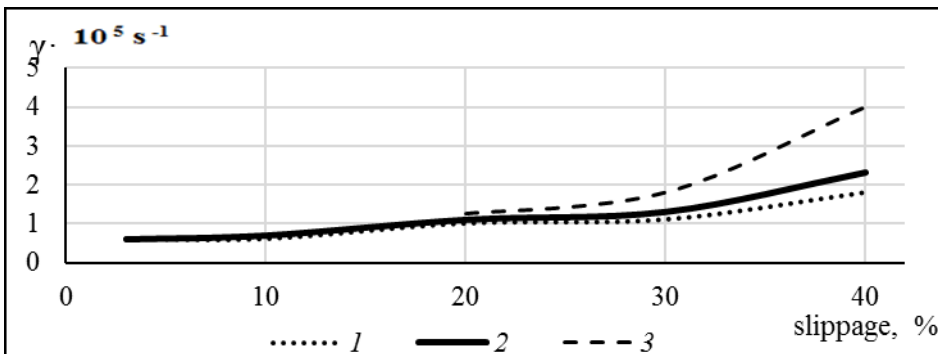


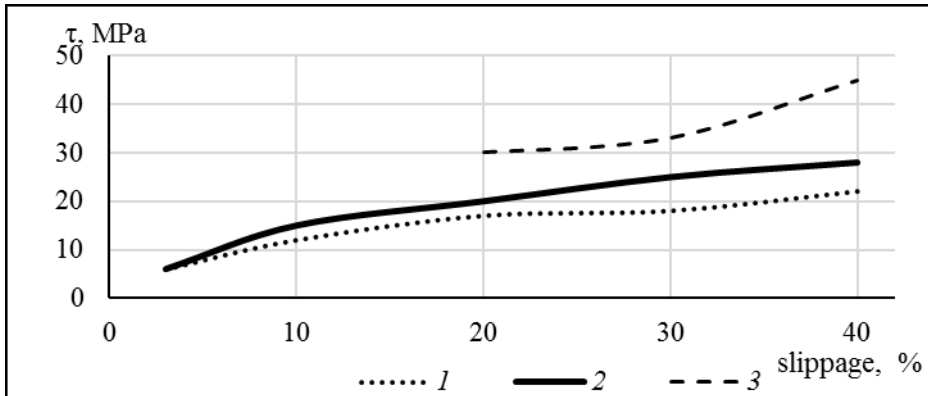
Fig. 3. Dependence of the friction coefficient on the sliding degree in conditions of limited lubrication: (1) pure rolling mode at start-up; (2) rolling-with-sliding mode; (3) moment of seizure during the rolling-with-sliding mode



(a)



(b)



(c)

Fig. 4. Influence of the sliding degree on the rheological characteristics of lubricant: (1) abundant lubrication; (2) limited lubrication; (3) forced removal of lubricant; *a* is the change in the effective viscosity  $\eta$  in contact; *b* is the change in gradient of shear rate,  $\gamma$ , in lubricating layers; *c* is the change in the shear stress,  $\tau$ , of lubricant material



The absence of excess lubricant does not significantly affect the gradient of shear rate ( $\gamma$ ) of the lubricating layers: the increase in this parameter was by 1.02–1.1 times, regardless of the degree of sliding of contact surfaces. The increase in sliding from 3 to 40 % causes growth of the shear rate gradient by 4.6 times (Fig. 4, *b*). It is this parameter that influences the stability of the formed boundary films: the increase in  $\gamma$  from  $10^5$  to  $2.3 \cdot 10^5 \text{ s}^{-1}$  with increasing sliding degree from 20 to 40 % leads to the complete abrasion of the formed boundary layers during 30 % of the operation cycles on surfaces with maximum sliding (Fig. 5).

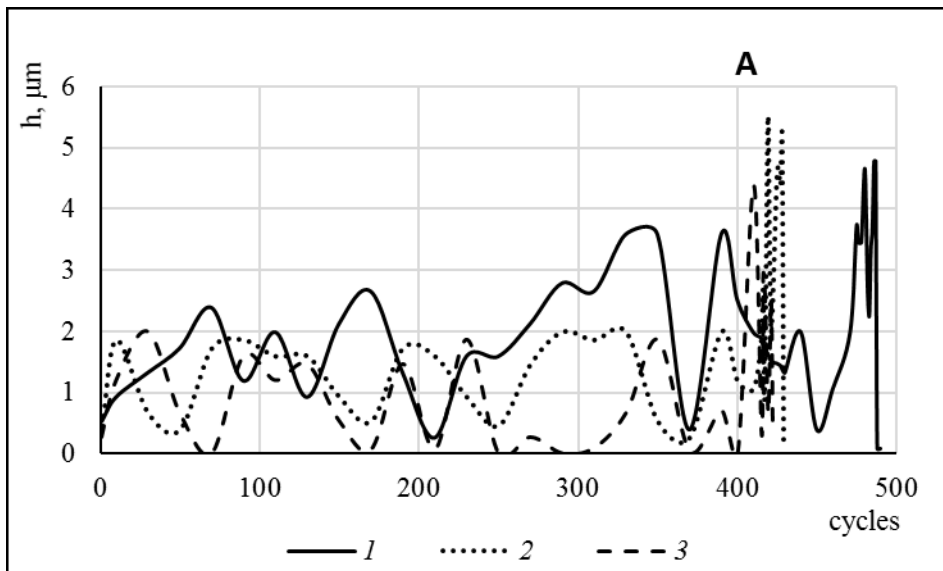


Fig. 5. Kinetics of change in the lubricating layer thickness in the stick-slip mode of operation during rolling-with-sliding at the sliding degree (1) 20 %, (2) 30 %, and (3) 40 %. Point A indicates forced removal of lubricant

With an increase in the sliding degree from 10 % ( $\gamma = 0.5 \cdot 10^5 \text{ s}^{-1}$ ) to 40 % ( $\gamma = 2.3 \cdot 10^5 \text{ s}^{-1}$ ), the effective viscosity in contact decreases from 260 to 180 Pa s (Fig. 4*a*). Up to 400 operation cycles, when the oil had not yet forcibly removed from the studied surfaces, the shear stress of the lubricating layers at a sliding degree of 10 to 40 % increased by on average 1.25 times as compared with abundant supply of lubricant. In conditions of limited supply of oil, which leads to a decrease in the lubricating layer thickness, an increase in the effective viscosity of lubricant was revealed, which was by 1.13; 1.27; 1.31, and 2.0 times for sliding degrees of 10, 20, 30, and 40 %, respectively (Fig. 4*a*).

After 400 operation cycles, the lubricant was forcibly removed from friction surfaces by wiping the rollers. For contact surfaces with a degree of sliding of 3 and 10 %, the normal operation of tribocoupling was established to the 500th cycle, the friction coefficient was stable, which testifies to high antifriction properties of boundary lubricating films. The increase in the degree of sliding to 20, 30 and 40 % leads to reduction in the period of normal operation of contact surfaces because of the appearance of the first signs of seizure corresponding to 490, 430 and 415 operation cycles, respectively. In such extreme conditions there is observed a sharp change in the antifriction characteristics of contact, which manifests itself in the periodic growth and reduction of the friction coefficient (Fig. 6).

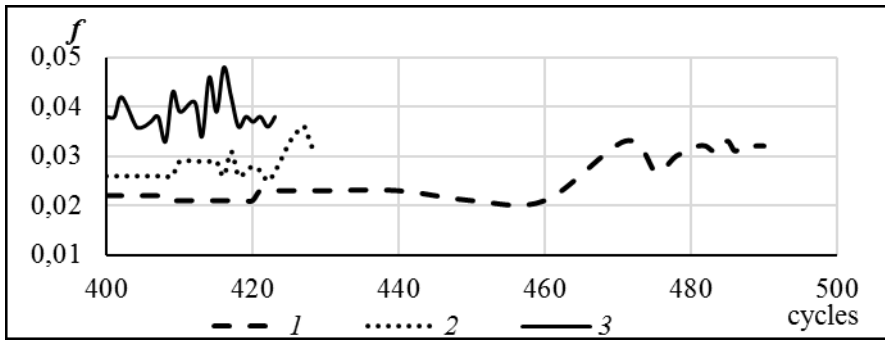


Fig. 6. Change in the friction coefficient during forced lubricant removal under rolling-with-sliding conditions at the sliding degree: (1) 20 %, (2) 30 %, and (3) 40 %

For contact surfaces that operate under minimal sliding of 3 %, no significant changes in the rheological properties of boundary lubricating layers were observed during the tribosystem transition to the mode of stopping the supply of lubricating material: stability of the shear stress of lubricating layers indicates weakness of cohesive forces of interaction.

However, with the evolution of cycles within  $400 < N < 500$ , in conditions of forced lubricant removal, no sign of seizure was detected in spite of the formation by transmission oil of boundary layers of physical nature during their structural adaptation. Two factors contribute to the preservation of normal work of friction pairs. Firstly, thanks to the reserve of lubricant in the roughness cavities of contact surfaces (operation  $R_a = 0.39 \mu\text{m}$ ,  $R_z = 1.71$ ), the lubricating layer thickness begins to grow from the 430th cycle of operation up to  $3 \mu\text{m}$ , followed by its cyclic fluctuations of  $0.2\text{--}3 \mu\text{m}$ , which prevents frictional seizure (Fig. 7).

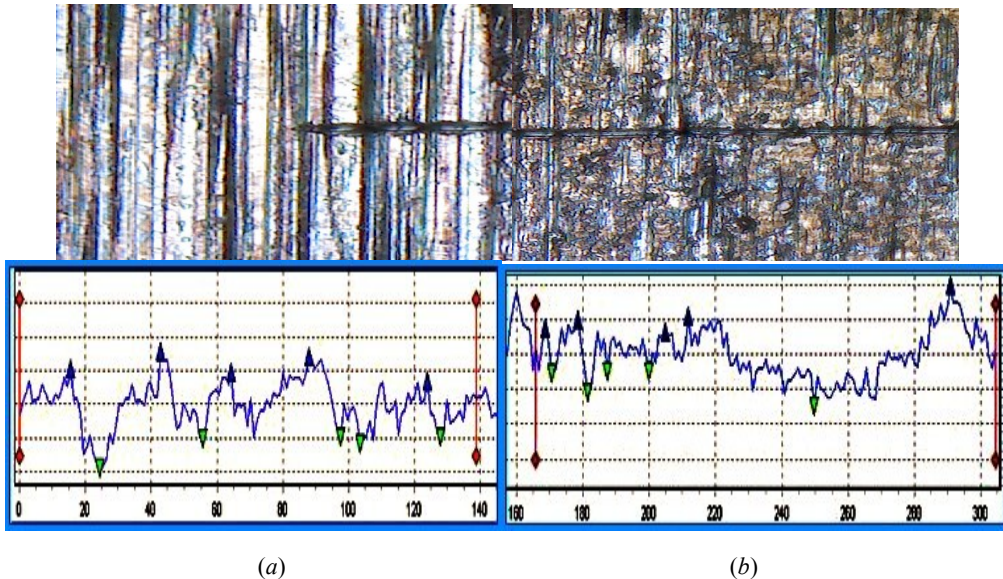


Fig. 7. Roughness of contact surfaces: (a) initial ( $R_a = 0.57 \mu\text{m}$ ,  $R_z = 2.33$ ); (b) operational ( $R_a = 0.39 \mu\text{m}$ ,  $R_z = 1.71$ )

Secondly, strongly weakened surface layers of metal due to the plasticizing Rebinder effect are characterized by low resistance to tensile shear stresses. The microhardness of surface layers decreases, by on average 1000 MPa, and the depth of amorphized weakened layer reaches 60  $\mu\text{m}$ . Thus, these processes in contact contributes to maintaining the stability of the adhesion component of the friction coefficient at the level of values established for the sufficient supply of lubricant to contact.

The friction coefficient is equal to the sum of two components: an adhesion component related to the molecular interaction of solids on the actual area of contact, and a deformation component related to the deformation of surface layers of solids under friction [34]. Therefore, analysis of the kinetics of changes in antifriction properties of contact should be based on the consideration of issues relating to both the lubricity of materials used and changes occurring in the surface metal layers under friction. In the presence of lubricant incontact, the adhesion component can be determined by the tensile shear stresses predominantly in the lubricating layer.

Let us consider the kinetics of changes in antifriction properties of transmission oil from the position of self-organization of boundary lubricating films during the tribosystem transition into extreme operation conditions, that is, without reserve of lubricant. During the start-up period, in conditions of pure rolling with increasing speed, the tensile shear stresses are localized in the central part of the lubricating layer thickness, since there is no gradient of shear rate ( $V_{sl} = 0$ ). Upon 400 cycles, owing to the growth of lubricant layer thickness due to the presence of free lubricant on the contact surfaces, the tensional stresses are localized mainly in this layer. Since the amount of lubricant is sufficient for the complete separation of the contact surfaces, in Region 1 no  $\tau$  change for surfaces with subsequent sliding of 3 to 20 % is observed,  $\tau$  is equal to 6–7 MPa like under abundant lubrication conditions. These processes contribute to the domination of the hydrodynamic mode of lubrication, in which the friction coefficient is determined by the volumetric properties of the liquid, smectic or nematic, phase of the lubricant [35]. The localization of tensile shear stresses in the liquid phase during a pure rolling process due to the minimal effect of solid surface provides the best antifriction characteristics of contact with a minimum friction coefficient at the level of 0.006–0.008 (Fig. 3).

On contact surfaces that operate at 30 and 40 % sliding, when the maximum velocity of sliding reaches 0.85 and 1.5 m/s, respectively, an intense lubricant spatter occurred. This resulted in decreasing the film thickness growth at the startup by on average 2 times, predominantly leading to a mixed lubrication mode. However, the shear stress of the lubricating layers during operation in Region I increases slightly (by on average 1.2 times) as compared with the conditions of abundant lubrication. With the lubricating film thickness decreasing, the effect of the solid metal phase on the lubricant components increases, which reduces antifriction properties: the friction coefficient increases to 0.01–0.013 (Fig. 3).

In conditions of sliding, the sliding velocity reaches maximum values, which results in appearing a gradient of the shear rate of lubricating layers through the thickness of lubricating film in contact. Because of the reduction of the hydrodynamic component of the lubricating layer thickness, the vector of tangential shear stress is localized mainly in the formed boundary layer, which is similar to the results obtained in [36].

Since the boundary layers are characterized by structuring and are subjected to the strong influence of the solid metal phase, the effect of rheological properties of lubricant on the kinetics of change in the adhesion component of the friction coefficient should be analyzed as well.

The analysis of lubricity of the investigated transmission oil in conditions of abundant lubrication revealed the formation of boundary lubricating layers of different nature. On contact surfaces with a minimum sliding of 3 %, predominantly physically adsorbed boundary films with weak Van der Waals interaction forces are formed which are characterized by low shear stresses similar to those in the bulk phase (Fig. 4c).

For contact surfaces with a sliding degree of 3 %, no change in the adhesion component of the friction coefficient was established in conditions of limited lubrication, unlike the case of abundant supply of lubricant to the contact area. First of all, this is due to low activation of surface metal layers at a slight sliding ( $V_{sl} = 0.07$  m/s). The domination of rolling speeds creates preconditions for minimum mechanical and thermal effects of the speed gradient [37], resulting in the formation of physically adsorbed boundary films. The absence of strong adhesive bonds of lubricant components with the metal surface causes frequent disorientation of the boundary layers, which manifests itself in cyclic fluctuations of the lubricating film thickness from 0.2 to 3.5  $\mu\text{m}$  in the course of 50 cycles. Such boundary layers are characterized by the rheological properties of the volumetric liquid phase of oil: their shear stress and effective contact viscosity are similar to those recorded in conditions of abundant lubrication (Fig. 4).

The increase in the degree of sliding and, consequently, in the gradient of shear rate, promotes formation of boundary lubricating layers of chemisorption nature. At a sliding of 10 to 40 %, the active lubricant components form chemisorption films and chemically modified layers on the surface, which are characterized by somewhat higher shear stresses as compared with the bulk phase owing to the action of cohesion forces.

According to [38], the shear rate gradient of about  $10^5 \dots 10^7 \text{ s}^{-1}$  leads to the destruction of lubricant components, which manifests itself in reducing lubricant viscosity. Similar results were obtained in the present studies as well: the effective contact viscosity decreased by 1.45 times with increasing the degree of sliding from 10 to 40 %. However, an analysis of rheological characteristics of structured boundary lubricating layers revealed another regularity of the kinetics of change in the effective viscosity in conditions of interrupted supply of lubricant: effective viscosity increased, and the degree of its increase correlated with the sliding (Fig. 4). These layers are characterized by increase in the shear stress due to the growth of cohesive interaction forces in the formation of a more dense and stressed structure. According to [39], polymorphism of organic molecules of lubricants, when they are structured on a solid surface, is a source of boundary layer formation by the type of netlike (reticular) heterogeneous structures. In such a boundary layer, under high pressures, reorganization of its molecular structure takes place with the formation of denser ordering, since the structure of chain molecules changes due to the axial compression of the chains because of deformation of tetrahedral angles between carbon atoms.

The used mineral transmission oil was produced on the basis of residual oil, low-viscosity distillate, and a set of multifunctional additives. Thus, it contained molecules of different stoichiometric structure. Under the action of high shear rate gradients, due to the mechanical destruction of the lubricant components, the formation of reticular boundary layers with multiple cross-links between the active centers of molecules occurs. Herein, along the boundary layer thickness, planes of slight sliding are formed, which are oriented in the plane of the action of the vector of tangential shear stresses. Such a layered netlike structure serves as a kind of reservoir for chaotically arranged inactive free molecules. The presence of such molecules is characteristic for fractions of saturated and non-polar compounds contained in the base during compounding mineral transmission oil.

As follows, ordered solid-crystalline reticular structures, similar to the framework of the dispersed phase of lubricants and containing different number of free molecules of oil, analogous to the dispersed component of lubricants, are formed on friction-activated contact surfaces. The number of inactive free components in such structures decreases with increasing degree of ordering and density of the framework (Fig. 8). Accordingly, the denser a solid-crystal structure, the higher its shear stress, which is manifested at the maximum sliding (Fig. 4).

The kinetics of change in the thickness of boundary lubricating layers after the 400<sup>th</sup> cycle of work, that is, upon forced removal of free lubricant from contact, confirms the above statement. For contact surfaces with 10 % sliding, the thickness of boundary lubricating layers varies from 0.5 to 3.2  $\mu\text{m}$ , while the rheological characteristics of chemisorption films remain unchanged.

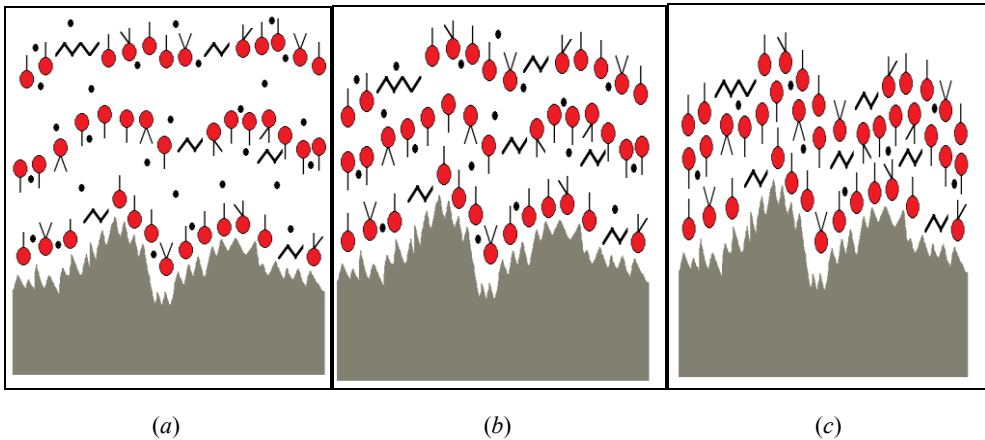


Fig. 8. Structure of boundary film from oil with multifunctional additives at the sliding degree: (a) 10 %; (b) 20 %; (c) 40 %

Stable values of the shear rate gradient and tangential stress indicate the presence of strong adhesion and cohesive bonds. The tangential stress vector is localized in the boundary layer of lubricant, the strength properties of which are high enough for the solid-crystal structure of boundary layers to remain stable. This structure of boundary films provides their high antifriction and wear-resistant properties. By the 500th cycle of operation, the normal operation of friction pairs had been established, no signs of seizure of contact surfaces were observed.

A similar peculiarities were revealed for sliding 20 % before the 450th cycle of work. Then there was observed a sharp increase in the lubricating layer thickness by 2.5 times, in shear stress by 1.5 times, and in shear rate gradient by 1.25 times. All this is connected with the destruction of cohesive and adhesive bonds in the solid-crystalline boundary layer, which leads to its disordering, melting, and transition to a smectic phase. Herein the effective viscosity in contact decreases by 1.2 times, whereas the friction coefficient drastically increases from 0.008 to 0.017 in the presence of lubricant in the friction zone. At the moments of breaking of the lubricating layer integrity, the friction coefficient increases to 0.032 and there appear the first signs of seizure of contact surfaces after the 490<sup>th</sup> operation cycle (Fig. 6).

A more intense destruction of boundary layers and earlier appearance of the first signs of seizure were revealed at 430<sup>th</sup> and 415<sup>th</sup> cycles under sliding of 30 and 40 %, respectively, after forced removal of free lubricating material from the friction zone. Thus, the strength properties of chemisorption boundary layers directly depend on the sliding velocity in contact, whose increase creates high shear rate gradients, which leads to mechanical and thermal destruction of the formed lubricating films. For example, at the moment of seizure of contact surfaces at a sliding of 40 %, the shear rate gradient increases by 2 times, the shear stress of lubricating layers does by 1.6 times, while the effective viscosity in contact decreases from 180 to 112 Pa s. In this case, there are observed sharp fluctuations in the lubricating layer thickness, whose increase can reach 4  $\mu\text{m}$ , which provides a low friction coefficient, about 0.006 (for comparison, 0.014 at the minimum film thickness). The mechanism of this process includes disorientation of boundary lubricating layers under the action of shear stresses and temperature rise in contact due to increasing sliding velocity rate, which causes melting of the solid-crystalline layer due to mechanical and temperature effects. The lubricant reacquires Newtonian properties, becomes liquid; cohesive, and adhesive forces of interaction weaken, and the volumetric liquid phase of oil increases in contact, which leads to a short-term manifestation of a hydrodynamic lubrication mode, characterized by high antifriction properties of lubricant.

The obtained data are in agreement with the postulates of the models for the first-order phase transitions between solid and liquid states [40, 41]. According to them, a sharp change in the friction force occurs at the moment of rapid change in the lubricant properties during the solid-liquid phase transition. Melting of boundary layers of lubricant can occur at the limit values of shear deformation, when the lubricant is in a liquid state, regardless of temperature [42]. In the moments of breaking the integrity of lubricating layer, the friction coefficient rapidly increases to 0.038 and 0.047 at a sliding degree of 30 and 40 %, respectively (Fig. 6).

As follows, sharp fluctuations of the friction coefficient in the manifestation of the first signs of seizure of contact surfaces are resulted from the destruction of boundary lubricating layers, which leads to a decrease in operation reliability of lubricant. These processes are characterized by the manifestation of two opposite effects. One of them causes an increase in the adhesion component of the friction coefficient owing to increasing degree of direct contact with surface metal. The other decreases the friction coefficient and provokes short-term hydrodynamic effects in contact due to local melting of boundary layers.

Reduction in the lubrication capacity of mineral transmission oil with increasing degree of sliding of contact surfaces from 3 to 40 % was established upon the tribosystem transition to extreme conditions of lubrication, which is related to decrease in the thickness of lubricating layer by 1.43 times when the tribosystem is transferred to the more rigid modes of operation, in which semi-dry and limited modes of lubrication dominate.

In conditions of pure rolling, high antifriction properties of contact are due to localization of the tensile shear stress in the liquid phase of lubricating film. In conditions of rolling-with-sliding, the vector of tangential stresses is localized in the boundary structured chemisorption films, which reduces the antifriction characteristics of contact.

The boundary layers of physical nature formed on contact surfaces with a sliding of 3 % are characterized by the identity of their rheological properties to those of the volume liquid phase of lubricant. Such structures have a strong plasticizing effect on the surface layers of metal, which manifests itself in reducing the adhesion component of the friction coefficient.

An increase in the sliding degree from 10 to 40 % causes the creation of high shear rate gradients of the lubricating layers and activates contact surfaces. These factors promote the formation of chemisorption films with an ordered layered crystalline reticular structure on friction surfaces.

An increase in the sliding degree promotes an earlier seizure of the contact surfaces, which manifests itself in the growth of the adhesion component of the friction coefficient under desorption of boundary layers. Locally, in extreme conditions, an oscillatory process of decreasing the friction coefficient was revealed, resulted from short-term hydrodynamic effects due to local melting of boundary layers.

#### РЕФЕРАТ

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#### **ХІММОТОЛОГІЧНІ, ТРИБОТЕХНІЧНІ ТА РЕОЛОГІЧНІ ВЛАСТИВОСТІ ГРАНИЧНИХ ЗМАЩУВАЛЬНИХ ШАРІВ ПРИ ОЦІНЦІ ЕКСПЛУАТАЦІЙНОЇ НАДІЙНОСТІ ТРИБОСИСТЕМИ**

Проаналізовано процеси фізико-хімічної взаємодії компонентів мастильних матеріалів з активованими в процесі тертя поверхневими шарами металу; розглянуто моделі, що враховують реологічні властивості мастильних матеріалів, що сприяють розкриттю механізмів формування граничних плівок мастильного матеріалу в триботехнічному контакті. Метою досліджень було встановлення залежності кінетики зміни коефіцієнта тертя від змащувальних та реологічних властивостей граничних плівок. Експериментально визначено закономірності зміни антифрикційних властивостей контакту від міцністних характеристик граничних плівок. Розглянуто механізми, які

приводять до підвищення адгезійної складової коефіцієнта тертя при порушенні суцільності змащувального шару або до її зниження при плавленні твердокристалічного змащувального шару.

**Ключові слова:** граничні плівки змащувального матеріалу, коефіцієнт тертя, напруга зсуву, ефективна в'язкість, градієнт швидкості зсуву.

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#### ХИММОТОЛОГИЧЕСКИЕ, ТРИБОТЕХНИЧЕСКИЕ И РЕОЛОГИЧЕСКИЕ СВОЙСТВА ГРАНИЧНЫХ СМАЗОЧНЫХ СЛОЕВ ПРИ ОЦЕНКЕ ЭКСПЛУАТАЦИОННОЙ НАДЕЖНОСТИ ТРИБОСИСТЕМЫ

Проанализированы процессы физико-химического взаимодействия компонентов смазочных материалов с активированными в процессе трения поверхностными слоями металла; рассмотрены модели, учитывающие реологические свойства смазочных материалов, способствующих раскрытию механизмов формирования граничных пленок смазочного материала в триботехнических контакте. Целью исследований являлось установление зависимости кинетики изменения коэффициента трения от смазочных и реологических свойств граничных пленок. Экспериментально определены закономерности изменения антифрикционных свойств контакта от прочностных характеристик граничных пленок. Рассмотрены механизмы, обуславливающие повышение адгезионной составляющей коэффициента трения при нарушении сплошности смазочного слоя или ее снижение при плавлении твердокристаллического смазочного слоя.

**Ключевые слова:** граничные пленки смазочного материала, коэффициент трения, напряжение сдвига, эффективная вязкость, градиент скорости сдвига.

#### ABSTRACT

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#### CHEMMOTOLOGICAL, TRIBOTECHNICAL AND RHEOLOGICAL PROPERTIES OF BOUNDARY LUBRICATING LAYERS USED IN EVALUATION OF OPERATION RELIABILITY OF TRIBOSYSTEMS

The processes of physicochemical interaction of epy components of lubricating materials with friction surface layers of metal are analyzed; models that take into account the rheological properties of lubricants that facilitate the discovery of mechanisms for the formation of boundary films of a lubricant in tribotechnical contact are considered. The aim of the studies is to determine the dependence of the kinetics of the coefficient of friction on the lubricating and rheological properties of the boundary films. The regularities of the change in the antifriction properties of the contact from the strength characteristics of the boundary films are determined experimentally. The mechanisms responsible for the increase in the adhesion component of the coefficient of friction when the continuity of the lubricating layer is disturbed or its decrease during melting of the solid-crystalline lubricating layer are considered.

**Key words:** boundary films of a lubricant, friction coefficient, shear stress, effective viscosity, gradient of shear rate.

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## **2.2 FORMATION OF COPPER (II) COORDINATION COMPOUNDS UNDER THE FRICTION PROCESS AND THEIR IMPACT ON THE TRIBOTECHNICAL CHARACTERISTICS OF THE LUBRICATING COMPOSITIONS**

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If we take into consideration the friction as a fundamental matter property, characterized by the energy transfer of physical bodies in contact, gravitational, electromagnetic and nuclear fields, then, for example, physicists point of view concerning friction, only as a complex physico-mechanical process of contacting bodies, is quite simplified. Therefore, for today, there are no definite friction laws and, as a sequence, it is revealed the difficulty of the complete physical concept representation of the frictional forces origin, as well as their quantitative evaluation and determination of all forces dependent factors. In fact, when friction forces are taken into consideration, mainly, laws of the qualitative origin are used and represented only by some approximations, which are related to actual laws, for example, the law of Amonton (1699) and the more definite law of Coulon's experimental studies (1781). According to these facts, it is logical to consider tribology (P. Jost, 1966) as an applied science of the friction, wear, lubrication and interaction of contacting friction pairs, which is based on a vast array of practical physico-chemical, mechanical, material science, technological and design studies. In this context, tribochemistry should be considered as a tribology component, which studies the chemical and physico-chemical solid surfaces transformations under the friction process, which are sequentially activated by the mechanical friction energy.