

**Mikosianchyk O.A.,
Mnatsakanov R.G.,
Khimko A.N.,
Kichata N.N.,
Yakobchuk A.E.**

National Aviation University, Ukraine
E-mail: oksana.mikos@ukr.net

**THEORETICAL ASPECTS
OF THE STRUCTURAL AND RHEOLOGICAL
STATE OF BOUNDARY LUBRICATING
LAYERS IN FRICTION PAIRS**

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Are analyzed the processes of physicochemical interaction of epy components of lubricating materials with friction surface layers of metal; are considered the 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 analyzed the modern investigations of tribological processes of dissipative structures formation at friction on atomic and molecular levels. Are systematized the results of the properties of the boundary lubricant layers, which differ significantly from their bulk characteristics, due to the supramolecular self-organization of boundary films. Is noted, that such boundary layers play a significant role in the processes of energy dissipation in the friction nodes, which operate in the modes of mixed and boundary modes of lubrication. Are considered the modern approaches of creating the structural and thermodynamic friction model under the boundary mode of lubrication. The formation and destruction processes of adsorbed and chemically modified films under the boundary lubrication mode should be considered as processes of self-organization in the thermodynamic friction system. Is presented the possibility of using the redistribution of absorbed heat as a criterion for evaluating thermal transformations in the lubricant and the processes of self-organization during thermoacidification. Are offered the general aspects of the choice of methods of tribological researches and modeling of friction processes.

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

Introduction

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.

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.

Physico - chemical properties of boundary lubrication films under the boundary mode of lubrication

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.

Rheological characteristics and orientational ordering of boundary lubricating films

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 μ_0 – is the lubricant dynamic viscosity at contact input;

α – is the piezoviscosity coefficient;

p – is the contact pressure;

U_Σ – 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 ($\tau \leq \tau_{lim}$); the hypothesis of oil solidification for calculation of lubricant viscosity ($\mu \leq \mu_0 \exp(\alpha p_{sol})$); 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 ν) 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 γ (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.

Processes of self-organization in the thermodynamic friction system

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.

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.

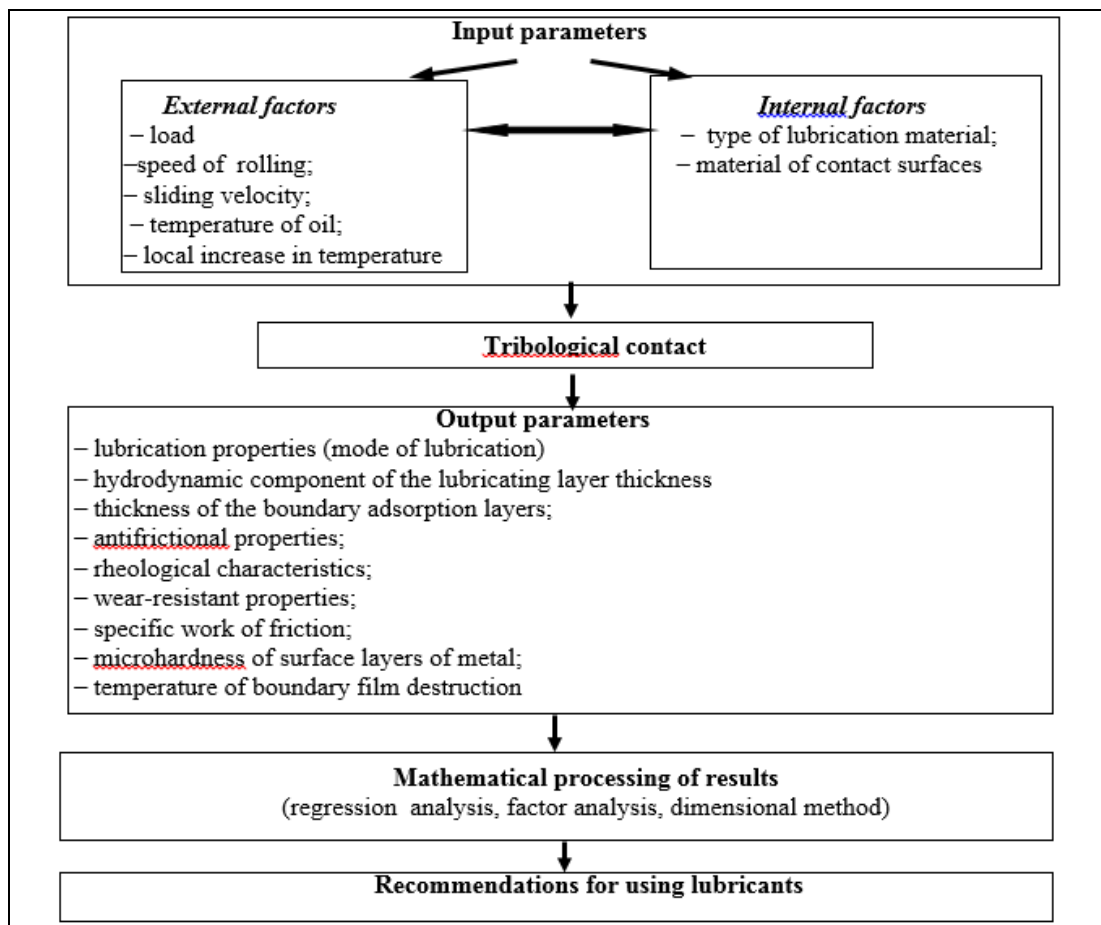


Fig. 1 – Scheme of evaluation of tribological parameters in contact

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; here in 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.

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.

Conclusions

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.

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Мікосянчик О. О., Мнацаканов Р. Г., Хімко А. М., Кічата Н. М., Якобчук О. Є. **Теоретичні аспекти структурно – реологічного стану граничних змащувальних шарів в парях тертя.**

Проаналізовані процеси фізико-хімічної взаємодії компонентів мастильних матеріалів з активованими в процесі тертя поверхневими шарами металу; розглянуті моделі, що враховують реологічні властивості мастильних матеріалів, які сприяють розкриттю механізмів формування граничних плівок мастильного матеріалу в триботехнічному контакті. Проаналізовані сучасні дослідження трибологічних процесів утворення дисипативних структур при терті на атомарному і молекулярному рівнях. Розглянуті сучасні підходи щодо створення структурно-термодинамічної моделі тертя при граничному режимі мащення. Запропоновані загальні аспекти вибору методик трибологічних досліджень і моделювання процесів тертя.

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

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