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DEVELOPMENT OF A RHEOLOGICAL MODEL OF STRESS RELAXATION IN THE STRUCTURE OF AN OIL FILM ON THE FRICTION SURFACE WITH FULLERENE ADDITIVES

Andrii Kravtsov
Corresponding author
PhD, Associate Professor
Department of Transport Technologies and Logistics*
E-mail: kravcov_84@ukr.net
Anastasiia Suska
Doctor of Economic Sciences,
Associate Professor, Head of Department
Department of Woodworking Technologies and System Engineering of the Forest Complex*
Abliatif Biekirov
PhD
Department of Aviation Engineering Support
Ivan Kozhedub Kharkiv National Air Force University
Sumska str., 77/79, Kharkiv, Ukraine, 61023
Dmytro Levkin
PhD, Associate Professor
Department of Higher Mathematics*
*Kharkiv Petro Vasylenko
National Technical University of Agriculture
Alchevskykh str., 44, Kharkiv, Ukraine, 61002

A rheological model of stress relaxation in a thin lubricant film, which is formed on the friction surface under the influence of the force field of the friction surface in the presence of fullerene compositions in lubricants, was developed. Analysis of the model made it possible to establish that the existence of elastic or viscous properties in surface structures depends on the ratio of two parameters. This is the time of stress relaxation in the structure on spots of actual contact and the duration of stress action on these spots, which is termed the lifetime of an actual contact spot.

It was shown that an increase in the sliding rate reduces the time of relaxation of stresses in the surface structure. This is due to the destruction of aggregates in the structure of gel and the appearance of rotational movements of separate units – flocs. An increase in the load on the tribosystem significantly increases the value of relaxation time. This is due to squeezing the viscous component out of the structure of a surface film. It was established that if the relaxation time exceeds the duration of actions of stresses on actual contact spots, the structure of a surface film behaves like an elastic solid. Conversely, if relaxation time becomes shorter than the duration of stress action, the film behaves like a viscous medium.

Theoretically, it was shown that in the range of sliding and loading rates, when a film behaves like an elastic solid, a decrease in stresses on actual contact spots does not exceed the values of 1.1–22.8 %. This property provides the bearing capacity of a film. The development of the model will make it possible to simulate elastic and viscous properties of "stitched" structures and substantiate the rational concentrations of additives to lubricants, as well as the ranges of their use

Keywords: tribosystem, fullerene compositions, dynamic viscosity, structural viscosity, clusters, micelles, rheological model, stress relaxation, lubricant film, lubricants

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1. Introduction

With the development of nanomaterials, in tribology, it was possible to effectively use functional additives in the form of geomodifiers [1, 2]. It was revealed that oils modified with nanomaterials, which form dispersed systems in lubricants, increase the durability of working surfaces of different types of tribosystems. The study of the formation of the optimal composition of tribological additives to base lubricants showed the possibility of improving the characteristics of lubricants. It was found that the use of nanomaterials makes it possible to form functional surface layers that enhance the resource of tribosystems.

The existence of dispersed phase in the lubricant material in the form of clusters and micelles, which are formed around fullerene molecules, creates wear-resistant structures on the surface of friction that take and relax stresses on actual contact spots.

The use of fullerenes as additives to lubricants opens a new direction in the creation of lubricants with special properties. Works on the creation of lubricants with fullerene additives are underway in all countries. Such lubricants respond to external effects on a tribosystem and are able to change the structure of surface layers, adapting to operating conditions. Control over the process of formation of such structures will improve durability and decrease friction losses of various machines and mechanisms, which will help to save energy during operation.

One of the ways to solve this problem is to develop a model of relaxation of stresses on spots of actual contact and to simulate such processes in friction-based structures formed on the surface. The results of the simulation will make it possible to substantiate the composition and the content of fullerene additives to lubricants for various purposes.

2. Literature review and problem statement

Paper [3] provides an overview of the literature on lubricants with the addition of nanoparticles. The effect of nanoparticles on tribotechnical characteristics of lubricants was analyzed. It was noted that the use of nano-additives leads to an increase in viscosity of the base medium, high bearing capacity of mating, a decrease in friction factor, and an increase in durability. Analysis of the conclusions of the paper reveals the prospects of applying nano additions to liquid lubricants. However, the problem of optimal concentrations of nanoparticles in the basic lubricant remains unresolved.

Paper [4] deals with additives of carbon nanoparticles to plastic lubricants. It was shown that the effect of the injection significantly depends on the type of lubricant and can both improve and worsen the tribotechnical indicators of a lubricant. At the same time, the authors received an increase in a load of burr, regardless of the type of a lubricant-base, in all types of explored lubricants during the introduction of carbon nanoparticles. Such contradictory data lead to a conclusion about a number of unresolved issues related to the technologies of the introduction of carbon nanoparticles into basic materials.

The authors of paper [5] also conclude that the use of nanomaterials in lubricants is promising. The research contains conclusions that the characteristics of lubricants can be improved by the use of nano-additives. The paper is devoted to an information review of the use of nano-additives to liquid lubricants and the prospects for their use in the production of lubricants. However, the paper does not provide data on the mechanisms of lubricants when forming structures on friction surfaces.

Some important aspects of the use of nanoparticles, which address different technologies and concentrations, are presented in paper [6]. When modifying lubricants, the authors take into consideration the following factors: the size of the nanoparticles, shape, structure, modification of the friction surface, particle concentration, physical and chemical properties of friction surfaces It was observed that when nano-additives are added to lubricants, even at low concentrations, friction and wear rate decreased significantly. In addition, carbon nanotubes and graphene are environmentally friendly and safe materials. Based on the studies, the authors positively assess the prospects of using these nanomaterials to modify lubricants.

In articles [7, 8], the studies of high-purification fullerenes and their addition to the base lubricant were carried out. The research was performed on a four-ball friction machine. The results showed that the use of fullerenes reduces the diameter of the wear spot at different concentrations in the base lubricant. There was a film on the friction surface. The authors of the paper recommend such lubricants for sliding bearings, as well as for solid lubricants. At the same time, the concentrations of additives in the base lubricant varied in the interval of 0.5–2.0 % by weight.

Further development of various nanomaterial technologies is presented in research [9]. The authors examined both solid lubricants with fullerene additives, including graphene (G), fullerene (C60), and carbon nanotubes, as well as liquid lubricants. The authors note that the lubricant system, consisting of amorphous carbon and liquid lubricant, is a promising lubricant and has good tribological characteristics. Compared to pure base lubricant, the use of fullerenes reduces the friction factor, which provides the best anti-friction properties. The authors suggest that a block solid protective film is formed on the friction surface, which significantly smoothens the surface of friction, weakens intermolecular interaction, and thus promotes the mobility of lubricant molecules.

The authors of paper [10] note that when fullerenes are used, the friction factor is reduced to a greater extent than the wear rate. The authors examined a low-viscous compressor oil with the addition of fullerenes of 0.1 % vol. As a result, the friction factor decreased by 90 % compared to the base lubricant. The authors conclude that the use of fullerene additives for the operation of compressors is promising. Similar results in the use of fullerenes in compressor oils of household refrigerators were obtained in papers [11, 12]. The authors provide the data on an increase in the compressor efficiency ratio by 5.6 % and 5.3 %, respectively. The prospect of fullerene use in many technical fields is noted in research [13, 14]. However, the presented papers do not pay attention to thin surface structures on friction surfaces and their role in reducing friction and wear.

The approach to the study of surface structures is demonstrated in paper [15]. It contains a review on the tribology of ultra-thin films, which focuses on the tribological properties of Langmuir-Blodgett films, self-organizing monolayers, and films of molecular deposition. The results of studies of various factors influencing the tribological properties of films and progress in the application of molecular dynamics modeling to study the mechanisms of friction and lubrication were presented.

A similar approach to the self-organization of films is presented by the authors of paper [16]. Experimentally, the method for the improvement of tribological properties of lubricants by introducing fine powder fullerenes in basic technical oils is ineffective. The reason is the low solubility of fullerenes in technical oils. The option of eliminating such a shortcoming was proposed in the abovementioned paper. To do this, it is necessary to pre-disperse fullerenes in vegetable high-olein oils, such as rapeseed, and then add this composition to technical greases. Experimental studies on changing the friction factor, which decreases by 86 %, which proves the effectiveness of this approach, were presented.

This approach, which uses the pre-dispersion of fullerenes in vegetable oils, is used in studies [16, 17]. According to the findings of these works, the aggregates of the disperse phase, united by external electrostatic forces into a solid grid (frame) on the friction surface, acquire the properties of a "solid body". A small external load causes elastic deformation of the frame. At a fairly high load, the frame is destroyed, and separate aggregates are disintegrated.

Article [18] presents a macro-rheological model of stress relaxation on spots of actual contact of a tribosystem in the presence of a solid grid of fullerene molecules on the surface of friction. The microrheological model is presented in the form of differential equations of the second order and their solutions. A working hypothesis of reducing friction forces in tribosystems in the presence of a disperse phase in the lubricant material was formulated.

Articles [21, 22] focus on the practical features of the use of lubricants with functional additives that provide a positive effect both in the manufacturing process and during the operation of tribosystem parts. At the same time, the mechanisms of action of lubricants on operational factors were not analyzed.

The presented analysis of the use of fullerenes in lubricants suggests the need to study thin structures in the form of films on friction surfaces containing fullerene molecules. It is necessary to establish the mechanisms of their formation and criteria for the transition of elastic properties of such structures into viscous properties. Such structures are involved in the relaxation of stresses on spots of actual contact during the operation of tribosystems, thereby reducing the wear rate and friction factor, which will enhance the resource of machines and decrease friction losses.

The development of a rheological model of stress relaxation will make it possible to simulate elastic and viscous properties of "stitched" structures and substantiate the rational concentrations of additives to lubricants, as well as the ranges of their application.

3. The aim and objectives of the study

The aim of this study is to develop and investigate a rheological model of stress relaxation in the structure of a lubricant film on the surface of friction of tribosystems in the presence of fullerenes in lubricants. This will make it possible to develop new lubricants containing nano-additives and determine the rational areas of their application.

To accomplish the aim, the following tasks have been set:

– to substantiate the structure of the rheological model of stress relaxation in a lubricant film on the surface of friction of tribosystems in the presence of fullerenes in lubricant material;

– to perform simulations of the stress relaxation process at different concentrations of fullerenes in lubricants and different conditions of operation of tribosystems;

– to substantiate the criteria for the transition of elastic properties into viscous in the structures of a lubricant film on the friction surface containing fullerene compositions.

4. The study materials and methods

In the development of a rheological model of stress relaxation in the structure of a lubricant film on the friction surface in the presence of clusters and micelles of fullerenes in basic lubricant material, the following assumptions were accepted.

The dispersion of clusters and micelles near the friction surface (in the field of action of electrostatic surface forces), is accepted as the structure of the gel [18]. In this case, the forces of electrostatic interaction operate between the micelles and the friction surface. Electrostatic forces contribute to the formation of a frame from aggregates, the cavities between which are filled with viscous liquid. This structure has elastic-viscous properties. Inter-micellar forces can relax, respectively, the structure behaves like Maxwell's body, Fig. 1 [18]. In this structure, stresses are received by elastic elements of aggregates and transferred to a viscous liquid medium. Due to the existence of viscous liquid between clusters and micelles, the elastic deformation during the load does not occur instantly but proceeds with lagging.

The scheme of the elastic-viscous gel is shown in Fig. 1. The presented scheme is borrowed from paper [18], where *N* is the load, dimensionality, *N*.

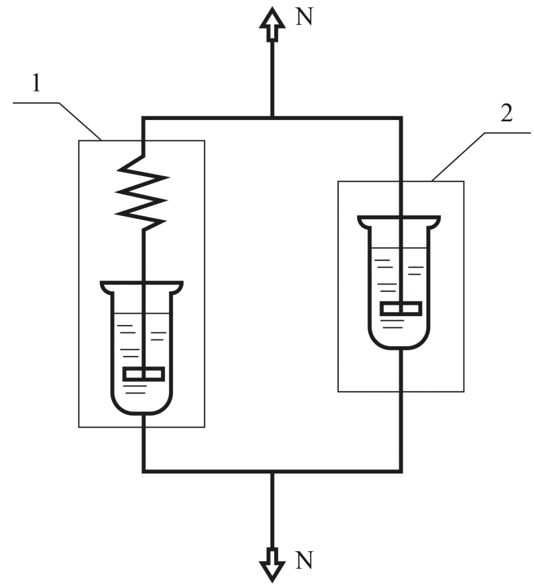

Fig. 1. The scheme of the rheological model of elastic-viscous gel: 1 – Maxwell's body; 2 – Newton's body; N – load

The methodical basis of the research is obtaining a rheological equation according to the scheme shown in Fig. 1. The solution of the rheological equation is a mathematical model of stress relaxation in the structure of a lubricant film on the surface of friction of tribosystems in the presence of fullerenes in lubricants. Analysis of the model made it possible to obtain data on the existence of elastic or viscous properties in surface stitched structures and the criterion of transition of elastic properties into viscous and, conversely, viscous properties into elastic ones.

5. Results of the study of stress relaxation in the lubricant film structure on the friction surface

5. 1. Substantiation of the structure of the rheological model of stress relaxation in a lubricant film on the friction surface of tribosystems in the presence of fullerenes in lubricant material

The substantiation of the structure of the rheological model will be based on the presented scheme, Fig. 1. Using the methodical approach outlined in paper [18], we will write down the expression for the rheological equation in the form of a second-order differential equation:

$$\sigma_{acs} + \frac{d\sigma_{acs}}{dt} \cdot T_{rel,g} = 2\mu_g \left(\frac{d\varepsilon_g}{dt} + \frac{d^2\varepsilon_g}{dt^2} \cdot T_{lag,g} \right), \quad (1)$$

where σ_{acs} is the stresses on spots of actual contact, Pa;

σ_{acs}/dt is the rate of a change in stresses on spots of actual contact, Pa/s;

t is the time of action of stresses on spots of actual contact, s;

$T_{rel,g}$ is the time of stress relaxation in the structure of elastic-viscous gel, s;

μ_g is the dynamic structural viscosity of the structure of elastic-viscous gel, Pa·s;

$d\varepsilon/dt$ and $d^2\varepsilon/dt^2$ are the rate and acceleration of deformation in the structure of elastic-viscous gel, 1/s and 1/s^2;

$T_{lag,g}$ is the time of a lag of deformation in the structure of elastic-viscous gel, s.

The solution to differential equation (1) is the expression:

$$\sigma_{acs}(t)=\sigma_{acs}\cdot\exp\left(-\frac{t}{T_{rel.g}}\right), \qquad (2)$$

where $\sigma_{acs}(t)$ is the stresses on actual contact spots as a time function, Pa.

The solution to differential equation (2) makes it possible to simulate the process of relaxation of stresses on spots of actual contact in tribosystems. This will determine the magnitude of stresses that the structure of the elastic-viscous gel receives during the operation of a tribosystem and the time of stress influence.

The next step of the study is to determine the parameters that are part of the solution of differential equation (2).

Stresses on spots of actual contact are determined according to research [19].

The time of relaxation of stresses in the structure of the gel, which is formed by clusters and micelle of fullerenes on the friction surface in the region of action of electrostatic forces of the friction surface, is determined from the following expression:

$$T_{\text{rel,g}}=\frac{\mu_g}{G},\ \text{m}. \qquad (3)$$

The dynamic viscosity of the formed gel structure on the friction surface $\mu_г$ is determined from formulas that are shown in paper [20].

The given module of gel structure is found from the following expression:

$$G_{\text{red,g}}=\frac{E_{\text{red,g}}}{(2+2\cdot\nu_P)},\ \text{Pa}, \qquad (4)$$

where $E_{\text{red,g}}$ is the reduced module of gel structure elasticity, determined from the formulas presented in research [20]; ν_P is the Poisson factor of disperse phase equal to 0.3.

The existence of an actual contact spot was called in this research the "life" time of an actual contact spot. The lifetime depends on the diameter of a contact spot and sliding rate, it is calculated from formulas of paper [19]:

$$t_{sr}=\frac{d_{acs}}{\nu},\ \text{s}, \qquad (5)$$

where d_{acs} and v are diameters of an actual contact spot, dimensionality – m, and sliding rate, dimensionality – m/s.

5. 2. Simulation of the stress relaxation process at different concentrations of fullerenes

Analysis of expression (2), which is the solution to the differential equation of a rheological model (1), makes it possible to draw the following conclusion. The process of relaxation of stresses in a thin surface structure is affected by the time of relaxation and the lifetime of an actual contact spot.

The dependences of a change in relaxation time of $T_{\text{rel,g}}$ by a structure of a thin oil film on the friction surface (the gel structure), are shown in Fig. 2, 3.

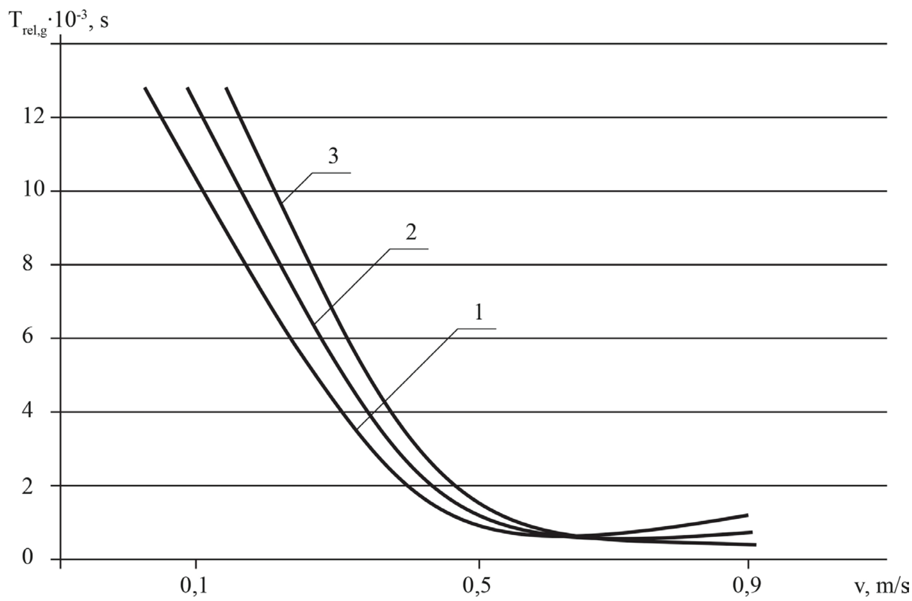

Fig. 2. Dependences of change in relaxation time in the gel structure on sliding rate and concentration of fullerenes (*F*) in the base lubricant medium: 1 – 0.5 % F; 2 – 1.0 % F; 3 – 1.5 % F

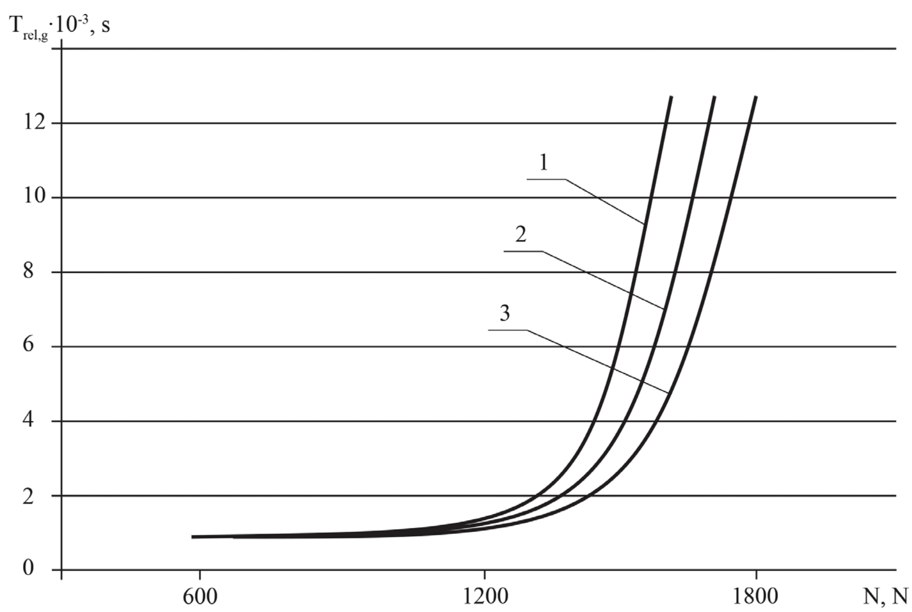

Fig. 3. Dependences of a change in relaxation time in the gel structure on load and concentration of fullerenes (*F*) in the base lubricant medium: 1 – 0.5 % F; 2 – 1.0 % F; 3 – 1.5 % F

The following tribosystem was chosen for simulation: movable triboelement steel 40X (HRC52); non-movable triboelement Br.AZh 9-4 (HB 100). The friction area of movable triboelement F_{fr}=0.0003 m^2, that of non-movable element F_{fr}=0.00015 m^2. The load on the tribosystem is 1,000 H; the basic lubricating medium SAE 10W30 with fullerene compositions of different concentrations of 0.5–1.5 % by weight. The scheme of mating "ring – ring". For this tribosystem, according to the procedure presented in paper [19], we calculated: d_{acs}=6.825·10^{-5} m; t_l=13.65·10^{-5} s (for v=0.5 m/s).

As it follows from the presented theoretical dependences in Fig. 2, an increase in sliding rate significantly reduces stress relaxation time in the gel structure. This structure is formed on the friction surface in the form of a film under the influence of a force electrostatic field created by the friction surface.

It follows from the dependences shown in Fig. 3 that an increase in load significantly increases the time of stress relaxation in the structure of the gel. This increase can be explained by squeezing the viscous liquid under the load of the gel structure. The concentration of fullerenes (F, % by weight) in the base lubricant does not have a significant effect on the magnitude of relaxation time in the gel structure.

The dependences of changes in stresses on spots of actual contact in the gel structure on time of stress action at different loads on a tribosystem are shown in Fig. 4. For the design of a tribosystem, which was chosen for research, the lifetime of an actual contact spot, at a sliding rate v=0.5 m/s, is 10.5·10^{-5}–12.9·10^{-5} s. A smaller amount of time corresponds to a lower load.

As the load increases, the spot diameter increases slightly, the number of spots increases to a greater extent. Relaxation time for such conditions of functioning is 39·10^{-5}–305·10^{-5} s. A large magnitude of relaxation time refers to a greater load.

Analysis of the simulation results makes it possible to conclude that the duration of stresses influence, which is equal to the lifetime of an actual contact spot t_l is less than relaxation time $T_{rel,g}$. During the lifetime of a contact spot, stresses decrease by 2.5–22.8 %, (Fig. 4).

Dependencies of a change in stresses on spots of actual contact at different sliding rates are represented in Fig. 5. The resulting dependences suggest that the lifetime of an actual contact spot at a constant load N=1,000 N is 59.4·10^{-5}–6.6·10^{-5} s. A larger magnitude of time corresponds to a lower sliding rate.

As the sliding rate increases, the lifetime of an actual contact spot decreases. Relaxation time for such operating conditions is 913·10^{-5}–28.1·10^{-5} s. A larger magnitude of relaxation time refers to a lower sliding rate.

The simulation results show that in the explored range of rates and loads, the duration of action of stresses is smaller than the relaxation time. During the lifetime of action spot, stresses decrease by 1.1–19.18 %, Fig. 5.

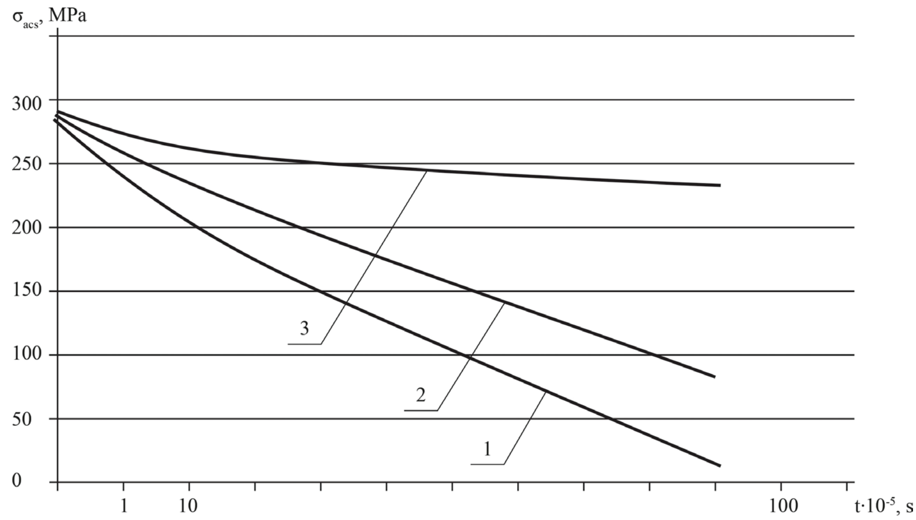

Fig. 4. Dependences of a change in stresses on actual contact spots in the gel structure on the duration of action of stresses at different loads N on a tribosystem: 1 — 500 N; 2 — 1,000 N; 3 — 1,500 N

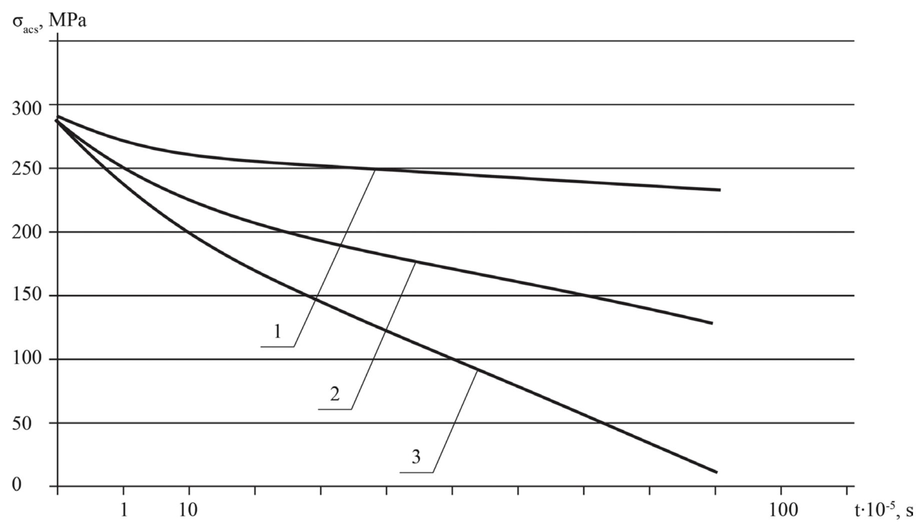

Fig. 5. Dependences of a change in stresses on actual contact spots in the gel structure on the duration of action of stresses at different sliding rates v: 1 — 0.1 m/s; 2 — 0.5 m/s; 3 — 0.9 m/s

5. 3. Substantiation of criteria for the transition of elastic properties of a lubricant film into viscous properties

The simulation results make it possible to substantiate the criteria for the transition of elastic properties into viscous and, vice versa viscous into elastic in the presence of fullerene compositions of different concentrations.

The ratio of the lifetime of an actual contact spot (the duration of action of stresses) to the time of stress relaxation in the structure is a criterion for assessing the properties of a structure. This dimensionless magnitude characterizes the transition of elastic properties of a lubricant film into viscous and vice versa, viscous into elastic. If this magnitude is smaller than unity, then, according to paper [20], the structure of a film behaves like a solid. Conversely, if it is more than unity, the structure of a film behaves like a viscous medium.

The simulation results show that the duration of action of stresses is less than relaxation time in the entire chosen range of changes in sliding and load rates. Consequently, the gel structure behaves like a solid elastic body.

At a sliding rate of more than 1 m/s (load 1,000 N), there is a loss of elastic properties and the gel structure behaves like a viscous medium.

6. Discussion of results of studying the rheological properties of structures that are formed on the friction surface of tribological systems

The developed mathematical model (2) of relaxation of stresses in the structure of an oil film on the surface of friction of tribosystems makes it possible to explore the properties of surface structures in the presence of fullerene compositions in lubricants. For example, analysis of dependences presented in Fig. 2 reveals that an increase in sliding rate decreases the magnitude of stress relaxation time in the structure of a surface film. If the relaxation time becomes equal to the time of stress influence (lifetime of a contact spot), then, according to paper [20], there is a transition from elastic properties in the gel structure to viscous ones. Conversely, if the relaxation time of the gel structure far exceeds the duration of the action of stresses, the gel structure has elastic properties and the structure behaves like a solid body.

The obtained result differs from the well-known model [18] because it makes it possible to determine the concentration of fullerene composition, a sliding rate, and load during operation, where elastic properties of surface stitched structures will be ensured.

The presented results continue the studies that are outlined in papers [16–20]. They differ from those known by the fact that they represent the dependences and the criterion for assessing elastic or viscous properties of surface structures, taking into consideration technological and operational factors that influence a tribological system.

The developed methodological approach can be used in studying small-scale systems of different origins. Such compounds are introduced into lubricants in the form of additives. Using the developed model, it is possible to determine the optimal concentrations of nano-additives in the basic lubricants, ranges, and operating conditions of these materials. This will make it possible to ensure the formation of friction-resistant films with elastic properties on the friction surface. The creation of next-generation lubricants, such as motor or transmission oils, will increase the time it takes to replace them, reducing the cost of operating machines.

According to the presented studies, fullerene compositions have rational modes of application. Such modes are determined using the proposed criterion for the transition of elastic properties of surface films into viscous films. That is why it is necessary to calculate for specific tribosystems the magnitudes of loads and sliding rates, where surface structures will have elastic properties. If the values of these factors are exceeded, the use of fullerene compositions will not create a positive effect. This explains the failures of some researchers in the use of fullerenes in lubricants.

Further research involves the creation of sedimentary resistant fullerene compositions in lubricants of different groups of operation. This requires additional research into the use of different solvents of fullerene, their concentrations, and their influence on the tribological characteristics of base lubricants.

7. Conclusions

1. The structure of the rheological model of stress relaxation in a thin lubricant film was substantiated, which is the result of the solution of the first task of research. The rheological model differs from the known ones because it takes into consideration the structure of the formed film on actual contact spots accounting for the lifetime of a contact spot. It was shown that the lifetime of an actual contact spot depends on the structural, technological, and operational factors of a tribosystem. The formulas for calculating the values of the lifetime of contact spots (5) were presented. It was shown that for the studied tribosystem $t_l=6.6\cdot10^{-5}-59.4\cdot10^{-5}$ s.

2. The rheological model was analyzed as a solution to the differential equation. It was shown that a change in operational factors significantly affects the results of the simulation. For example, an increase in sliding rate decreases the time of stress relaxation in the surface structure. This is due to the destruction of aggregates in the structure of the gel and the appearance of rotational movements of separate units – flocs. An increase in the load on a tribosystem significantly increases the value of relaxation time. This is due to squeezing the viscous component out of the structure of a surface film. It was shown that the concentration of fullerenes in the base lubricant does not have a significant effect on the magnitude of relaxation time in the gel structure.

3. The criterion for the transition of elastic properties into viscous ones in the structures of a lubricant film on the friction surface containing fullerene compositions was substantiated. This is the ratio of time of stress action on actual contact spots to stress relaxation time in the film structure. This dimensionless magnitude characterizes the transition of elastic properties of a lubricant film into viscous ones and vice versa, viscous properties into elastic. If this magnitude is smaller than unity, the structure of a film behaves like a solid. Conversely, if it is more than unity, the structure of a film behaves like a viscous medium.

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