

**MONOGRAPH**

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Sergii Boichenko, Olexander Aksionov,  
Petro Topilnytskyi, Andrii Pushak, Kazimierz Lejda

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**SELECTED ASPECTS  
OF PROVIDING THE CHEMMOTOLOGICAL  
RELIABILITY  
OF THE ENGINEERING**

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**MINISTRY OF EDUCATION AND SCIENCE OF UKRAINE  
NATIONAL AVIATION UNIVERSITY**

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**Monograph**

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*The Monograph selectively represents the Aspects of providing the Chemmotological Reliability of the Transport through the investigation of the Quality Parameters of Fuel and Lubricants. Is devoted to the Modern Problems of of Rational use of Traditional and Alternative Fuels, Lubricants and other operational materials during exploitation of Transport.*

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## BASIC ABBREVIATIONS

PC – ProfiLab software	DERD – Directorate of Engine Research and Development
ETC – electron transport chain	KMKO – Complex of Methods for Qualitative Assessment
DM – decision makers	GDP – gross domestic product
ICAO – International Civil Aviation Organization	EGR – exhaust gas recirculation
CAAF – Conference on the Use of Alternative Fuels	ECH – engine cylinder head
MEA – more electric planes	EG – exhaust gas
ODS – Oil dispersed systems	PM – particles mater
ARPD – asphalt-resin-paraffin deposits	ER – emission reduction
PPVO – product of processing of vegetable oils	ID – indicator diagram
PA – polycyclic arenes	CPG – cylinder-piston group
MAC – maximum allowable concentrations	OBD – On-Board Diagnostic
TWUO – tar from West-Ukrainian oil	GPS – Global Positioning System
EPC – European Pellet Council	EASA – European Aviation Safety Agency
EIPS – European Industry of Pellet Suppliers	CAA – Civil Aviation Authority
IBTC – International Biomass Torrefaction Council	FAME – fatty acid methyl esters
CIR – coumarone-indene resin	MIC – Microbial Induced Corrosion
AER – air excess ratio	TCA – tricarboxylic acid
DTA – differential-thermal analysis	IPCC – Intergovernmental panel on Climate Change
IBP – initial boiling point	BFFM2 – Boeing Fuel Flow Method 2
EBP – end boiling point	EE – engine emission
WPhF – wide phenolic fraction	EAPA – European Asphalt Pavement Association
EMF – electromagnetic field	PMB – polymer modified bitumen
MF – magnetic field	SBS – styrene-butadiene-styrene
L – lubricants	SIS – styrene-isoprene-styrene
IATA – International Air Transport Association	SEBS – styrene-ethylene / butylene-styrene
CamO – camellina oils	TE – Thermoplastic elastomers
JF – jet fuel	EPL – Environmental Protection Law
FAEE – fatty acids ethyl esters	OOU – obszar ograniczonego użytkowania (in Polish), noise zone (in English)
FIP – flash point	
ASTM – American Society for Testing and Materials	



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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.



Nowadays, 20–25 % of the world's produced energy is consumed / absorbed, under the functioning of any mechanical systems related to the friction. This leads to the formation of the important tribological tasks, which should improve the economic and environmental efficiency, durability and reliability of the machinery, mechanisms and technological equipment, which are used for various purposes. In addition, nowadays, the modern world has the fast-track developing nanotechnology related to obtaining of new materials with the unique, physico-chemical properties, micro-optoelectronic and microelectromechanical systems, which are characterized by the new friction and wear conditions, applied in the contacting microspheres sector [1–3]. This formed another developing tendency of tribology, specifically, nanotribology (G. Newbauer, 1990), which studies bodies friction of micro- and nanoscale. Thus, in order to study the friction of the thresholding layers with the width from 0.04  $\mu\text{m}$  to 0.08  $\mu\text{m}$  (critical width) [4], in our opinion, first of all, it is necessary to take into consideration an atomic and molecular, and adhesive / chemisorption interaction of the friction surfaces, which forms the derivation of the highly structured, liquid-crystal layers, which have high, wear-resistant properties and a low friction coefficient. Thus, in this case, under the investigation of the friction and wear, first of all, it is necessary to take into account the adhesion / chemisorption, atomic and molecular transformations, which occur on the friction surfaces, and only then, the friction pair volume properties variation. To prove this thesis, there is the discovery of the selective transfer phenomenon (Garkunov D. M., Kragelsky I. V., 1956), which is characterized by the formation of a nano-sized servo-film in the friction pair «bronze – glycerol – steel» [5–6]. This can be applied as fundamentals for considering the selective transfer phenomenon from the outlook of nanotribology, taking into account the chemisorption phenomena and chemical transformations, which occur on the surface of the contacting friction pairs. Thus, in the study [7] the authors explain the selective transfer mechanism by a series of consecutive probable triboelectrochemical mechanoactivation reactions. The authors explain the formation on the friction surface of the servovitic film by the work of macroscopic galvanic cells, in which the surface of steel can be represented as anode, and bronze – as cathode. The reaction also proceeds on the cathodic and anodic surfaces of micro-galvanic cells. In our opinion, it is given more accurate consideration of the chemical transformations in the tribochemical system «bronze – organic additive / medium – steel» from the point of view of the direct mechanically activated dissolution of the more active metal in the donor-acceptor electron transport chain (ETC) system, which is a matrix for the formation of the corresponding copper(II) coordination complexes [8]. According to this thesis, this study is carried out concerning the investigation of the possibility of the direct synthesis of copper(II) coordination complexes with thioamide ligands in the tribochemical system «bronze BrAZ 9-4 – thioamide – organic solvent – oil I-20A – steel 45» and their influence on tribotechnical characteristics of the lubricating compositions.

Methods of the lubricating compositions preparation and investigation of their tribotechnical characteristics are given below.

*Preparation of lubricating compositions 1–5.* 3 ml of an organic solvent (except composition 3) was added to 97 ml of industrial oil I-20A, and the mixtures were thoroughly mixed, using a magnetic stirrer until homogenization. The mixing process, as a rule, was carried out under the room temperature.

*Preparation of lubricating compositions 7–11.* 0.06 g of thioamide ( $\text{HL}^1 - \text{HL}^5$ ) was added to 99.9 ml of industrial oil I-20A, and it was heated to 70–90  $^\circ\text{C}$ , and mixed until dissolved. The obtained lubricating compositions were cooled and the tribotechnical investigations were carried out.

*Preparation of lubricating compositions 12, 15.* 0.06 g of thioamide ( $\text{HL}^1 - \text{HL}^5$ ) was added to 3 ml of DMF, then it was mixed until the full dissolution of thioamide and added to 97 ml of industrial oil I-20A. If it was necessary, the homogenization of the lubricating composition was



carried out with heating to 70–90 °C, then cooled and the tribotechnical investigations were performed.

*Preparation of lubricating compositions 13, 16.* 0.06 g of coordination compound ( $\text{CuL}_2^1 - \text{CuL}_2^5$ ) was added to 3 ml of DMF, then heated to 70 °C and stirred until the solution was homogenized. The obtained homogeneous mixture was added to 97 ml of industrial oil I-20A that was preheated to 70–80 °C. The mixture was stirred until it was completely homogenized, cooled and the tribotechnical investigations were performed.

*Preparation of the lubricating compositions 14, 17* were carried out similarly to the lubricating compositions 13, 16. In this case, for example, instead of the coordination compound  $\text{CuL}_2^1$ , a mixture of  $\text{HL}^1 + \text{CuL}_2^1$  in a ratio of 1 : 1 was taken.

*Methodology of the investigation.* The lubricating compositions were investigated using a friction machine SMC-2 (Fig. 1) with the friction pairs «shoe-roller», sliding velocity was 3.0 m/s and friction displacement –  $3 \cdot 10^3$  m. Material of the roller was steel 45, shoe material – bronze BrAZ 9-4. The initial roughness was from 0.30  $\mu\text{m}$  to 0.62  $\mu\text{m}$  applied for a steel sample and from 0.62  $\mu\text{m}$  to 0.80  $\mu\text{m}$  for a bronze sample [9]. The duration of testing of one composition was 40 minutes. The temperature change in the friction zone was determined by the chromel copel thermocouple and marked on the corresponding strip chart curve of the electronic potentiometer KSP-4. The frictional force was determined with the means of a strain gauge sensor. The wear of the sample was indicated by a weighting method, which was applied on the analytical balance of the second accuracy classe VLR-200 type (GOST 24104–80). The change of mass of the samples was determined:

$$\Delta m = m_n - m_k \quad (1)$$

where  $m_n$  – initial mass of the sample, g;  $m_k$  – mass of the sample at the end of the experiment, g.

From the friction torque curve in a strip chart was taken the value of  $l$  in mm, corresponding to the deviation of the movable carriage of the KSP-4 potentiometer at the beginning and at the end of the experiment (the mode of stable selective transfer in the friction couple). With the calibration chart, it was determined the corresponding friction torque  $M_{fr}$  by the  $l$  (mm) deviation magnitude of the potentiometer's carriage. The friction coefficient was determined by the formula:

$$f = \frac{M_{fr}}{r \cdot N} \quad (2)$$

where  $M_{fr}$  – friction torque in the investigated friction pair, N·m;  $r$  – radius of a movable roller, m;  $N$  – total load in the friction pair, N.

The installation schematic diagram of the investigated tribotechnical characteristics of the lubricating compositions in a friction pair «bronze– steel» is given in Fig. 1.

*Investigation of the tribotechnical system «bronze BrAZ 9-4 – organic solvent – oil I-20A – steel 45» [11].* It is known [12, 13] that in the friction pair «bronze-steel», the friction mechanical effect leads not only to the wear of its surface, but also due to the presence of complexing reagents in the oil medium, the friction mechanical effect leads to the formation of the metal-complex compounds, which are different in the composition and structure. Initially, this phenomenon was observed in the friction pair «copper alloy – steel» in glycerine, which was oxidized as a ligand, and formed the multiplex metal-complex compounds [14, 15].



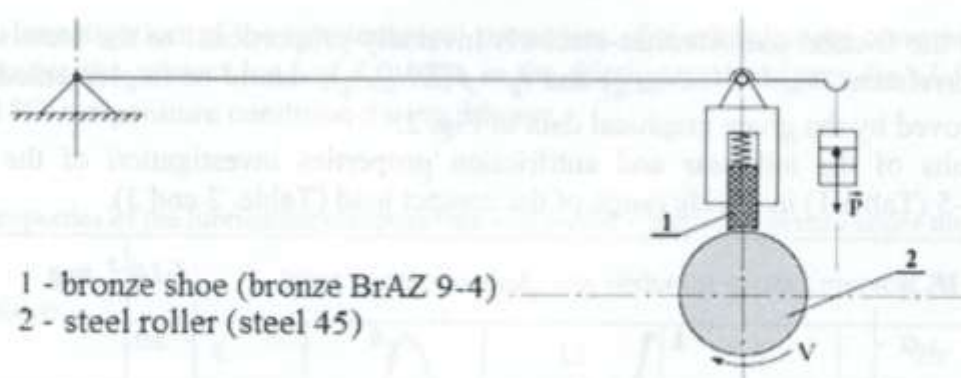


Fig. 1. The installation schematic diagram of the investigated tribotechnical characteristics of the friction pair bronze BrAZ 9-4 – steel 45, using a friction machine of SMC-2 type

The next persistent investigations of this friction pair, under the presence of N-, O-containing organic ligands, were proved the formation of copper(II) coordination complexes, the presence of which, substantially improved the tribotechnical properties of the lubricating compositions [12, 16, 17]. Specifically, the wear ( $I_g$ ) and the friction coefficient ( $f_f$ ) decrease was unambiguously associated with the oxidation of metals during the friction in the oil medium and with the formation of their coordination compounds [18].

A sufficient number of scientific surveys is devoted to the problem of the metals oxidation in the aprotic solvents. In the studies [21–23] it was frequently considered that the metals oxidative solubility, especially, the direct organometallic and coordination complexes synthesis, is determined both by the nature of the metal (the ionization potential  $U$ , the standard redox potential  $E^0$ , the resonant potential  $I_r$ ), and by the nature of the organic aprotic solvent (dipole moment  $\mu$ , dielectric permittivity  $\epsilon^{2\theta}$ , donor number  $DN_{SbCl_5}$ ). In the researches [23–26] it is stated that metals oxidation rate  $W$  depends on the donor numbers  $DN_{SbCl_5}$  of the organic solvents and has an extreme character, thus,  $W = f(DN_{SbCl_5})$  correlation can be applied. This can be explained by the selective adsorption on the metallic surfaces, which have the diverse nature. Thus, Kuprin V. P. [27, 28] demonstrated the correlation of copper and bronze adsorption of the organic substances / solvents and their resonant potential ( $I_r$ ), which also has the similar extreme character. However, it can be assumed, that there is a direct correlation between the organic solvents donor force ( $DN_{SbCl_5}$ ) and their resonant potentials ( $I_r$ ), if there is an adsorption of organic substances / solvents on the metal surface, the metals oxidation of this surface, where  $M^{n+}$  cations are formed, and the probability of their complexation in the solution is maximum (triple conditional «resonance») then, as a consequence, the friction pair has the minimum values of the wear ( $I_g$ ) and friction coefficient ( $f_f$ ), in the case of using such tribotechnical systems. An example of this assumption is the analysis and comparison of the different authors' investigation results, which are given in Fig. 2. The curve I represents the extreme correlation of the cadmium oxidation rate ( $I_r = 7.4$  eV [25]) and diphenylbismuthchloride, under involving of the organic aprotic solvents, and of their donor numbers [24]. It was found that the maximum dissolution rate ( $W_{rel}$ ) of cadmium is observed in solvents with the highest donor activity: DMSO and DMF. The II curve shows the correlation between the obtained wear ( $I_g$ ) of the friction pair «bronze BrAZ 9-4 – steel 45» of the lubricating composition «oil I-20 A + organic solvent», and the donor numbers of the investigated organic solvents.

According to the study [27], the resonant potential is equal to  $I_r = 7.2$  eV for zero-valent copper and for its oxides CuO and  $Cu_2O$   $I_r = 7.9$  eV, and for bronze with the different composition, for example, with the tin composition,  $I_r^1 = 7.25$  eV and  $I_r^2 = 8.00$  eV. If we assume that with the same donor force of the investigated organic solvents (curves I and II, Fig. 2) and approximately the same values of the resonant potentials for cadmium and copper, their dissolution rate coincides,



and the wear in the friction pair «bronze-steel» is inversely proportional to the metals dissolution rate, then the correlation  $W_{rel} = f(DN_{SbCl_5})$  and  $I_g = f(DN_{SbCl_5})$ , should be implemented, i.e.,  $W_{rel} = 1/I_g$ , which is proved by the given graphical data in Fig. 2.

The results of the antiwear and antifricition 'properties investigation of the lubricating compositions 1-5 (Table 1) in a wide range of the contact load (Table. 2 and 3).

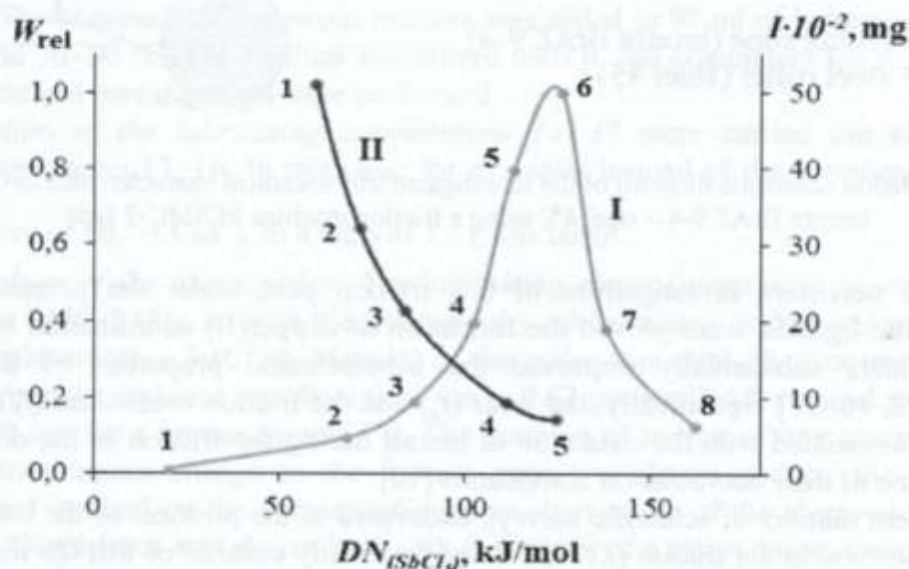


Fig. 2. The correlation of the relative dissolution rate between cadmium and bismuth diphenyl chloride and the donor numbers of the used organic solvents (I, conditions:  $t = 20\text{ }^{\circ}\text{C}$ ,  $C_{ox} = 0,4\text{ mol/l}$ ): 1 - *p*-xylene; 2 - ethyl acetate; 3 - tetrahydrofuran; 4 - diglyme; 5 - DMF; 6 - DMSO; 7 - pyridine; 8 - hexamethylphosphoramide and the correlation of the wear in the friction pair «bronze BrAZ 9-4 - steel 45» with the lubricating composition «oil I-20 A + organic solvent» and donor numbers of the used organic solvents (II, conditions:  $t = 20\text{ }^{\circ}\text{C}$ ,  $\tau = 3\text{ h}$ ,  $P = 8\text{ MPa}$ ): 1 - dipropyl oxalate; 2 - ethyl acetate; 3 - TCM : DMF = 1 : 1; 4 - DMF; 5 - DMSO

Table 1

The compounds of the lubricating compositions «oil I-20A + organic solvent» and their physical, antiwear and antifricition properties

Composition	Compounds of the composition			Physical properties [29]			Tribotechnical properties	
	Organic solvent		Base oil I-20A	$\mu_s$ $C \cdot m \cdot 10^{-30}$	$e^{20}$	$DN_{SbCl_5}$ , kJ/mol	$I_g$ , mg	$f_{fr}$
	Title	% mass						
1	dipropyl oxalate	3,0	до 100	-	-	60,00	0,5106	0,32
2	ethyl acetate	3,0	до 100	6,03	18,51	71,57	0,3215	0,23
3	TCM	1,5	до 100	-	-	84,00	0,2128	0,19
	DMF	1,5						
4	DMF	3,0	до 100	12,70	36,7	111,33	0,0928	0,12
5	DMSO	3,0	до 100	13,03	48,9	124,73	0,0733	0,10
6	I-20A	100	-	-	-	-	0,6004	4,24

Note. Investigation of the tribotechnical properties of the lubricating compositions 1–5 was carried out under the contact load of 8.0 MPa in the friction pair «bronze BrAZ 9-4 – steel 45» under the 20 °C temperature condition during 3 hours.

Table 2

Antiwear properties of the lubricating compositions «oil I-20A + organic solvent» under the various loads

Composition	Wear of the sample $I_p$ , mg, under the contact pressure $P$ , MPa					
	4	8	12	16	20	24
1	0,4134	0,5106	0,5747	0,6933	0,8892	–
2	0,2200	0,2856	0,3805	0,4301	0,5408	0,6647
3	0,1402	0,2008	0,2567	0,3200	0,4001	0,5066
4	0,0433	0,0928	0,1867	0,2534	0,3332	0,4607
5	0,0267	0,0733	0,1005	0,1800	0,2267	0,2940
6	0,6003	0,6004	0,7406	0,9002	–	–

Table 3

Antifriction properties of the lubricating compositions «oil I-20A + organic solvent» under the various loads

Composition	The friction coefficient $f_f$ under the contact pressure $P$ , MPa					
	4	8	12	16	20	24
1	0,41	0,32	0,33	0,51	0,96	1,41
2	0,37	0,23	0,19	0,27	0,48	0,80
3	0,34	0,19	0,15	0,21	0,40	0,63
4	0,29	0,12	0,06	0,08	0,19	0,41
5	0,27	0,10	0,03	0,05	0,15	0,32
6	3,76	4,24	4,85	5,20	5,15	5,64

The results of the investigation (Table 2, 3) are represented graphically in Fig. 3. The data shown in Fig. 3a represent that in the range of contact load from 4 MPa to 24 MPa, the lubricating compositions, which contain organic solvents with high value of donor numbers  $DN_{SbCl_5}$  (compositions 4 and 5), have the best antiwear properties in the friction pair «bronze BrAZ 9-4 – steel 45», which unequivocally indicates the determining donor activity character of the organic aprotic solvents under the oxidation of metals. The determining influence on the wear decrease of the donor activity of organic solvents in the compounds of the lubricating compositions remains unchanged irrespectively to the contact load (4–24 MPa) in the investigated friction pair. It should be noted that the additional DMF dilution with 50 % of tetrachloromethane ( $DN_{SbCl_5} = 0$ ) reduces the overall value of the donor activity ( $DN_{SbCl_5} = 84,4$ ; the value is obtained by the graphic data interpolation (Fig. 2) and increases the wear in 1.6 times compared to «pure» DMF, which again proves the important character of the aprotic solvents and their donor activity during the metals oxidation.



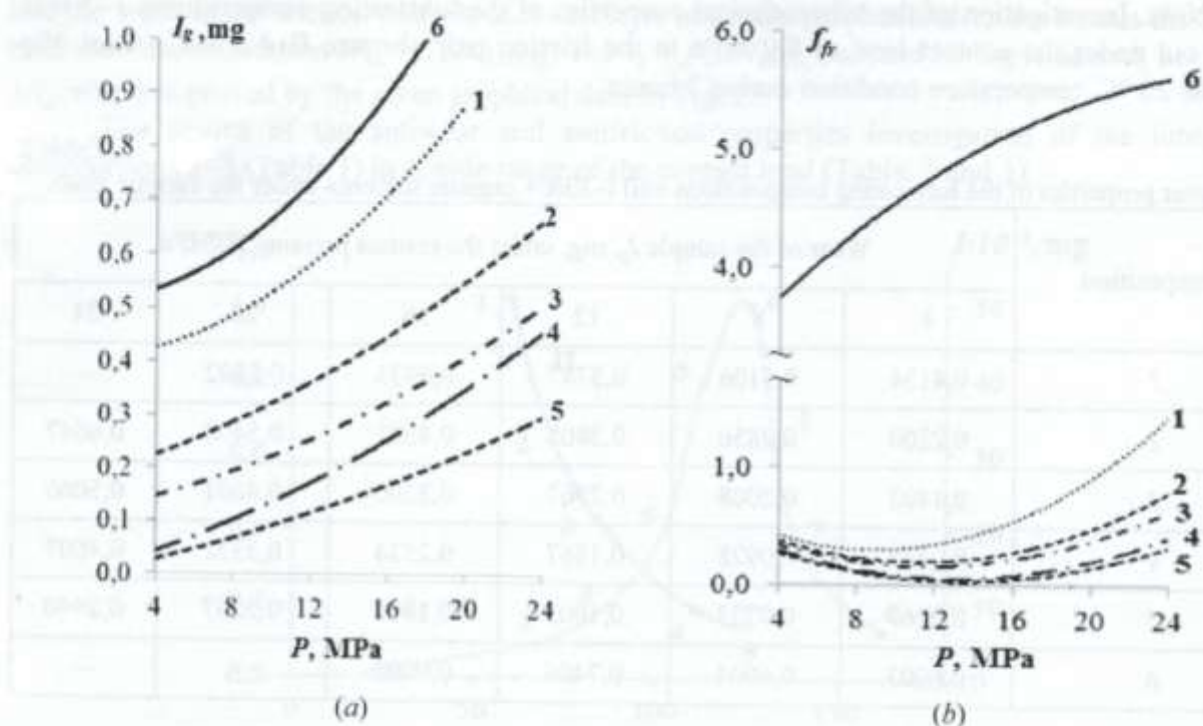
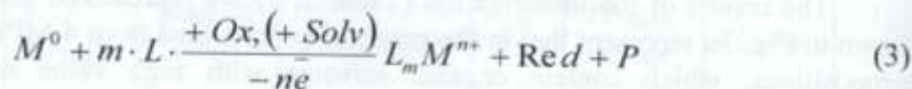


Fig. 3. The correlation between the wear (a) and the friction coefficient (b) under the contact load in the friction pair «bronze BrAZ 9-4 – steel 45» with the lubricating composition «oil I-20A + organic solvent» (conditions:  $t = 20\text{ }^{\circ}\text{C}$ ,  $\tau = 3\text{ h}$ ): 1 – dipropyl oxalate, 2 – ethyl acetate, 3 – TCM : DMF = 1 : 1; 4 – DMF; 5 – DMSO; 6 – «pure» oil I-20A

It was found out a similar correlation between the decreasing of the friction coefficient under the growth of the donor activity of the organic aprotic solvents within the pressure range from 4 MPa to 24 MPa for the investigated lubricating compositions (Fig. 3b). It should be considered a significantly less range of the friction coefficient change in the definite range of contact pressure. It is obviously, that it is revealed due to another mechanism of the antifriction action of the compounds of the compositions 1–5 in comparison to the mechanism of antiwear action of the same solvents.

*Investigation of the tribotechnical system «bronze BrAZ 9-4 – thioamide – oil I-20 A – steel 45» [8, 30].* In addition to the nature of aprotic solvents (Solv), the metals dissolution rate is greatly influenced by the presence of a complexing agent in the investigated systems:



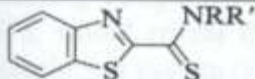
where  $M^0$  – zero-valent metal;  $L$  – complexing agent;  $Ox$  – oxidant;  $L_m M^{n+}$  – formed coordination complex;  $Red$  – reduced oxidant form;  $P$  – by-products of the chemical interaction.

In our case, different substituted thioamides have been investigated as complexing agents. The thioamides, as additives to industrial oils, have been studied in detail in the following surveys [19, 31–36], however, the improvement of the tribotechnical properties of the friction pair «bronze-steel» was previously related only to the implementation of copper (II) selective transfer from the bronze surface to the steel surface without taking into consideration the character of the organic solvents in this process. In the studies [9, 19] there were investigated the lubricating compositions, which are based on the industrial oil I-20A, complexes of the mixed ligand thioamides and DMF, as an aprotic solvent. However, DMF was used to homogenize the lubricating composition and to

improve the copper(II) metal-chelates dissolution, and not as an active component of additional dissolution of zero-valent copper in the friction pair «bronze – steel». In the context of these studies tasks, which are related to the direct synthesis of copper(II) thioamide complexes, we conducted an additional investigation of the effect of the aprotic solvents on the tribotechnical properties of the industrial oil I-20A. At first, for this purpose, the system «bronze BrAZ 9-4 – thioamide – oil I-20 A – steel 45» was investigated. In this case, the obtained data (Table. 4) and their corresponding graphical correlation is represented in Fig. 4. The investigated thioamides HL<sup>1</sup> – HL<sup>5</sup>, containing benzothiazole and amine –NRR' fragments, as additives to the oil I-20A, in various ways influence on the antiwear properties of the investigated lubricating compositions 7–11. We have determined that the obtained data in the range of the 8–16 MPa contact pressure are the most important and informative.

Table 4

Compounds of the lubricating compositions «oil I-20 A + thioamide» and their antiwear and antifricition properties

Composition				Compounds of the composition, %			Wear of the sample $I_x \cdot 10^{-1}$ , mg					Friction coefficient $f_f \cdot 10^{-2}$				
	R	R'	design ation	thioam ide	DMF	oil I-20A	contact pressure $P$ , MPa									
							8	12	16	20	24	8	12	16	20	24
7	H	C <sub>6</sub> H <sub>5</sub>	HL <sup>1</sup>	0,06	–	до 100	0,43	0,88	1,33	4,95	8,50	2,3	2,4	2,5	2,9	3,2
8	CH <sub>3</sub>	CH <sub>3</sub>	HL <sup>2</sup>	0,06	–	до 100	0,33	0,50	0,67	3,50	6,00*	2,5	2,6	2,7	5,1	8,0
9	(CH <sub>2</sub> CH <sub>2</sub> ) <sub>2</sub> O		HL <sup>3</sup>	0,06	–	до 100	0,00	1,30	2,67	4,31	5,87	3,0	3,1	3,3	5,2	7,3
10	H	C <sub>6</sub> H <sub>4</sub> Cl-4	HL <sup>4</sup>	0,06	–	до 100	0,00	1,28	2,50	3,88	5,25	3,3	5,0	6,7	6,6	6,4
11	H	C <sub>6</sub> H <sub>4</sub> Br-4	HL <sup>5</sup>	0,06	–	до 100	0,00	0,93	1,80	2,35	3,00	5,7	5,8	5,9	5,7	5,5

Note: \* – the value was obtained under the 22 MPa contact pressure.

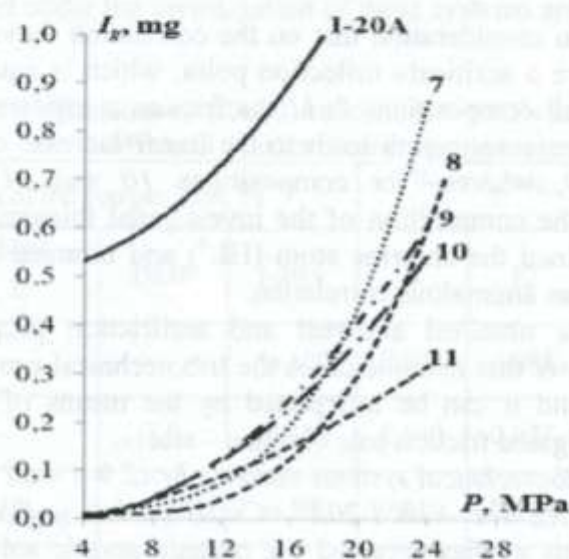


Fig. 4. The correlation between the wear and the contact pressure in the friction pair «bronze BrAZ 9-4 – steel 45» with the lubricating composition «oil I-20A + thioamide» ( $t = 20$  °C,  $\tau = 3$  h)

It should be highlighted that thioamides with *N*-aryl fragment (HL<sup>1</sup>, HL<sup>4</sup>, HL<sup>5</sup>) from 8 MPa to 16 MPa contact pressure range have the best results, while under the increasing of the contact pressure to 24 MPa, the antiwear activity series changes into this:

$$HL^5 > HL^4 > HL^3 > HL^2 > HL^1,$$



that at the first approximation can be explained by the thermicpermanence increase of the investigated heterocyclic thioamides. Thus, in the heat-resistant correlation, in the given series, *N,N*-dimethylamidobenzothiazole-2-thiocarboxylic acid (HL<sup>2</sup>) is the least stable compound and is the latest in the given antiwear activity.

Fig. 5 illustrates the graphical correlation between the antifriction properties of the investigated lubricating compositions 7–11 (Table 4) and the contact load.

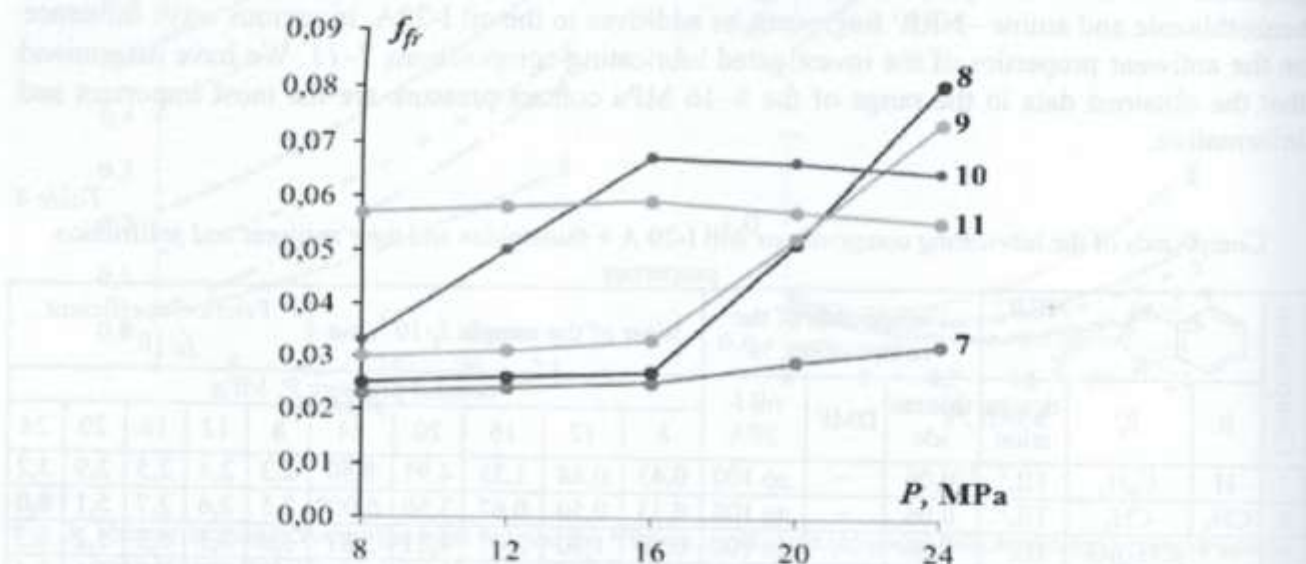


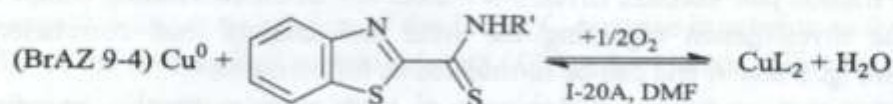
Fig. 5. The correlation between the friction coefficient and the contact pressure in the friction pair «bronze BrAZ 9-4 – steel 45» with the lubricating composition «oil I-20A + thioamide» ( $t = 20^\circ\text{C}$ ,  $\tau = 3\text{ h}$ )

It should be taken into consideration that on the correlation curves  $f_f - P$  the investigated lubricating compositions have a «critical» inflection point, which is equal to 16 MPa. Under the growth of contact load, for all compositions 7–11 the friction coefficient linearly increases up to 16 MPa. A further contact pressure growth leads to the linear increase of the friction coefficients only for compositions 7–9, whereas for compositions 10 and 11 the friction coefficient unexpectedly decreases. In the composition of the investigated thioamides, which revealed such properties, there were contained the chlorine atom (HL<sup>4</sup>) and bromine atom (HL<sup>5</sup>) in the *N*-aryl fragment, that explains such an anomalous correlation.

A comparison of the obtained antiwear and antifriction properties (Table 4) of the lubricating compositions shows that in some cases the tribotechnical properties improvement does not occur simultaneously, and it can be interpreted by the means of different mechanisms of additives action in the investigated friction pair «bronze – steel».

*Investigation of the tribotechnical systems «bronze BrAZ 9-4 – oil I-20A – thioamide – DMF – steel 45» and «bronze BrAZ 9-4 – oil I-20A – Cu(II) chelate – DMF – steel 45».* Previous, simpler tribotechnical systems studies revealed the organic aprotic solvents effect on the direct dissolution of zero-valent copper, which is contained in bronze BrAZ 9-4, and impact on the improvement of the antiwear and antifriction properties of the lubricating compositions, based on industrial oils. During the investigation of the same friction pair and aprotic solvent replacement with heterocyclic thioamides of the different substitutions, which included the benzothiazole fragment, their high efficiency was demonstrated, concerning the improvement of the antiwear and antifriction properties of the «bronze – steel» friction pair. According to Kuzharov A. S. [12] and our previous studies, this can be explained due to the fact, that in the friction process, coordination complexes of general formula  $\text{CuL}_2$  are formed. At the same time oxygen is the oxidant, and zero-valent copper is the reductant, which is contained in bronze:





However, the composition of the previously studied tribotechnical system «bronze BRAZ 9-4 – thioamide – oil I-20A – steel 45» (tables 4 and Fig. 4, 5) did not include organic aprotic solvents with high donor activity, for instance, DMF ( $DN_{\text{SbCl}_5} = 111.33 \text{ kJ} \cdot \text{mol}^{-1}$ ), which significantly increase the metallic copper dissolution rate, and the formation of  $\text{CuL}_2$  metal chelates, and, accordingly, improve the tribotechnical properties.

In order to eliminate this disadvantage, we conducted investigations of such tribotechnical systems:

– «bronze BRAZ 9-4 – oil I-20A – thioamide ( $\text{HL}^1, \text{HL}^5$ ) – DMF – steel 45». It was assumed that a comparison of the obtained results concerning the antiwear properties of the lubricating compositions, which contain active additives ( $\text{HL}^1, \text{HL}^5$ ), ( $\text{HL}^1 + \text{DMF}$ ) and ( $\text{HL}^5 + \text{DMF}$ ), would reveal the character of the aprotic DMF solvent, as an active component of the tribotechnical system (additional factor), with the direct copper dissolution in the friction process;

– «bronze BRAZ 9-4 – oil I-20A – chelate ( $\text{CuL} \frac{1}{2}, \text{CuL} \frac{5}{2}$ ) – DMF – steel 45». It was assumed that in the case of similar results, concerning the antiwear properties, obtained by the investigation of the given systems: ( $\text{HL}^1 + \text{DMF}$ ) and ( $\text{CuL} \frac{1}{2} + \text{DMF}$ ), ( $\text{HL}^5 + \text{DMF}$ ) and ( $\text{CuL} \frac{5}{2} + \text{DMF}$ ), it is possible not only indirectly to prove the formation of metal chelates  $\text{CuL} \frac{1}{2}, \text{CuL} \frac{5}{2}$ , but also to reveal the active character of the aprotic solvents, because such investigations have not been carried out previously.

The results, obtained under the investigation of these systems are given in Table 5.

Table 5  
Compounds of the lubricating compositions «I-20A oil + thioamide + DMF» and «I-20A oil + chelate Cu(II) + DMF» and their antiwear properties

Composition	The compounds of the composition, %				Wear $I_g$ , mg, under the contact pressure $P$ , MPa				
	thioamide HL	complex $\text{CuL} \frac{n}{2}$	DMF	I-20A	8	12	16	20	24
7	$\text{HL}^1, 0,06$	–	–	до 100	0,043	0,088	0,133	0,495	0,850
12	$\text{HL}^1, 0,06$	–	3,0	до 100	0,040	0,027	0,030	0,038	0,039
13	–	$\text{CuL} \frac{1}{2}, 0,06$	3,0	до 100	0,010	0,014	0,018	0,027	0,013
14	$\text{HL}^1, 0,03$	$\text{CuL} \frac{1}{2}, 0,03$	3,0	до 100	0,024	0,034	0,026	0,041	0,034
11	$\text{HL}^5, 0,06$	–	–	до 100	0,000	0,093	0,180	0,235	0,300
15	$\text{HL}^5, 0,06$	–	3,0	до 100	0,035	0,022	0,011	0,032	0,030
16	–	$\text{CuL} \frac{5}{2}, 0,06$	3,0	до 100	0,000	0,011	0,016	0,025	0,021
17	$\text{HL}^5, 0,03$	$\text{CuL} \frac{5}{2}, 0,03$	3,0	до 100	0,000	0,019	0,034	0,040	0,033



In the friction pair «bronze BrAZ 9-4 – steel 45» of the lubricating compositions 12–17, the results of the investigation regarding the wear and contact load correlation (Table 5) and represented in Fig. 6 and 7, and can be formulated as following:

– under the condition of the contact load change, in the investigated lubricating compositions, in the friction pair «bronze BrAZ 9-4 – steel 45», the wear magnitude variation is represented as a constrained values range  $I_g$ : for the lubricating compositions 12–14 (Fig. 6) the magnitude variation is 0.013–0.041 mg, and for compositions 15–17 (Fig. 7) the magnitude variation is 0.011–0.04 mg;

– in the friction pair «bronze BrAZ 9-4 – steel 45», given intervals of the wear magnitude variation are practically identical, thus, as a sequence, we obtained the same antiwear effect mechanism of the investigated additives and lubricating compositions (Table 5);

– similar  $I_g$  values of the investigated compositions 12–17, for example, under the condition when  $P = 16$  MPa demonstrate, that the active substances, which provide the antiwear properties of the investigated friction pair, are absolutely  $\text{CuL}_2^1$  and  $\text{CuL}_2^5$  coordination compounds, which are present in the I-20A industrial oil;

– the difference in  $I_g$  values is 4.4 times for compositions 7 ( $\text{HL}^1$ ) and 12 ( $\text{HL}^1 + \text{DMF}$ ) (Table. 5, Fig. 6), for instance, under the condition when  $P_c = 16$  MPa, and for compositions 11 ( $\text{HL}^5$ ) and 15 ( $\text{HL}^5 + \text{DMF}$ ), Table. 5, Fig. 7, under the same contact pressure, the difference in  $I_g$  values is 16.4 times, and it can be considered as a very significant aprotic DMF solvent effect regarding the process of zero-valent copper dissolution according to the previously given scheme (4)

It was demonstrated the series of the antiwear and antifriction action of the investigated organic solvents under the  $P = 8$  MPa contact load.

$f_f$	DMSO	>	DMF	>	(DMF + $\text{CCl}_4$ )	>	$\text{CH}_3\text{C}(=\text{O})\text{OC}_2\text{H}_5$	>	$(\text{C}(=\text{O})\text{OC}_3\text{H}_7)_2$
$I_g, \text{mg}$	DMSO	>	DMF	>	(DMF + $\text{CCl}_4$ )	>	$\text{CH}_3\text{C}(=\text{O})\text{OC}_2\text{H}_5$	>	$(\text{C}(=\text{O})\text{OC}_3\text{H}_7)_2$
$DN_{\text{SbCl}_5}$ kJ/mol	124,73		111,33		84,00		71,57		60,00

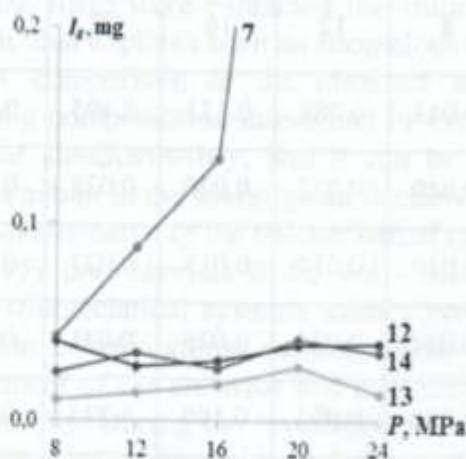


Fig. 6. The correlation between the wear and contact load in the friction pair «bronze BrAZ 9-4 – steel 45» with the lubricating compositions «oil I-20A + thioamide  $\text{HL}^1 + \text{DMF}$ » and «oil I-20A + chelate  $\text{CuL}_2^1$ » ( $t = 20^\circ\text{C}$ ,  $\tau = 3$  h)

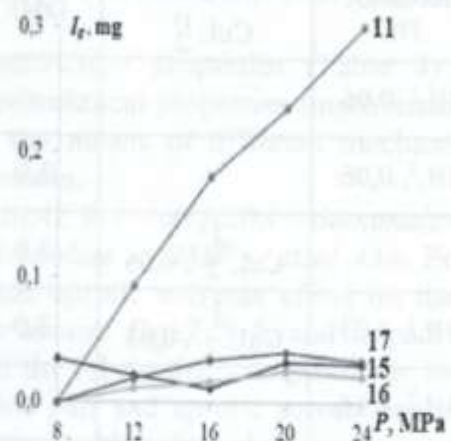


Fig. 7. The correlation between the wear and contact load in the friction pair «bronze BrAZ 9-4 – steel 45» with the lubricating compositions «oil I-20A + thioamide  $\text{HL}^5 + \text{DMF}$ » and «oil I-20A + chelate  $\text{CuL}_2^5$ » ( $t = 20^\circ\text{C}$ ,  $\tau = 3$  h)



These data specify a linear correlation of the  $I_g$  and  $f_{fr}$  decrease in relation to the increasing of the donor activity of the investigated aprotic solvents ( $DN_{SbCl_5}$ ) and their ability to, additionally, increase the dissolution of zero-valent copper in the friction pair «bronze – steel» with the formation of the corresponding coordination compounds. It was found that the serial complication of the lubricating compositions 1–5, 7–17 by the addition of the organic solvents, thioamide complexones ( $HL^1 - HL^5$ ), copper(II) metal chelates, based on them ( $CuL_2^1, CuL_2^5$ ), to oil I-20A allowed not only to find out a number of antiwear activity of additives, for example, for the aprotic solvent DMF:

Additives to the oil I-20A	I-20A	>	DMF	>	HL <sup>1</sup>	>	HL <sup>1</sup> +DMF	>	CuL <sub>2</sub> <sup>1</sup> + DMF
$I_g, mg$	0,6004		0,0928		0,0430		0,0400		0,0100

but also to investigate the influence of each of the factors on the tribotechnical characteristics in the friction pair «bronze BrAZ 9-4 – steel 45».

Thus, the conducted investigations allow us to draw the following conclusions:

1. Organic solvents, which are included in the lubricating compositions «oil I-20A + organic solvent» in 1,2–8,2 and 13,3–42,4 times improve, respectively, the antiwear and antifriction properties of the friction pair «bronze BrAZ 9-4 – steel 45» in comparison with «pure» oil I-20A.

2. Antiwear properties of the lubricating compositions «oil I-20A + organic solvent» in the friction pair «bronze BrAZ 9-4 – steel 45» are determined by the donor activity ( $DN_{SbCl_5}$ ) of the organic solvents:  $I_g = f(DN_{SbCl_5})$ . The minimum wear values are observed during there were used organic solvents (DMF, DMSO), which have the maximum values of  $DN_{SbCl_5}$ .

3. The injection of the organic solvent (DMF) into the lubricating composition «oil I-20A + thioamide HL<sup>1</sup>» provides the wear decrease in the «bronze BrAZ 9-4 – steel 45» friction pair in 4.4 times, under the contact pressure of 16 MPa. Thus, the organic solvent (DMF) is an active dissolution zero-valent copper component, which is contained in bronze.

4. Similar values of the wear in the friction pair «bronze BRAZ 9-4 – steel 45», within the presence of the lubricating compositions «oil I-20A + thioamide HL<sup>1</sup> + DMF», «oil I-20A + chelate  $CuL_2^1 + DMF$ », «oil I-20A + thioamide HL<sup>1</sup> + chelate  $CuL_2^1 + DMF$ » (according to 0.030, 0.018 and 0.026 mg under the contact pressure of 16 MPa) prove that in all cases the active antiwear additive to oil I-20A is the coordination compound  $CuL_2^1$ .

5. Coordination compounds of the general formula  $CuL_2$  are the effective antiwear and antifriction additives to industrial oils and can be recommended for industrial use and can be applied for the general friction pairs «bronze – steel».

#### РЕФЕРАТ

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#### ФОРМУВАННЯ КООРДИНАЦІЙНИХ СПЛУК МІДІ (II) ПІД ЧАС ПРОЦЕСУ ТЕРТЯ ТА ЇХ ВПЛИВ НА ТРИБОТЕХНІЧНІ ХАРАКТЕРИСТИКИ ЗМАЩУВАЛЬНИХ РЕЧОВИН

Досліджено трибохімічну систему «бронза БраЖ 9-4 – органічний додаток – олива I-20A – сталь 45», у якій під дією механічного активування металевих поверхонь пари тертя та органічних



додатків утворюються координаційні сполуки купруму (II), що забезпечують високі протизношувальні й антифрикційні властивості мастильних композицій. Послідовне ускладнення мастильних композицій шляхом додавання до базової оливи I-20A органічних розчинників, тіоамідних лігандів та метал-хелатів купруму (II) на їх основі дозволило встановити ряд протизношувальної активності додатків до індустриальної оливи I-20A та дослідити вплив кожного із чинників на триботехнічні характеристики в парі тертя «бронза БрАЖ 9-4 – сталь 45». На основі отриманих результатів зроблено висновок, що покращення експлуатаційних характеристик мастильних композицій визначається утворенням у процесі трибохімічних реакцій координаційних сполук купруму (II).

**Ключові слова:** трибохімія, мастильні композиції, координаційні сполуки купруму(II), протизношувальні і антифрикційні властивості

#### РЕФЕРАТ

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#### **ФОРМИРОВАНИЕ КООРДИНАЦИОННЫХ СОЕДИНЕНИЙ МЕДИ (II) ВО ВРЕМЯ ПРОЦЕССА ТРЕНИЯ И ИХ ВЛИЯНИЕ НА ТРИБОТЕХНИЧЕСКИЕ ХАРАКТЕРИСТИКИ СМАЗЫВАЮЩИХ ВЕЩЕСТВ**

Исследована трибохимическая система «бронза БрАЖ 9-4 – органическая присадка – масло И-20А – сталь 45», в которой под действием механического активирования металлических поверхностей пары трения и органических присадок образуются координационные соединения меди(II), обеспечивающие высокие противозносные и антифрикционные свойства смазочных композиций. Последовательное усложнение смазочных композиций путем добавления к базовому маслу И-20А органических растворителей, тиамидных лигандов и металл-хелатов меди (II) на их основе позволило установить ряд противозносной активности присадок к индустриальному маслу И-20А и исследовать влияние каждого из факторов на триботехнические характеристики в паре трения «бронза БрАЖ 9-4 – сталь 45». На основании полученных результатов сделан вывод, что улучшение эксплуатационных характеристик смазочных композиций определяется образованием в процессе трибохимических реакций координационных соединений меди(II).

**Ключевые слова:** трибохимия, смазочные композиции, координационные соединения меди (II), противозносные и антифрикционные свойства

#### ABSTRACT

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

It has been investigated the tribochemical system «bronze BrAZ 9-4 – organic additive – oil I-20A – steel 45» in which due to mechanical activation of metal particles of the surfaces of friction pair the copper(II) coordination compounds have been created. These complexes provide high antiwear and antifricition properties of the lubricating compositions. The sequential complication of lubricating compositions by adding organic solvents, thioamide ligands and metal chelates of copper(II) based on them to base oil I-20A allowed to establish an order of anti-wear activity of additives to industrial I-20A oil and investigate the influence of each of the factors on tribotechnical characteristics in friction pair «bronze BrAZ 9-4 – steel 45». Based on the obtained results, it is concluded that improvement of the operational characteristics of lubricating compositions is determined by the formation of copper(II) coordination compounds during the tribochemical reactions.

**Key words:** tribochemistry, lubricating compositions, copper (II) coordination compounds, antiwear and antifricition properties.

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### 2.3 DEVELOPMENT OF ALTERNATIVE JET FUELS MODIFIED WITH CAMELINA OIL BIO-ADDITIVES

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Constant increase of aircraft fleet and exhausting crude oil deposits promote worsening of the world energy crisis. As a result we observe rise in prices for jet fuel (JF) that today comprise about 25–30 % of passenger travel [1]. Moreover, products of fuel combustion cause detrimental impact on environment. Thus, the task of search and development of alternative JFs became especially important.

This work is devoted to the study of possibilities of partial replacement of conventional JFs with components of biological (plant) origin. It will allow decreasing the dependence on exhaustible energy sources and minimizing negative impact of aviation on environment.

Today alternative fuels from various renewable feedstock are actively developed and studied. Among them are fuels made of biomass, plant oils, animal fats, microalgae, waste from agriculture, wood processing industry, municipal waste etc [2]. JFs produced from biomass via FT-



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*Scientific edition*

**Sergii BOICHENKO,  
Olexander AKSIONOV,  
Petro TOPILNYTSKYI,  
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Kazimierz LEJDA**

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