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WEAR RESISTANCE IMPROVEMENT OF LINEAR BLOCK-POLYURETHANES UNDER CONDITIONS OF LIQUID FRICTION

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Abstract:
A significant disadvantage of parts made of linear block polyurethanes under conditions of friction contact in the presence of an aqueous medium is the intensification of hydrolysis processes and decrease in volume strength. It was proposed to slow down the hydrolysis of polyurethanes in three ways: by directed changes in chemical structure, by electromagnetic radiation of different intensities and by creation of composite materials, based on the principle of additional intermolecular crosslinking at the manufacturing and hot processing stages. The conditions for the formation of a smooth wear-resistant surface that protects the material from rapid destruction are revealed.

Key words: *linear block-polyurethane, wear resistance, crystallinity, smooth wear-resistant surface, electromagnetic radiation, composite block-polyurethanes*

INTRODUCTION

Polyurethanes are among the most universal and practically used polymers, due to the wide possibilities of changing their chemical structure and properties [1]. They are processed by high-performance injection molding, extrusion, blowing, pneumatic molding, pressing, etc. Absolutely all known types of materials and products can be obtained from polyurethanes: filled, reinforced, foamed, laminated and others in the form of plates, sheets, blocks, profiles, fibers, films, etc. [2]. Recently, polyblock polyurethanes of linear structure are widely used in industry because of their unique combination of properties: high level of strength and elasticity, oil and gas resistance, resistance to shock loads and vibration. Products and constructions based on linear block-polyurethanes (BPU) are used in all industries without exception.

A significant disadvantage of BPU is their low hydrolytic stability under friction contact conditions. Previous studies have shown that the magnitude of wear rate of the oligesters-based BPU when adding water to the contact zone increases more than an order of magnitude [3]. Therefore, the development of more perfect and technological methods of increasing the hydrolytic stability and creation of wear-resistant composite BPU on their basis remains an urgent task today.

LITERATURE REVIEW

Today there are hundreds of brands of polyurethanes in the world, thousands of names of the initial products from which they are synthesized [2, 3]. This diversity, on the one hand, has great potential for variations with the composition of macromolecules, and hence their properties, and on the other hand, it significantly complicates the systematization of data on their relationship [4]. It is especially difficult to understand this variety for specialists in their practical application in industry. To improve the tribological characteristics, it is generally recommended to introduce solid dispersed (nanodispersed) compounds, liquids (lubricants), high modulus fibers [5, 6, 7, 8], exposure to a wide range of radiation (from thermal to electromagnetic) [9], modification of the friction surface [10, 11,

12, 13, 14, 15] etc. There are a large number of publications on this topic [16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26], but their results are scattered and not generalized, especially when it comes to creating polyurethanes for concrete operating conditions. In addition, the development of compositions of materials based on polyurethanes that are optimal for these production conditions occur mainly empirically, and the understanding of the relationship between structure and properties, as a rule, is incomplete, sometimes contradictory [11]. Thus, assessment of the polyurethanes tribolodical characteristics depending on the hardness index is commonly accepted today [11]. However, polyurethanes of the same structure but of different ability of the oligoglycolic component for crystallization can have almost twice different values of hardness, and, accordingly, values of elasticity and coefficient of friction, or have the same hardness but completely different morphology. Using many years of experience, the authors of this article recommend slowing down the processes of hydrolysis of polyurethanes in three ways, which have shown a particular efficiency of application in industry [27, 28, 29, 30, 31, 32, 33, 34, 35]: a directed change in the chemical structure, electromagnetic radiation of varying intensity, the creation of composite materials based on the principle of additional intermolecular crosslinking at the stage of manufacturing and hot processing [36].

GOAL OF WORK

The goal of this work is to develop efficient technologies and composite materials based on linear block-polyurethanes of improved durability for conditions of friction in liquid.

OBJECTS AND METHODS OF RESEARCH

The molecular structure of the polyurethanes was varied by changing the nature of the oligoglycols. As oligomeric glycols (OG) such esters were used: with a regular structure – oligobutylene glycol adipate of molecular weight ~2000 ($OBGA_{2000}$), with a variable identity period – oligoethylene glycol adipate of molecular weight ~2000 ($OEGA_{2000}$) and their mixture at equimolar ratio ($OEBGA_{2000}$). The urethane groups were formed from 4,4'-diphenyl methane diisocyanate (MDI), and low molecular weight glycol – 1.4 butanediol (butylene glycol) (BD) was introduced to obtain the block structure of the BPU.

Samples were obtained under laboratory conditions by one-step synthesis in mass at a NCO: OH = 1.0:1.0 ratio.

The morphological structure was varied due to changes in the ratio of low molecular weight glycol (butanediol) to oligoglycol during synthesis. With the increase in the amount of butanediol, there is an increase in the content of hard blocks (P_c) in block-polyurethanes, which was determined as the fraction of a molecule with a low molecular weight diol [12]:

$$P_c = \frac{90 \cdot n + 250(n+1)_2}{a \cdot 2000 + 90 \cdot n + 250 \cdot (n+1)} \cdot 100\% \qquad (1)$$

where:

P_c – content of hard blocks, %;

2000 – molecular weight of oligoglycols;

90 – molecular weight of low molecular weight butanediol;

250 – molecular weight of diisocyanate;

a – number of moles of oligoglycol;

n – number of moles of butanediol;

n+1 – number of moles of diisocyanate.

Naphthalene formaldehyde resin were used as filler [9] as well as epoxy resin ED-20 [13].

Ultraviolet irradiation was carried out using a 400-watt lamp PRK-400 with a treatment during 2 hours at a distance of 0.1-0.6 m. X-ray irradiation was performed at RUM-17 machine using a 2.64 kW source DTR-1000 with different exposure times.

To determine the elasticity modulus at tensile (E_p), standard methods of physical and mechanical tests were used.

Measurement of elasticity modulus at tensile (E_p) of the BPU was carried out in the mode of stretching at 293 K in the section of linear viscosity-elastisity ($E_p < E_k$, where E_k is the limit of elasticity of the BPU of given composition) on universal machines INSTRON-1122 and TT-DM-4-INSTRON.

Studies of wearing intensity of the BPU were carried out on a disk friction machine according to the scheme "disk-finger sample" at the speed range of 0.2-2.3 m/s at a specific load of 0.2 MPa both in the friction conditions without external supply of oil, as well as in the presence of liquid [14].

An "Epiguant" structural analyzer was used to study the topography of friction surfaces and the nature of their rupture.

The degree of crystallinity (C_k) was determined by Mathews method as the ratio of area under crystalline reflexes (S_k) on the radiograph to the total area under coherent scattering curve ($S_k + S_a$).

Previous investigations have found that molecular weight of the BPU is the most important characteristic that determines their technological and operational properties [12]. Thus, polyurethane block-copolymers of molecular weight corresponding to the characteristic viscosity index [η] = 0.8-1.1 dl/g have the most susceptible structural order, minimum values of wear intensity, friction coefficient and high heat resistance. Therefore, BPU of molecular weight that corresponds to characteristic viscosity index [η] = 0.8-1.1 dl/g were used for further studies.

FEATURES OF BLOCK-POLYURETHANES OF VARIOUS CHEMICAL STRUCTURE WEARING

The influence of chemical structure of BPU synthesized from oligoester of molecular weight 2000 and diols of different nature (ethyleneglycol (EG), butylene glycol (BG), mixture EG + BG = 1:1) on the parameters of supramolecular structure (degree of crystallinity) and properties was studied (Fig. 1).

It is revealed that BPU synthesized from butylene glycol ($C_k \approx 20\%$) have the highest degree of crystallinity with the minimum content of rigid blocks ($P_c \approx 11\%$) (Fig. 1a, curve

2). The high ability of BPU $OBGA_{2000}$ to crystallize provides a high level of elasticity modulus at tensile and wear resistance, unlike $OEGA_{2000}$ and $OEBGA_{2000}$ in the whole concentration range of changing the content of hard blocks (Fig. 1b,c, curve 2).

At a content of 30% ≤ P_c ≤ 50% there is such a structural organization, under which elastic phase works together with hard phase. Block-polyurethanes of these parameters of geometric phase morphology provide low values of wear rate under friction without lubrication (Fig. 1c). Another character of the BPU wearing is observed in the conditions of friction in water. In this case, when the influence of temperature in the friction zone is minimized, the high ordering of the flexible blocks of the BPU $OBGA_{2000}$ plays a significant role.

Thus, with a minimum content of rigid blocks P_c = 11-15%, wear intensity of polyurethanes is not of catastrophic character, as it is observed in the case of friction conditions without lubrication (Fig. 1c, curve 2).

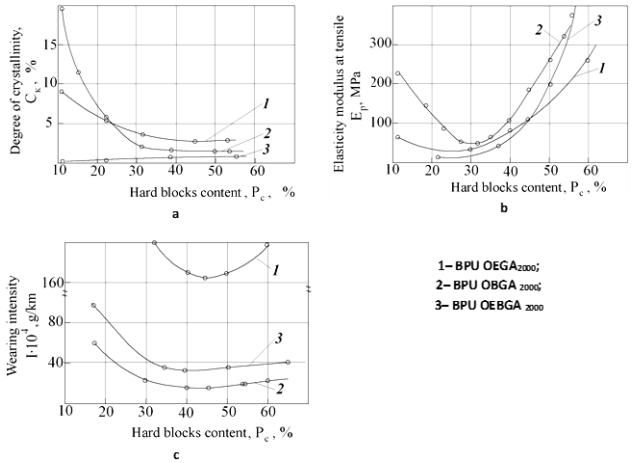
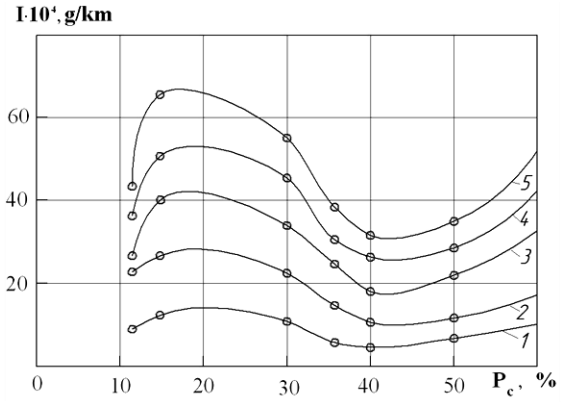


Fig. 1 Influence of BPU chemical structure on the degree of crystallinity (Ck) and mechanical properties

The decrease in the degree of crystallinity of the system from 20% to 13% causes a rapid increase in the amount of wear in three times (Fig. 2).



1– ƒ = 0,1; 2– ƒ = 0,2; 3– ƒ = 0,3; 4– ƒ = 0,4; ƒ = 0,5

Fig. 2 Dependencies of the wear rate (I) of the BPU based on $OBGA_{2000}$ under conditions of friction in water (P = 0.2 MPa, V = 2.3 m/s) on the content of hard blocks (P_c) at fixed values of the coefficient of friction (f)

Optical analysis of the friction surface of the BPU based on $OBGA_{2000}$ with P_c = 11-15% revealed the formation of a smooth wear-resistant surface (Fig. 3), which quickly adapts to the friction conditions.

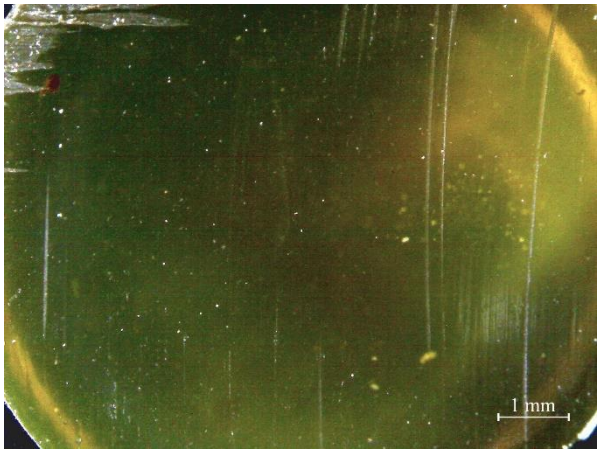
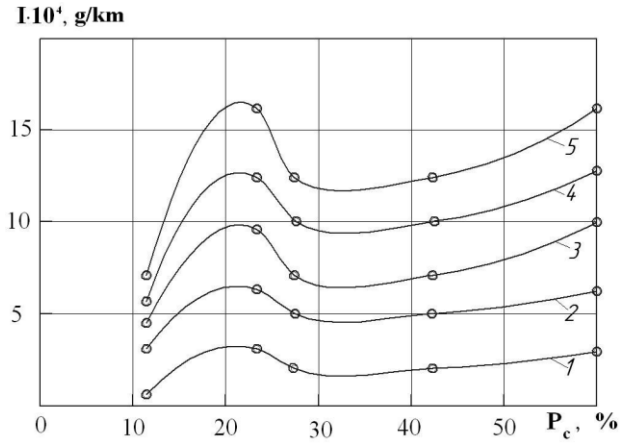


Fig. 3 The surface of the BPU based on $OBGA_{2000}$ under friction in water at P_c = 11-15%

The further growth of P_c contributes to formation of grid of hard blocks, which can take the main load and provide low values of tribological characteristics in the conditions of friction without lubrication.

The wearing intensity of polyurethanes based on $OEGA_{2000}$ with P_c in the range of 11%, as well as of BPU based on $OBGA_{2000}$, has minimal values in the conditions of friction in water (Fig. 4). With increase of P_c to 23%, the degree of crystallinity decreases from 9% to 4%, and the wear becomes catastrophic. A further downward trend in wear rate is associated with the creation of continuous microphase of rigid blocks.



1– ƒ = 0,1; 2– ƒ = 0,2; 3– ƒ = 0,3; 4– ƒ = 0,4; 5- ƒ = 0,5

Fig. 4 Dependencies of the wear rate (I) of BPU on the basis of $OEGA_{2000}$ under friction in water (P = 0.2 MPa, V = 2.3 m/s) on the content of rigid blocks (P_c) at fixed values of the coefficient of friction (f)

When comparing data for two rows of materials, the correlation between degree of crystallinity and wear rate of the BPU under friction in water is obvious. Thus, at 20% crystallinity of $OBGA_{2000}$ wear is of an order of 3.0 mg/km, and at ≈ 9% crystallinity of $OEGA_{2000}$ wear is 2.5-3 times higher (influence of hard blocks is insignificant, because

their content in both compositions is no more than 11%). That's why, the higher crystallinity of the microphase of elastic blocks is, the lower intensity of material wear under friction in water (Fig. 2, Fig. 4) is observed.

Optical analysis of the friction surface of the BPU based on $OEGA_{2000}$ with P_c = 11-15% also reveals the formation of a smooth wear-resistant surface, but with elements of incompleteness. Thus, traces of micro-cuts in the form of grooves are observed on the background of a smooth surface (Fig. 5).

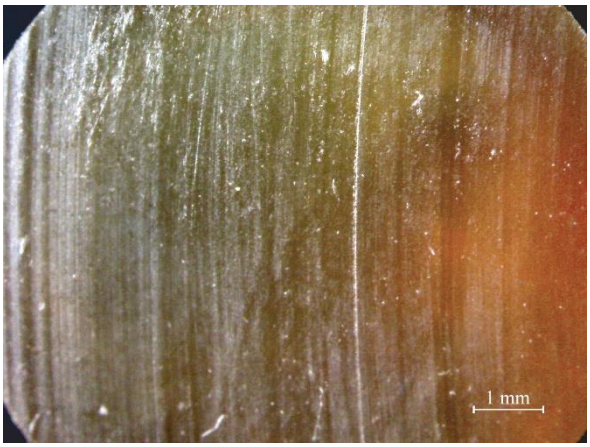
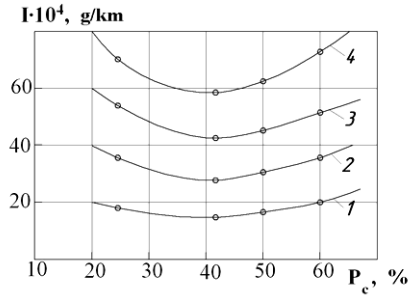


Fig. 5 The surface of BPU based on $OEGA_{2000}$ under friction in water at P_c = 11-15%

For the BPU $OEBGA_{2000}$, with minimum values of P_c from 11% to 24%, rigid blocks represent separate structures and aggregations. A material consisting predominantly of elastic oligomeric blocks with a low glass transition temperature and low crystallinity (C_k = 0.1-0.2%) is not capable to resist external impacts. That is why low values of tensile modulus (E_p) are observed in this concentration range (Fig. 1b, curve 3).

The low values of the elasticity modulus at tensile can be explained by low resistance of the elastic blocks microphase to mechanical loads and, accordingly, high contribution of irreversible deformations to the scattering of mechanical energy. In this concentration range, at any type of friction (friction without lubrication, friction in water), catastrophic wear and maximum values of the wear intensity are observed (Fig. 1c, curve 3, Fig. 6).



1– f = 0,1; 2– f = 0,2; 3– f = 0,3; 4– f = 0,4

Fig. 6 Dependencies of the wear rate (I) of BPU on the basis of $OEBGA_{2000}$ under friction in water (P = 0.2 MPa, V = 2.3 m/s) on the content of rigid blocks (P_c) at fixed values of the coefficient of friction (f)

Optical analysis of the friction surface of the BPU on the basis of $OEBGA_{2000}$ with minimal P_c did not reveal, unlike the previous series of BPU, the formation of a smooth wear-resistant surface of the samples, and it represents a matte field of wave-like formations with traces of micro-cuts (Fig. 7).

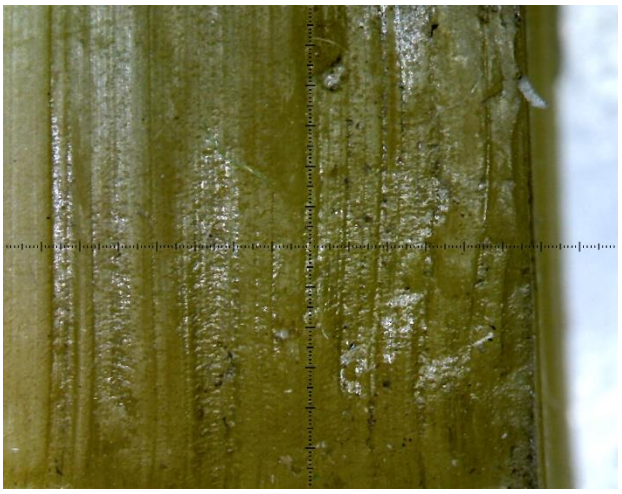


Fig. 7 The friction surface of the BPU based on $OEBGA_{2000}$ in water at P_c = 11-24%

Thus, detected smooth wear-resistant surface provides high deformation-strength and tribological characteristics of BPU on the basis of $OBGA_{2000}$ at $P_c \approx 11\%$, and protects the material from rapid destruction.

THE INFLUENCE OF ELECTROMAGNETIC FIELD OF DIFFERENT INTENSITY ON WEAR RESISTANCE OF BLOCK-POLYURETHANES IN THE PRESENCE OF WATER ENVIRONMENT

The influence of ultraviolet and X-rays on the wear resistance of BPU under hydrodynamic contact was studied in this investigation. BPU of optimal morphology, based on oligoesters were selected for the study: $OEGA_{2000}$ (40) and $OEBGA_{2000}$ (39) (values of P_c are given in parentheses). It is revealed that ultraviolet and X-ray irradiation of BPU samples significantly influences the level of their tribological properties: block polyurethanes after ultraviolet irradiation (with a radiation dose of 31.5 K·lm·s) and after X-ray irradiation (with power 2.64 kW during 6 seconds) have a higher level of wear resistance compared to the wear resistance of control BPU, both under friction without lubrication and under friction in aqueous medium. Moreover, wear resistance of the BPU under friction in liquid increases by almost an order, and with friction without lubrication - in two to three times. The increase in wear resistance is probably due to the process of structural transformation under the influence of radiation. There is an increase in the melting temperature of the BPU by 20-30 K, and in values of the characteristic viscosity of the BPU solution after irradiation by 10-50%. After irradiation, gel fraction was detected in the BPU, indicating the presence of mesh structures.

The dependence of the wear resistance of polyurethanes on the duration and dose of both ultraviolet and x-ray radiation is presented in Table 1 and in Fig. 8.

Table 1
Wear intensity of BPU in water from the dose of ultraviolet radiation

Distance from source, m	Dose of ultraviolet radiation*, K·lm s	Wear intensity** ($I \cdot 10^4$), g/km
0.1	126.0	5/4
0.2	31.5	3/3
0.4	7.9	4/5
0.6	3.5	6/6

Notes:
*1 – * at power 400 W and duration 2 hours.*
*2 – ** In the numerator – for BPU $OEGA_{2000}(40)$, in the denominator – for BPU $OEBGA_{2000}(39)$*

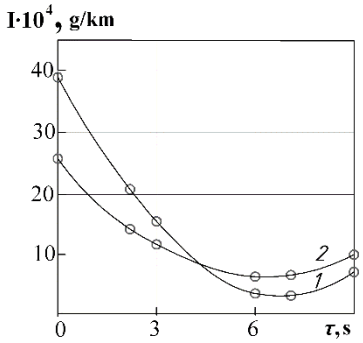


Fig. 8 Dependencies of the wear intensity in water (I) of the BPU based on $OEGA_{2000}$ (40) (1) and $OEBGA_{2000}$ (39) (2) on the time τ of their X-ray irradiation

The effect of electromagnetic irradiation of different intensity on the character of friction surface was also studied. It is established that irradiated samples of block-copolyurethanes form a wear-resistant surface similar to that found on the BPU on the basis of $OBGA_{2000}$ with a minimum content of P_c (~ 11%) and maximum elastic block capacity for crystallization (crystallinity $C_k \approx 20\%$) (Fig. 5).

It has been found that, unlike ultraviolet radiation (there is a change in the properties only in the surface layer), X-ray radiation changes the properties in the volume of the BPU. When irradiated with ultraviolet radiation, which has less ability to penetrate, the formation of a cross-linked structure can be observed in the surface layer of approximately 2-3 mm depth, providing increased durability. physical-mechanical properties of the BPU samples irradiated with ultraviolet radiation are at the level of the control samples. During X-ray irradiation (under optimal modes), the modulus of elasticity and conditional strength increase by 15-20% compared to their values for the control samples.

Thus, as a result of conducted researches, it is established that the radiation exposure of the BPU, using ultraviolet and X-rays, allows to affect their structure, which provides increase of durability of these materials almost by an order during exploitation in water.

COMPOSITE MATERIALS BASED ON BLOCK-POLYURETHANES OF INCREASED WEAR RESISTANCE IN CONDITIONS OF LIQUID FRICTION

The creation of materials based on the principle of additional intermolecular crosslinking at the stage of manufacture and hot processing is effective. So, adding from 5 to 20 mass % of naphthalene formaldehyde resin (NFS) with 5-10 mass % epoxy dian resin (EDS), due to the reaction of the interaction of epoxy or methylol groups with urethane, allows to change the spatial structure of BPU, significantly reduce the friction coefficient under conditions without lubrication and in the liquid. The presence of rigid phenolic and naphthalene aromatic nuclei in the polymer chain contributes to the formation of structure on the principle of additional intermolecular crosslinking at the stage of manufacture and hot processing. The epoxy resin has high wettability, low water absorption and increased mechanical properties [15]. The physical and mechanical characteristics of the initial and composite based on resins are given in Table 2.

It is significant that, under conditions of processing of proposed composition by injection molding, heating the resin layer, that is formed on the granules of the polyurethane block-copolymer in the first stage of processing, intensifies the heating rate of the polymer particles, as well as reduces the viscosity and improves fluidity. That is, naphthalene formaldehyde and epoxy dean resins in this case act as a high-temperature plasticizer, which allows to improve the molding of products and reduce the number of macro defects in the structure.

Table 2
Physical-mechanical characteristics of composite materials based on BPU and resins

Properties of compositions	Composition: BPU $OEGA_{2000}$ (40)	BPU $OEGA_{2000}$ (40) + 25 mass % NFS*	BPU $OEGA_{2000}$ (40)+ 15 mass. % NFS + 10 mass. % EDS**
Conditional strength at stretching (f_p), MPa	47.0	52.0	53.3
Wearing intensity (**I**), g/km:			
– friction without lubrication (**P** = 0.2 MPa, **V** = 0.4 m/s);	0.0190	0.0030	0.0022
– friction in water (**P** = 0.2 MPa, **V** = 1.2 m/s)	0.0040	0.0040	0.0035
Coefficient of friction (**f**):			
– friction without lubrication;	1.7	0.46	0.25
– friction in water	0.5	0.45	0.1

Notes:
1 – NFS – naphthalene formaldehyde resin*
*2 – EDS** – epoxy dian resin*

CONCLUSIONS

1. It was found that the introduction of water into the contact zone intensifies the wearing of polyurethanes by more than an order of magnitude. At the same time, the wear process is accompanied by the destruction of polyurethane macromolecules and separation of large particles and flakes of material in the friction zone.
2. The choice of initial polyurethanes was justified taking into account their chemical structure, peculiarities of the supramolecular structure, which made it possible to reveal the conditions for the formation of a smooth wear-resistant surface under friction in a liquid, which effectively protects the material from rapid destruction.
3. Processing the BPU with electromagnetic fields of different intensities (ultraviolet and X-ray irradiation) allows more than an order of magnitude increase of the wear resistance of polyurethanes during friction in water, as well as correct choice of optimal radiation modes, such as type and time of exposure to radiation, allows a directional approach to the creation of materials with preset characteristics.
4. Introduction of 5-20 mass. % of naphthalene formaldehyde resin and 5-10 mass. % of epoxy resin into original polyurethane, due to the reaction of interaction of epoxy or methylol groups with urethane, contributes to the increase of tribological characteristics both under friction without lubrication and friction in water.

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