

ABOUT WEAR RESISTANCE OF LINEAR BLOCK- POLYURETHANES

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Linear block-polyurethanes (BPU) are finding more and more practical applications in industry, and their recipe range is rapidly expanding. It is shown that the evaluation of the tribotechnical characteristics of BPU depending on hardness, as suggested by ISO 16365-1:2014 P1, is inaccurate and requires further research to establish the relationship between structure and properties of polyurethanes, taking into account their chemical structure and the type of initial components. The dependences of the tribotechnical characteristics of linear block-polyurethanes under friction with steel without external lubrication and in the liquid on the parameters of structural organization of the polymer were studied: at the molecular level (oligoethers, oligoesters), at the topological level (change in the molecular weight of oligomers from 500 to 2000), at supramolecular level (influence of crystallinity), at the morphological level (content of hard phase). The change in the roughness of steel counterbody during friction of polyurethanes with the content of hard blocks 20%, 50%, and 70% was also studied. A method is proposed for increasing the wear resistance of polyurethanes by thermal diffusion saturation of the surface with silicon carbide particles.

KEYWORDS

linear block-polyurethane, wear resistance, coefficient of friction, hardness, molecular weight, content of rigid blocks, counterbody roughness, thermal diffusion surface saturation

1 INTRODUCTION

Modern technology development is closely related to innovations in the area of new polymeric composite materials. Polymeric composites, generally, are used as materials of construction function. An actual problem of modern technology is the necessity of materials of high wear and abrasive resistance creation [Zia 2007, Król 2007, Chattopadhyay 2009, Shtompel 2011, Madbouly 2009, Thomas 2018].

The actuality of such polymeric materials development is determined, at first, by economic factors: necessity of ferrous and non-ferrous metals replacement, increasing the performance of machines and aggregates, expenses reduction. Construction plastics are used for replacement of bronze and other alloys of non-ferrous metals because of their high wear resistance (slip bearings, gears, rotors, clutches, face seals, etc.). The usage of polymeric materials also offers a number of

advantages: ability to create fundamentally new product designs, reduce mass of parts, reduce the noise level and increase durability, even in conditions of intensive abrasive wear and in an aggressive environment [Dyadyura 2016, Panda 2017, Duplakova 2018, Jurko 2012, 2016, Monkova 2013, Gombar 2013, Mrkvica 2012, Leško 2010, 2014, Balara 2018, Krehel 2013, Krenicky 2012, Olejárová 2016, Panda 2011, 2013, 2018, Prislupcak 2014, 2016, Ragan 2012, Valíček 2016,]. Widespread usage of polymer composites based on polyurethanes is due to the unique combination of high level of strength and elasticity, oil and gas resistance, shock and vibration resistance [Anisimov 2019, Anisimov 2019].

The actuality of presented research is primarily determined by ecologic factors - block structure polyurethanes (block-polyurethanes) provide a unique set of properties and, at the same time, they are recyclable, processed by waste-free technology on high-speed automated equipment.

However, development of recommendations of justification the choice of block-polyurethanes, which is optimal for these operating conditions, is empirical, and the concept of "composition-structure-properties" is incomplete and contradictory. Considerable amount of researches on block polyurethanes is highly specialized and scattered [Shtompel 2011, Madbouly 2009, Thomas 2018, Rimar 2016, Vojtko 2014, Zaborowski 2007, Straka 2013, 2014, Markulik 2016, Michalik 2014, Janekova 2014, Sebo 2012, Bielousová 2017, Dobránsky 2019, Panda 2012, Mačala 2012, Pollák 2018]. Therefore, development of scientific basis for the creation of block-polyurethanes of high wear and abrasion resistance, taking into account the influence of the structural organization parameters of all levels of polymeric material on the properties is an urgent scientific and technical problem and is of great practical importance for usage in industry.

2 MATERIALS AND METHODS

2.1 Materials

Block-polyurethanes of different molecular structure are selected as objects of the study. Synthesis was carried out from oligomeric esters (synthesized from adipic acid and glycols of methylene series of different nature: oligoethylene glycol adipate of molecular weight ~2000 (OEGA₂₀₀₀), oligobutylene glycol adipate of molecular weight ~500 and ~2000 (OBGA₅₀₀, OBGA₂₀₀₀), oligoethylene butylene glycol adipate of molecular weight ~2000 (OEBGA₂₀₀₀) and oligomeric ether – oligoxy tetramethylene glycol of molecular weight ~1000 (OOTMG₁₀₀₀), synthesized from tetrahydrofuran). Urethane groups were created from 4,4'- Methylene diphenyl diisocyanate (MDI), a low molecular weight glycol – 1,4 butanediol (butylene glycol) (BD) was inserted for obtaining block structure of BPU. Block-copolymer molecule consists from parts that differ in flexibility and repeat. Elastic blocks are formed from flexible parts (oligoethers or oligoesters). Rigid blocks are formed as a result of the self-organization of urethane groups. Molecular weight of all BPU samples is ~50000±70000 (characteristic viscosity [η] = 0.8÷1.1 dl/g of BPU in dimethylformamide).

2.2 Methodology of research

Molecular structure was regulated by changing the nature of oligoesters between simple and complex ones. Simple oligoesters are synthesized from tetrahydrofuran (OOTMG). Complex oligoesters were synthesized from adipic acid and low molecular weight diol of different nature: oligoester with a irregular period of identity (OEGA), oligoester of the regular structure (OBGA), mixtures of OEGA and OBGA at equimolar ratio (OEBGA). The topological structure was regulated by

change in molecular weight of oligoether in the range of 500–2000. The morphological structure of BPU macromolecule was regulated by changing the ratio of oligoester with low molecular weight diols. The number of butandiol varied from 0 to 7 moles. In this study, standard methods were used to determine the physico-mechanical characteristics, to study the structure, and also the special methods of studying the tribological characteristics [Anisimov 2019].

3 RESULTS AND DISCUSSION

It is known that BPU based on oligoesters (OEGA₂₀₀₀, OBGA₅₀₀, OBGA₂₀₀₀, OEBGA₂₀₀₀) have higher physico-mechanical characteristics, better resistance to light and thermal oxidative degradation, are easier to process into products and have lower price. BPU based on oligoethers (OOTMG₁₀₀₀) have increased hydrolytic resistance, frost resistance, are more resistant to the action of microorganisms [Anisimov 2019]. These features of linear block polyurethanes must be first of all taken into account when developing wear-resistant parts based on them. It is also important to take into account the degree of crystallinity of both individual components as well as all block material. It is known that under conditions of intensive dynamic impact BPU of a high degree of crystallinity have a tendency to “vitrification”. This leads to cracking, loss of strength and failure of critical parts of friction units of machines and aggregates (Fig. 1). High values of crystallinity are provided by an ability of a number of oligoglycols (for example, OBGA₂₀₀₀) to self-organize or hard phase formation [Anisimov 2019].



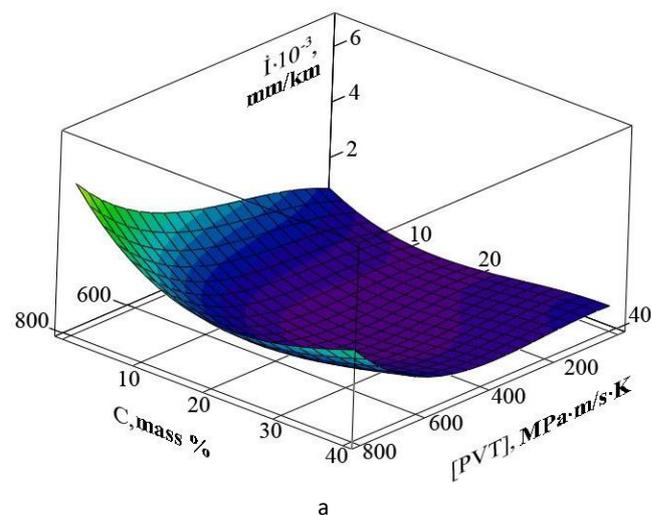
Figure 1. The characteristic “vitrification” of highly crystalline products from block-polyurethanes during exploitation under conditions of intensive dynamic impact

In our opinion, an accepted assessment of tribological characteristics of BPU from hardness or elasticity, which does not take into account the peculiarities of their molecular structure and the nature of initial monomers, is also erroneous. Block-polyurethanes of same structure but of different molecular weight or of different ability of oligoglycols to crystallize can have hardness values that differ in 2 times and, accordingly, different wear and friction coefficient values [Anisimov 2019]. Pretty much such judgments are promoted by the fact that, according to current standards, hardness values are necessarily given in the designations of trademarks of industrially produced linear block-polyurethanes [ISO 16365-1:2014]. The usage of polyurethanes gives a significant win in price when taking into account such factors as product reliability, durability and absence of downtimes, but not only the price of product. In order to determine tribological characteristics in relation to structural and exploitation parameters, the semi-industrial studies were carried out on friction machine 2070 SMT-1, according to the scheme of «shaft–partial bearing» under both non-lubrication conditions and friction in water [8]. As initial materials, BPU of hard blocks content $P_c = 20\text{--}60\%$ and molecular weight which corresponds

characteristic viscosity values $[\eta] = 0.8\text{--}1.1$ dl/g were chosen. Previous studies have shown that the initial BPU of this structure have high deformation characteristics as well as sufficient hardness [Anisimov 2019]. They are BPU based on OEGA₂₀₀₀. As studied parameters linear wear intensity and friction coefficient were chosen, and as variable factors – a few exploitation parameters: sliding velocity, specific load, temperature in the contact zone and amount of antifriction filler. The intervals of variable factors for different friction conditions are given in Table. 1. Results of experiments are presented in the form of three-dimensional plots (fig. 2, fig. 3), which show that, independently from friction conditions, influence of exploitation parameters and structural factors is decisive, so they should be taken into account during modeling and calculations of machine parts of tribological destination.

Table 1. Intervals of variable factors

Parameters	Intervals of variation				
	Sliding velocity (V), m/s	Specific load (P), MPa	Temperature (T), K	Hard blocks concentration (P _c), %	Amount of antifriction filler (C), mass %
Variation interval under friction without lubrication	0 – 2	0 – 1	273 – 400	40	0 – 40
Variation interval under friction in water	0 – 3	0 – 9	300	20 – 60	–



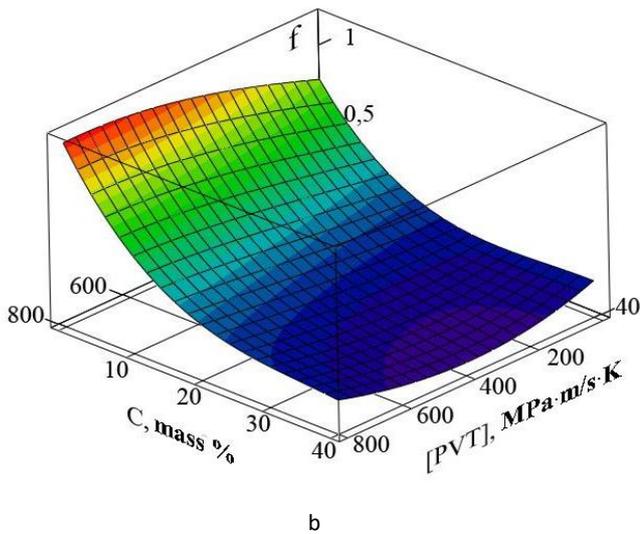


Figure 2. Dependencies of wear intensity (I) (a) and friction coefficient (f) (b) of BPU on exploitation parameter of usability [PVT] and amount of antifriction filler (C) under conditions of friction without lubrication

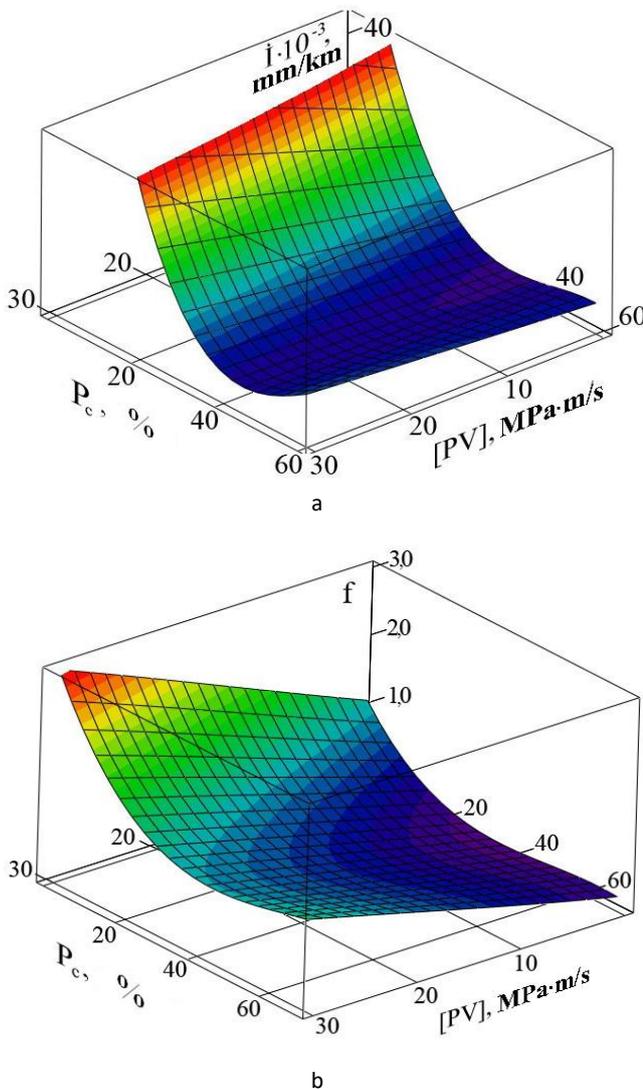


Figure 3. Dependencies of wear intensity (I) (a) and friction coefficient (f) (b) of BPU on exploitation parameter of usability [PV] and hard blocks content (P_c) under conditions of friction in water

The change in roughness of the steel counterbody during the friction of polyurethanes with the content of hard blocks 20%,

50%, and 70% was also studied. The initial roughness of steel counterbody was formed by grinding it on abrasive paper. The arithmetic average of absolute values of initial profile deviations R_a within the base length of 0.8 mm was equal to 1.0-0.85 μm (fig. 4, profilograms 1, 3, 6). For polyurethane with hard blocks concentration $P_c = 20\%$ the parameter R_a decreased to 0.59 μm during the first kilometer of friction path. There is a somewhat smoothing of peaks radiuses of the microprotrusions of the counterbody profile from 0.05 μm to 0.1 μm on profilogram 2. The further friction of BPU of a content of $P_c = 20\%$ occurred without noticeable changes in the roughness of the steel disk.

After 1 km of friction path of BPU with a content $P_c = 50\%$, its roughness decreased from $R_a = 1 \mu\text{m}$ to $R_a = 0.58 \mu\text{m}$, the profilogram 4 shows the smoothing of peaks radius of the microprotrusions from 0.05 to 0.1 μm . After 15 km of friction path, the roughness decreased to $R_a = 0.5 \mu\text{m}$, and the radius at the peaks of the microprotrusions decreased to 0.5 μm (profilogram 5). Further testing (more than 100 km of friction path) did not reveal any significant changes in the microgeometry of the friction surface of the steel counterbody, which indicates the establishment of equilibrium roughness with $R_a = 0.5 \mu\text{m}$. Profilograms 7, 8 of the surfaces of steel counterbody during friction with BPU of concentration $P_c = 70\%$ after 1 km and 15 km of path corresponded to profilograms 4, 5 for friction of BPU with $P_c = 50\%$.

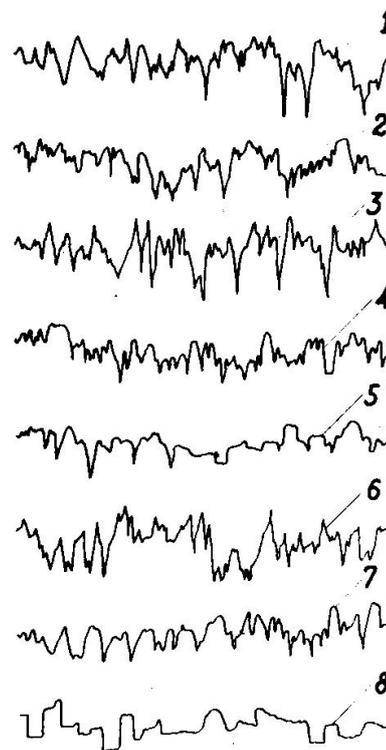


Figure 4. Profilograms of the steel counterbody surface (horizontal scale - 200, vertical scale - 10000):

- 1, 3, 6 – initial surface of the steel counterbody after grinding;
- 2 – surface after 1 km of friction path for polyurethanes of $P_c=20\%$;
- 4 – surface after 1 km of friction path for polyurethanes of $P_c=50\%$;
- 5 – surface after 15 km of friction path for polyurethanes of $P_c=50\%$;
- 7 – surface after 1 km of friction path for polyurethanes of $P_c=70\%$;
- 8 – surface after 15 km of friction path for polyurethanes of $P_c=70\%$.

However, noticeable longitudinal stripes and even scratches appeared on the counterbody, they are relatively evenly covering the entire surface, which indicates its wear during friction. A similar situation is observed after prolonged dynamic wear (more than 100 km of the friction path) of BPU with hard blocks content 70%. Roughness $R_a = 0.5 \mu\text{m}$ is maintained.

Thus, when developing wear-resistant block-polyurethanes, in addition to the molecular and topological structure, their morphological structure must be taken into account. To improve wear resistance, accelerate initial lapping and create an optimal surface roughness, a method of coating products with BPU has been developed [Anisimov 2019]. In this way, finished BPU product (part) is placed in a container with an abrasive silicon powder of a particle size 5-30 μm and undergoes heat impact at 160-180 $^{\circ}\text{C}$ for 60-80 min. The usage of silicon carbide particles, which have a polarized covalent bond molecule, ensures their strong adhesion to the polymer surface. The application of a powder layer on the surface of the product occurs as a result of thermal diffusion of the particles to surface as well as due to their partial sticking. In the last case, due to the high adhesion, strength is quite high and particles do not crumble under friction, providing high lapping speed and wear resistance under operating conditions. Products from BPU, when processing in a given temperature range, do not lose their shape and do not deform. According to the results of the analysis of BPU samples with such a layer, under conditions of friction with the steel surface, the presence of abrasive powder allows to reduce the lapping time of metal-polymer pair more than in 10 times. Surfaces with same roughness are formed in the shortest time and increase wear resistance of the friction pair in 5-6 times. After the lapping process, in fact, friction occurs between the metal and original polyurethane material. The physico-mechanical characteristics remain at a high level.

Choosing of the optimum depth of penetration of silicon carbide powder particles into the surface layer is given in table. 2, and the intensity of polyurethanes wearing with a surface layer saturated with abrasive particles of silicon carbide of different dispersion is given in table 3. The given range of time and temperature regimes of processing is chosen due to the fact that at temperatures below 433 K and time less than 60 min the processes of thermodiffusion saturation were insignificant and no important impact on the material properties was found. At temperatures above 453 K and time more than 80 min some irreversible changes occurred in the material due to the processes of thermal degradation and the related processes of strength and deformation stability decreasing (table 2).

Table 2. Penetration depth of silicon carbide powder particles into the surface layer of polyurethane

Average size of particles of silicon carbide powder, μm		1	5	20	30	60	
Time of processing, min	40	413	0	0.5	4	4	11
		433	0	0.5	5	7	15
		443	0.35	1.5	5	12	20
		453	0.5	2	6	13	20
		463	0.35	2	6	15	20
	60	413	0	1.5	6	11	18
		433	0.5	2	7	14	25
		443	0.5	2.5	8	15	30
		453	0.5	2.5	8	15	30
		463	0.5	2.5	8	15	30
	70	413	0	1.5	6	11	18
		433	0.5	2	7	14	25
		443	0.5	2.5	8	15	30
		453	0.5	2.5	8	15	30
		463	0.5	2.5	8	15	30
	80	413	0	1.5	7	11	18
		433	0.5	2	8	14	25
		443	0.5	2.5	9	15	30
		453	0.5	2.5	9	15	30
		463	0.5	2.5	9	30	30

100	413	0.3	2	8	13	25
	433	0.5	2.5	9	15	30
	443	0.5	2.5	9	15	30
	453	0.5	2.5	9	15	30
	463	0.5	2.5	9	15	30

Data in table 2 shows, that the usage of silicon carbide powder fractions of a particle size about 5–30 μm is optimal. The particles of larger sizes are poorly fixed in the surface layer of the material and quickly crumble in the process of friction. Such large particles occurrence in contact area leads to the appearance of liftings, splits on the surface and is accompanied by intensive material wear.

The saturation of the friction surface with silicon carbide particles of size less than 5 microns also does not produce significant positive results due to the intense crumbling under the influence of micro-roughness of the steel counter body.

The usage of proposed method for wear resistance increasing is recommended in the repair and mechanical shops (departments) of technological enterprises

Table 3. Intensity of wear of polyurethanes with a surface layer saturated with abrasive particles of silicon carbide

Indicator	Initial material*	Time of processing, min				
		60-80	60-80	60-80	60-80	60-80
		Temperature, K				
		453	453	453	453	453
Silicon carbide particles size, μm						
1**						
5						
20						
30						
60***						
Lapping time, min		—	60	100	110	—
Wear intensity during friction without lubrication, mg/km:	2,5 4,0	2,5 4,0	0,25 0,7	0,3 0,7	0,4 0,8	9,0 14,0
– BPU OEGA ₂₀₀₀ (P _c =40 mass %)						
– BPU OEBGA ₂₀₀₀ (P _c =39 mass %)						

Remarks:

1 – * Lapping time is compared to the full wear time of the applicated layer.

2 – ** The stage of lapping is absent. Due to the rapid crumbling of particles in the initial period, some friction is practically present between steel and the initial material.

3 – *** The stage of lapping is absent. There are deep friction traces and tears (ruptures), that are likely for abrasive wear.

4 CONCLUSIONS

Estimation of the tribological characteristics of BPU depending on hardness, as suggested by ISO 16365-1:2014 P1, is inaccurate and requires further research to establish the relationship between the structure and properties of polyurethanes, taking into account their chemical structure and the type of initial components. The nature of the oligomer (oligoether, oligoester) is the most important in case of BPU workability estimation under conditions of light and thermo-oxidative degradation, high temperatures and humidity, resistance to the action of microorganisms. Taking into account

the degree of crystallinity of both individual monomers and all the block material can let avoiding premature cracking and failure of polyurethane products during usage under severe dynamic conditions. The obtained dependences of wear intensity and coefficient of friction of the BPU on the sliding speed, specific load, temperature in the contact zone, amount of hard phase and antifriction filler showed that the influence of selected exploitation parameters and structural factors is decisive and should be taken into account when modeling the tribological parts. The analysis of the steel counterbody roughness during the friction of polyurethanes indicates the necessity to take into account their morphological structure. Proposed method of increasing wear resistance of BPU by the method of thermal diffusion saturation of the surface with silicon carbide particles can effectively control the lapping processes of the developed materials.

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