INCREASING OF WEAR RESISTANCE OF LINEAR BLOCK-POLYURETHANES BY THERMAL PROCESSING METHODS

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A significant imperfection of parts made from linear block-polyurethanes under conditions of friction contact in the presence of water medium is an intensification of hydrolysis processes and reduction of bulk strength. In order to slow down the hydrolysis process, samples of the polyurethanes were subjected to three types of thermal impacts: thermal processing during some time, infrared and laser irradiations. It has been established that processing in thermal fields of different intensity allows 5-10 times increase of wear resistance of these materials during friction in water medium.

KEYWORDS

linear block-polyurethane, friction in water, thermal processing, wear resistance, coefficient of friction

1 INTRODUCTION

Polyurethane elastomers are widely used as structural materials [Zonnenshteyn 2018]. A special place among them is occupied by linear block-polyurethanes (BPU), that combine a high level of strength and elasticity, oil and gas resistance, shock and vibration resistance [Shtompel 2008]. But under conditions of frictional contact in the presence of water medium the parts made of linear block-polyurethanes can be subjected to intensification of hydrolysis processes and decrease in their bulk strength. So the introduction of water into the contact zone intensifies the wearing of polyurethanes, ceteris paribus, more than an order of magnitude. The wearing process is accompanied by destruction of polyurethane macromolecules and separation of large particles and flakes of material in the friction zone [Anisimov 2019]. According to the authors, this is due to the intensification of the hydrolysis process and decrease in the bulk strength of the material [Panda 2014, 2018, 2019, Valicek 2016, 2017, Macala 2009, 2017, Pandova 2018, Balara 2018, Monkova 2013, Gombar 2013, Bielousova 2017, Dyadyura, 2017, Duplakova 2018, Krehel 2013, Krenicky 2012, Flegner 2019, 2020, Markulik 2016, Mrkvica 2012, Modrak, 2019, Chaus 2018, Pollak 2020, Olejarova 2017, Rimar 2016, Zaborowski 2007, Michalik 2014]. Therefore, the search for ways to increase the wear resistance of polyurethanes for different operating conditions is an urgent task and is of great practical importance for usage in industry.

2 PROPERTIES OF INITIAL POLYURETHANES

The study of relationship of structure and properties as well as assessment of various factors impact made it possible to optimize linear block-polyurethanes for wear resistance. It was found that main characteristics that determine properties of polyurethanes of block structure are their molecular weight and content of hard segments in the macromolecule [Anisimov 2019]. Materials with desired characteristics can be obtained by adjusting these parameters at the synthesis stage [Bochen 2009, Domanski 2016].

Linear block-polyurethanes based on oligomeric esters with a variable identity period - oligoethylene glycol adipate of molecular weight ~2000 (OEGA2000) and mixed - oligoethylenebutylene glycol adipate of molecular weight ~2000 (OEBGA2000) were chosen for studies. Urethane groups were formed from 4,4′-diphenylmethanediisocyanate (MDI). To obtain the block structure of BPU, a low molecular weight glycol of 1.4 butanediol (butylene glycol) (BD) was introduced. A block polymer molecule consists of parts that are flexible and repeatable. Elastic blocks are formed from flexible parts (oligoesters). Hard blocks are formed due to the self-organization of urethane groups. The abbreviated designation of the materials studied has the form: BPU OEGA2000 (40) and BPU OEGA2000 (39) (the values of the content of hard blocks in percentages are given in parentheses).

The molecular weight of all BPU samples was ~50000÷70000 (characteristic viscosity [η] = 0.8÷1.1 dl/g of BPU in dimethylformamide). The BPU of selected compositions have the most favorable combination of high deformation characteristics with sufficient frame hardness, as well as the global trend to stable production growth and the widest practical application [Murcinkova 2013, Anisimov 2019].

The main physico-mechanical characteristics of initial block-polyurethanes at normal temperature of 293 K are given in Table 1 and at different temperatures (from 223 K to 353 K) in Fig. 1.

Table 1. Physico-mechanical characteristics of BPU

Indicator	BPU $OEGA_{2000}$ (40)	BPU $OEBGA_{2000}$ (39)
Density, kg/m^3	1230	1160
Shore hardness, scale A, conventional units	90.2	84.2
Conditional tensile strength ($\mathbf{f_p}$), MPa	34.0	29.0
Elastic modulus ($\mathbf{E_p}$), MPa	45.0	10.0
Elongation at rupture, ($\boldsymbol{\varepsilon_p}$), %	850	675
Residual elongation, ($\boldsymbol{\varepsilon}$), %	50	85
Vicat softening temperature (T_p), K	463	438
Degree of crystallinity, ($\mathbf{C_к}$), % (vol.)	3.0	< 2.0

The behavior of initial polyurethanes at low (negative) temperatures is determined by glass transition temperature of the elastic blocks microphase. Analysis of the curves, shown in

Fig. 1, shows that there is a characteristic presence of a clear one low-temperature transition at T = 253 K for both initial materials, which corresponds to the glass transition temperature of elastic blocks microphase and at least one transition in the region of positive temperatures at T = 303÷323 K, which corresponds to melting point of crystalline regions of the elastic blocks microphase.

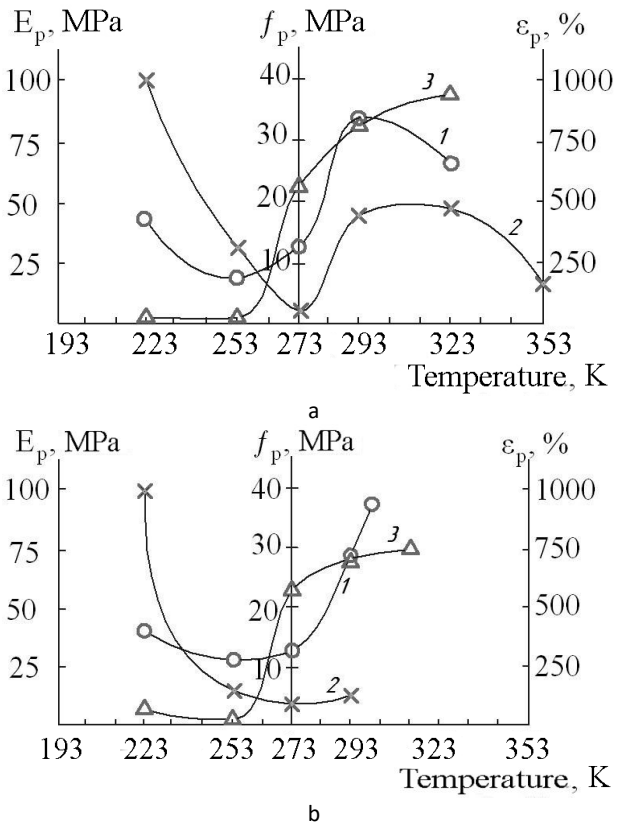

Figure 1. Physico-mechanical characteristics of initial polyurethanes at different temperatures (I) (a)based on BPU $OEGA_{2000}$(40); b – based on BPU OEBGГA_{2000}(39):
1 - conditional tensile strength f_p,
2 - elastic modulus E_p,
3 - elongation at rupture, (ε_p), %

Moreover, the greater complexity of oligoglycol component is, the less expressive transition in region of positive temperatures is observed, hence the degree of phase separation is lower and the temperature range for their processing into products is wider.

PROPERTIES OF LINEAR BLOCK-POLYURETHANES AFTER THERMAL PROCESSING OF DIFFERENT INTENSITY

To slow down the hydrolysis process, the samples of considered polyurethanes were subjected to thermal effects of three types: processing in a heat chamber, infrared and laser irradiations. The choice of thermal processing was determined by the simplicity of reproduction in any conditions (laboratory, industrial), as well as economic factors [Kestelman 1980]. When holding in a thermal chamber, both temperature and time intervals of processing were varied. Under infrared irradiation samples from various BPU were placed at distance of 1 m from the source (IRP-1000 lamp, radiation power of 1000 W), and processing time was changed so as not to cause their melting. The samples were processed similarly under laser irradiation, where the emitter was a standard CO_2 laser (LG-22) with a radiation wavelength of 10.6 μm and an output power of 20 W. To change the amount of absorbed thermal energy, the processing was carried out both by a focused and a defocused beam.

The duration of thermal field influence was in interval of 0.5 - 80 hours in the first and second methods, and 1 - 6 seconds in the third one. Accordingly, in the first two types of processing, the properties changed in all the volume of material, and in the third one - in surface layers.

The samples were tested for friction and wear at normal temperature on a disk friction machine with a specific load of 0.2 MPa and a sliding speed of 1.2 m/s. A counterbody was a disk made of steel 45, hardened to HRC 45-48, roughness of which corresponded to the parameter Ra = 0.2-0.8 μm [Tishchenko 1985, Cacko 2014, Krenicky 2015].

To determine an optimal temperature regime for processing in a heat chamber, polyurethane samples were kept for the same time interval with a stepwise change in temperature in the range 343-423 K.

As results analysis has shown, the wear resistance of thermally processed BPU $OEGA_{2000}$ (40) and BPU $OEBGA_{2000}$ (39) increases in 5-10 times. The minimum wear value was observed for the samples, that are processed at a temperature of 373-383 K. Changing the processing time at this temperature ambiguously affects the wear resistance of the studied polyurethanes. Figure 2 shows, that dependence of the BPU wear resistance on the thermal processing time has an extreme character. By the way, the shift of minimum value of wear towards longer processing time for polyurethane BPU $OEGA_{2000}$ (40) is obviously due to the presence of a diffusion stage of reactions in a more viscous medium.

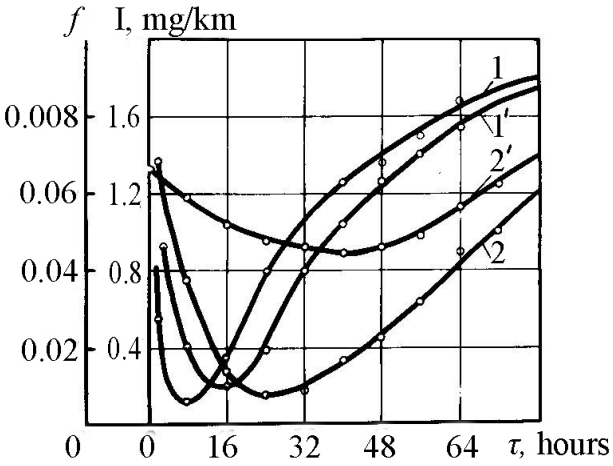

Figure 2. Dependence of wearing intensity **I** (1, 2) and coefficient of friction **f** (1^I, 2^I) of linear block-polyurethanes on heat processing time (P=0,2 MPa; V=1,2 m/s)
1; 1^I – BPU $OEBGA_{2000}$ (39);
2; 2^I – BPU $OEGA_{2000}$ (40)

Decrease in wear resistance of BPU $OEBGA_{2000}$ (39), when processing during more than 8-10 hours and of BPU $OEGA_{2000}$ (40) during 24-26 hours, is caused by the occurrence of thermal destruction processes there, decrease in molecular weight, and decrease in the mobility of molecules. The obtained results completely correlate with the data of P. Wright and A. Cumming [Wright 1973], according to which, when processing molded products from polyurethane based on polyethylene adipate (1.0 mol), MDI (2.4 mol) and diethylhydroquinone (1.21 mol) during 24 hours at 383 K, they achieved an improvement in the strength properties of this material, and obtained low values of residual deformation.

As shown by the results of thermomechanical and X-ray analyzes (Table 2), the softening start temperature during heat processing of the studied polyurethanes increases on 20-30 K, as well as for $OEGA_{2000}$ (40) the degree of crystallinity increases,

which indicates the occurrence of structural ordering processes inside the system and the expansion of workability temperature interval.

Table 2. Effect of thermal processing on properties of BPU

Parameters	Polyurethane					
	BPU $OEGA_{2000}$(40)			BPU $OEBGA_{2000}$(39)		
	1	2	3	1	2	3
Time of thermal processing, hours	0	24	72	0	8	72
Softening start temperature, K	463	488	473	438	468	478
Degree of crystallinity, $C_к$, % (vol.)	3.0	4.1	3.7	< 2	< 2	< 2

A similar dependence of wear resistance on processing time was also observed for subjected to infrared radiation samples (Fig. 3). However, in this case the wear value is slightly higher than when holding in a thermal chamber. Apparently, it is caused by a shorter processing time, which leads to some incompleteness of physicochemical processes in the block material.

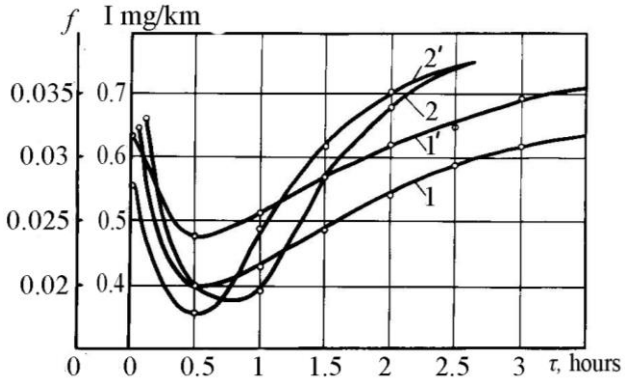

Figure 3. Dependence of wearing intensity **I** (1, 2) and coefficient of friction **f** (1^I, 2^I) of linear block-polyurethanes from the time of infrared radiation processing (P=0,2 MPa; V=1,2 m/s):
1; 1^I – BPU $OEBGA_{2000}$ (39);
2; 2^I – BPU $OEGA_{2000}$ (40);

Laser thermal processing of polyurethane samples leads to a change in their structure in the surface layers mainly. In this case, a smooth decrease in the wear value with an increase in the time of laser irradiation (Fig. 4) is characteristic. When processing polymer samples for more than 4 seconds, then intensive thermal destruction processes occur in the surface layers, accompanied by surface melting. In the process of friction, such a layer either wears out very quickly (typical for BPU $OEGA_{2000}$(40)), or participates in the formation of a thin film in the contact zone on the surface of the polymer sample (BPU $OEGA_{2000}$ (39)). Optical studies have shown that such a film is formed by friction of polyurethane samples processed in thermal chamber as well as samples, that are processed by infrared irradiation.

The nature of dependence of the friction coefficient on the type and time of thermal processing is similar to the nature of change in wear resistance (Figs. 2-4). Apparently, the formation of thin film during friction not only increases the resistance of materials to abrasion, but also contributes to sliding processes.

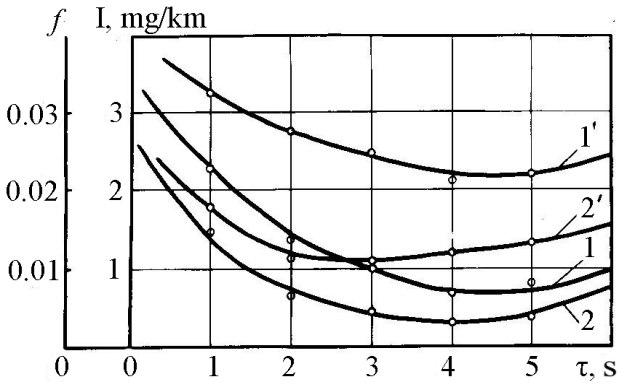

Figure 4. Dependence of wearing intensity (1, 2) and coefficient of friction (1^I, 2^I) of linear block-polyurethanes on the time of laser irradiation (P=0,2 MPa; V=1,2 m/s):
1; 1^I – BPU $OEBGA_{2000}$ (39);
2; 2^I – BPU $OEGA_{2000}$ (40)

Comparison of the results of friction and wear studies of thermal processed polyurethanes showed the complete identity of changes in their tribological characteristics, regardless of the type of thermal processing. That allows us to conclude that occurring in the BPU transformations are of the same nature. In our opinion, the increase in the wear resistance of thermal processed BPU is caused by crosslinking of linear polymer chains and a decrease in the structure defects. It is known [Buist 1978] that when polyurethanes are heated to the temperature of 343–353 K, the bonds of NH urethane group and carbonyl of urethane group are broken, and heating to temperatures of more than 423 K breaks the bonds between urethane group and carbonyl of the urethane group. Therefore, bond breaking should be accompanied by both an increase in the mobility of molecular chains and an initiation of the formation of additional hydrogen bonds. The change in the mobility of polyurethane macrochains throughout all the block of material completely depends on the power of the heat flow and its duration.

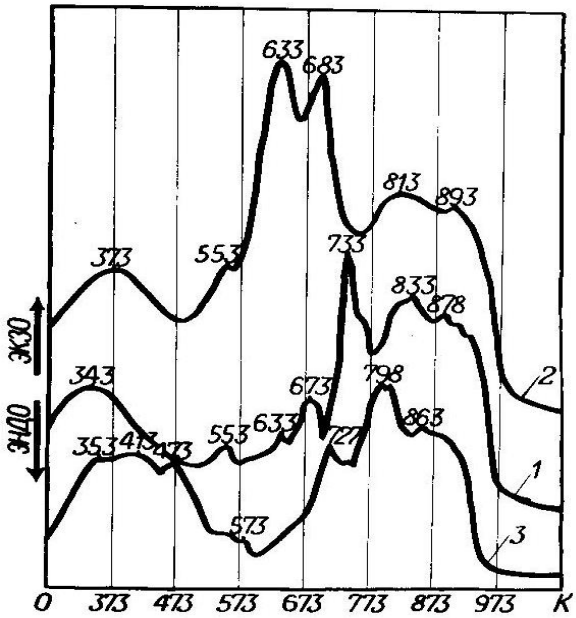

Figure 5. Diffractogram BPU $OEGA_{2000}$ (40):
1 – initial;
2 – thermal processing (24 hours, 373 K);
3 – thermal processing (72 hours, 373 K)

The results of differential thermal analysis show the formation of additional hydrogen bonds in the material and an increase in the upper temperature limit of workability of the parts made of polyurethane after thermal processing. When using the BPU

$OEGA_{2000}$ (40), as an example, it can be seen (Fig. 5) that the bonds breaking between the NH urethane group and the carbonyl of the polyester group in initial material is characterized by an exothermic peak in the region of 343 K. This exothermic peak shifts to the region of elevated temperatures by 30 degrees in subjected to thermal processing polyurethane. A further increase in temperature causes deep oxidative transformations of the degradation products.

Crosslinking formation leads to an increase in the density of the structure and an increase in the strength properties of polymers. Moreover, in the process of friction, the rate of diffusion of water molecules into the surface layers decreases and hydrolysis slows down, thereby some increase in the wear resistance of linear block polyurethane is observed. In addition, the rate of wear-resistant film formation on the friction surface and its stability significantly increase after thermal processing. It plays the role of a kind of membrane, which localizes the hydrolysis processes in thin surface layers and protects the material from rapid destruction, thereby contributes to increase in wear resistance.

Thus, the BPU processing in thermal fields of various intensities allows a 5-10 times increase in the wear resistance of these materials during friction in the water medium and it lets to recommend them for industrial usage in such pump and compressor equipment as cuffs and seals.

CONCLUSIONS

1. It was found that the introduction of water into the contact zone intensifies the wear of polyurethanes, ceteris paribus, more than an order of magnitude. Preliminary studies have shown that the wearing process is accompanied by the destruction of polyurethane macromolecules and the separation of large particles and flakes of material in the friction zone.
2. The choice of initial polyurethanes is justified, taking into account their molecular weight and content of hard segments in macromolecule, which have the most favorable combination of high deformation characteristics with sufficient frame stiffness.
3. To slow down the hydrolysis process, the samples of considered polyurethanes were proposed to be subjected to thermal effects of three types: processing in thermal chamber, infrared and laser irradiation.
4. It has been established that processing the linear block-polyurethanes in thermal fields of various intensities makes it possible to increase the tribological characteristics of these materials during friction in water medium in 5-10 times due to the formation of additional hydrogen bonds, and formed on the surface wear-resistant film plays the role of a kind of membrane, which localizes hydrolysis processes in thin surface layers and protects the material from rapid destruction, thereby contributes to increased wear resistance.

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