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POLYMER-COMPOSITES BASED ON AROMATIC POLYAMIDE AND ARAMID FIBER FOR HEAVY-DUTY FRICTION AND SEALING UNITS

***Oleg Kabat*[1,2], *Yulia Boiko*[2,✉]**

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Abstract. The paper substantiates the selection of a polymer and filler based on aromatic polyamide to create a material of friction pairs that operate in heavy-duty friction and sealing units of up-to-date vehicles and machinery. The effect of load and sliding speed on the tribological behavior of the initial aromatic polyamide and filled 15 %-aramid fiber was studied. As a result of tribological studies, the terminal operating conditions of products made of initial aromatic polyamide and a composite based thereon, filled with aramid fiber, have been determined. The basic physical and mechanical properties of the materials under study have been determined, and it is established that the introduction of aramid fiber into the aromatic polyamide contributes to a 10–15 % reduction in the level of the properties thereof due to the occurrence of micro- and macrodefects of its structure.

Keywords: aromatic polyamide, aramid fiber, tribotechnical properties, coefficient of friction, linear wear intensity, strength.

1. Introduction

The up-to-date vehicles and machinery, in most cases, cannot operate with a high level of reliability and durability without the use of polymers in friction and sealing units[1–5].

The advantages of using polymer materials in friction and sealing units include self-lubricating ability, lower density compared to metals and alloys thereof, high level of chemical stability, *etc.*[6, 7]. However, their positive properties are in conjunction with disadvantages. One of them is that in the event of changing the operating conditions (loading, sliding speed, temperature, *etc.*), the tribological characteristics of the friction units, where polymers operate, can significantly change[8].

So, the main purpose of the research is to create composite materials based on polymer that can work in friction units of modern vehicles and machinery, ensuring their high level of reliability and durability.

The studies of tribological polymers have shown that fluoropolymers (0.1–0.25), polyamides (0.21–0.25), polyetherketones (0.18–0.24), phenolic polymers, *etc.*, feature the lowest coefficient of friction[9].

Fluoropolymers feature one of the lowest coefficients of friction in the case of friction engagement with steel. In friction modes without lubrication, it ranges from 0.1 to 0.25. Such polymers can be used in friction units that operate in chemically active media at sufficiently high temperatures. However, fluoropolymers have a reasonably low level of physical and mechanical properties and are easily deformed under low loads[10, 11].

Friction units with aliphatic and aromatic polyamides (0.21–0.25) differ in quite small values of the coefficient of friction as well. These materials feature rather high values of physical, mechanical, and thermophysical properties and can be used in heavily loaded friction units over a wide temperature range (from 233 to 513 K)[12].

Polyetherketones are quite widely used as materials for friction units. These materials are capable of operating at high levels of load and temperature[13]. Due to their high chemical stability, they are often used in units contacting with aggressive media[14]. The disadvantages of these polymers are their high cost and the sophisticated technology of processing them into products.

Thermosetting (cross-linked) polymers, *i. e.*, phenolic resins, epoxy resins, polyester, organosilicon resins, *etc.*, have gained widespread use as matrix materials in tribological engineering. Thus, phenolic resins feature a stable modulus of elasticity of 10–20 GPa, maintaining the value up to 393 K, a low coefficient of thermal expansion, a high level of chemical stability, *etc.*[15, 16].

Based on the above, it is possible to conclude that the most optimal tribological materials are aromatic

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[1] State Higher Educational Institution, Ukrainian State University of Chemical Technology, Gagarin Ave., Dnipro, 49600, Ukraine
[2] Dnipro State Agrarian and Economic University, 25, Efremov str., Dnipro, 49600, Ukraine
✉ *julia.boyko@gmail.com*

polyamides, which, when used as matrix polymers for the realization of a material for friction units, feature the greatest number of advantages as opposed to others.

To reduce the disadvantages of aromatic polyamides and improve the level of properties, it is filled with materials of various nature and morphology[17, 18].

The well-known ones are the polymer composite materials based on aromatic polyamides, which are filled with dispersed carbon and silicon-containing materials[19,20]. Their advantages include a fairly high level of structural (σ_y up to 250 MPA) and thermophysical properties (T_{vc} up to 295 K). However, they feature a relatively high coefficient of friction and wear in the case of friction engagement with metal surfaces. Polymer composites based on aromatic polyamides, which are filled with fibrous materials, have gained widespread use[21, 22]. They feature a good level of tribological properties, along with a high level of physical-chemical and thermal-physical properties. Based on this, they are fibrous materials that are mostly used for filling aromatic polyamides.

The high status among polymer reinforcing fibers is given to aramid fibers based on aromatic polyamides, which include kevlar, armos, terlon fibers, and fibers based on aromatic heterocyclic polyarylene[23, 24]. These fibers have exceptionally high strength characteristics, high heat endurance and thermal resistance, and low flammability.

The high performance of aramid fibers is explained by the high degree of structural elements' order and the rigidity of their chains, which include a large number of aromatic fragments and a high degree of macromolecule orientation. These structural features, when combined with high crystallinity and the absence of flexible links between aromatic bonds, ensure their thermal resistance up to 523–623 K. A special feature of the structure of aramid fibers is the apparent anisotropy of their mechanical properties. Due to high adhesion to most binders, their use as reinforcing fillers in organic fiber composite ensures the production of materials with low porosity and exceptionally high strength characteristics[25]. Therefore, these particular fibers were used for the realization of polymer composite materials.

2. Experimental

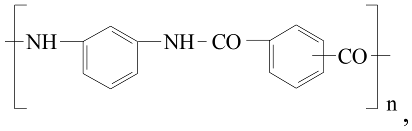
2.1. Materials

Aromatic polyamide (produced in China) poly-*m*-, *n*-phenyleneisophthalamide was used as the polymer matrix, which is a polymer of heterocyclic structure, consisting of aromatic fragments of various structures connected by amide bonds. It is a product of the polycondensation of aromatic diamine and isophthalic acid chlorohydride. Like other aromatic polyamides, it is characterized by glass transition (563 K) and melting temperatures (703 K) and a fairly high (up to 533 K) temperature of long-term operation, increased radiation, chemical stability, and other important properties. The molecular weight of the final polymer is 20000–70000.

The poly-*m*-, *n*-phenyleneisophthalamide is a powder-like polymer of pink-white color with a density of 200–400 kg/m^3 and a specific viscosity of 0.5 % in dimethylformamide with 5 % lithium chloride of at least 0.8, and the glass transition point of 543 K. The crystallization takes place when heated to 613–633 K (T_{pl} = 703 K).

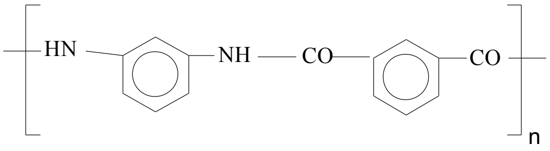
The poly-*m*-, *n*-phenyleneisophthalamide, like other aromatic polyamides, represents a linear heterogeneous-chain polymer, the macromolecules whereof are constructed from aromatic fragments of various structures that are connected by amide bonds.

The poly-*m*-phenyleneisophthalamide, a linear aromatic polyamide,



is known as Phenilon (Ukraine), Nomex (USA), Konex (Japan)[2].

The aramid fiber (Terlon), which is a product of the interaction of *poly*-n-phenyleneisophthalamide and terephthalic acid dichloroanhydride and which is manufactured by the DuPont Company, was used as a filler. This filler, capable of being used in a wide temperature range (from 23 to 548 K), features a high tensile strength, increased resistance to repeated strain, low heat shrinkage, electrical conductivity, and excellent chemical stability.



For the purpose of being used as a study object the aramid fiber was crushed into pieces about 1 mm long using scissors.

2.2. Methods

The study of tribological properties in the dry friction mode was carried out using a pin-disk machine according to the scheme "disk (steel 45, heat-treated to a stiffness of 45–48 HRC with a surface roughness *Ra* = 0.16–0.32 microns) – finger" (polymer or composite) in the range of specific loads of 0.2–0.8 MPa, sliding speeds of 1.0–2.0 m/s; the friction path was 1000 m.

The wear of the samples was determined by the weight-change method on VLR-200 analytical balance with an accuracy of 0.0002 g. After determination, it has been converted to the linear wear intensity.

The microstructure of the friction surface was studied using a NEOPHOT 32 light microscope equipped with the electronic AxioCam105 Color Camera.

The density was measured by the hydrostatic method according to ISO 1183-1:2019 – Plastics – Methods for determining the density of non-cellular plastics – Part 1: Immersion method, liquid pycnometer method and titration method. The measurement was performed using a modified VLR-200 analytical balance with a density measurement module.

The microhardness was measured using a PMT-3 microhardness meter in accordance with ISO 14577-1:2015 – Metallic materials – Instrumented indentation test for hardness and materials parameters – Part 1: Test method.

The stresses at the point of yield strength (σ_y), when compressing the initial polymers and composites based thereon, were determined in accordance with ISO 604 using the universal testing machine FP-100/1.

3. Results and Discussion

According to previous studies, the optimal fiber content in aromatic polyamide, from the point of view of physical-and-mechanical, tribological properties, and economic indicators, is 15 % by weight[26, 27]. Therefore, we will further study a polymer composite with precisely this filler content.

It is known that the friction units of vehicles and machinery operate at specific loads and sliding speeds. Therefore, it is interesting to study the effect of these external factors on the friction and wear of the initial aromatic polyamide. The results of such studies are shown in Fig. 1.

Graphs of the load-dependent coefficients of friction (Fig. 1, *a*) for all of the studied sliding speeds feature a similar nature. Pursuant thereto, with the increasing load, there is an increase in the coefficient of friction, which is a consequence of an increase in the resistance to movement in the friction pair. The value of the coefficient of friction increases from 0.390 to 0.435 with the increase of the load in the friction unit at the sliding speeds under study. This leads to an increase in the temperature of friction surfaces and can, under appropriate conditions, contribute to the sub-melting of the surface layers of the polymer, which, in turn, will lead to the transition of the friction unit to the emergency mode of operation with a future rapid loss of its performance capacity.

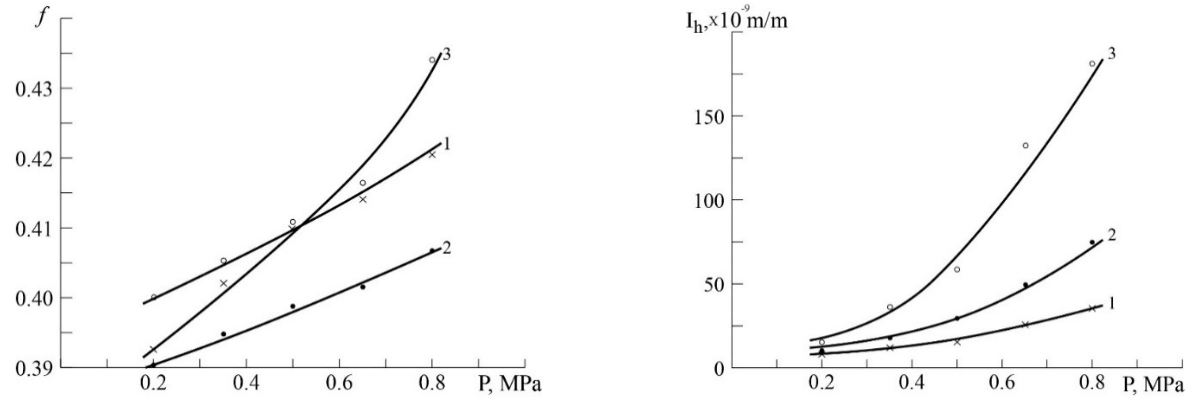
Also, a note should be made of the growth intensity of the coefficient of friction at various sliding speeds of the friction pair. So, at sliding speeds of 1 and 1.5 m/s, the intensity of increase in the coefficient of friction is approximately the same, and even at 2 m/s it significantly exceeds their values. The high intensity of the increase in the coefficient of friction indicates a significant increase in temperature in the friction zone, which can cause the sub-melting of the surface layers of the polymer sample, followed by the intensive failure.

To confirm this assumption, micrographs of the friction surfaces of aromatic polyamide after friction engagement with steel in the studied range of sliding velocities have been made (Fig. 2).

Graphs of load-dependent linear wear intensities (Fig. 1, *b*), at all the studied sliding speeds, feature a similar nature, as well. Pursuant thereto, with increasing load, there is an increase in wear of the polymer material in the friction pair. Thus, the minimum wear value of aromatic polyamide is observed at a load of 0.2 MPa and a sliding speed of 1 m/s ($I_h = 7\times10^{-9}$ m/m); the maximum values are observed at 0.8 MPa and 2.0 m/s ($I_h = 180\times10^{-9}$ m/m). It should be noted that such a significant increase in wear at the maximum loads and sliding speeds under study indicates the transition of the friction pair to emergency mode of operation, when further use of the friction unit is not appropriate. It is worth noting that there is a significantly higher intensity of wear growth in the friction pair, which operates at a sliding speed of 2.0 m/s compared to 1 and 1.5 m/s. This indicates that the terminal (maximum permissible) mode, at which the friction pair operates in normal mode, has been reached. According to the studies conducted, it is arguable that the friction unit under study operates in normal mode at sliding speeds of 1 and 1.5 m/s over the entire studied load range (from 0.2 to 0.8 MPa), and at a sliding speed of 2 m/s only at loads from 0.2 to 0.5 MPa.

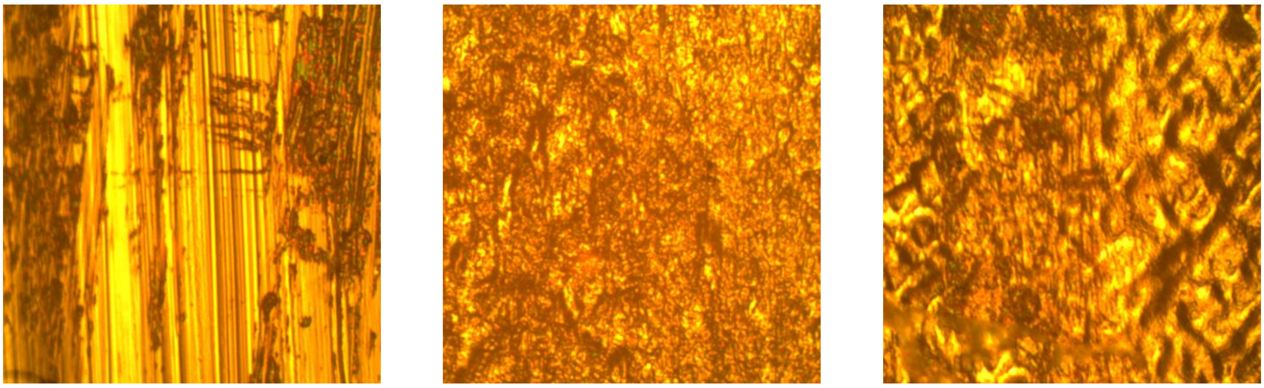
It raises the interest to study the effect of aramid fiber on the tribological properties of aromatic phenylone polyamide in a friction pair with steel. Fig. 3 represents experimental results of the influence of extraneous factors on friction and wear of a composite material based on aromatic polyamide filled with aramid fiber.

According to the studies conducted, it is arguable that graphs of the load dependencies of the friction coefficient of the composite feature have a similar nature. Thus, with an increase in the load from 0.2 to 0.8 MPa, an increase in the coefficient of friction is observed from 0.295 to 0.325 over the studied range of sliding speeds of the friction pair. It is worth noting that the values of the coefficient of friction are 24–26 % lower for the composite-steel friction pair compared to the same value in the friction of the initial phenylone. This is related to the reinforcing effect of the fiber on the polymer matrix and the presence of a new component in the friction pair, which contributes to changes in the friction nature. Reduction of the coefficient of friction will lead to a decrease in the temperature on the friction surface, which makes it possible to use the engineered material under higher loads and sliding speeds as opposed to the original phenylone. To confirm this assumption, micrographs of the friction surface of polymer composites after friction engagement with steel were obtained (Fig. 4).



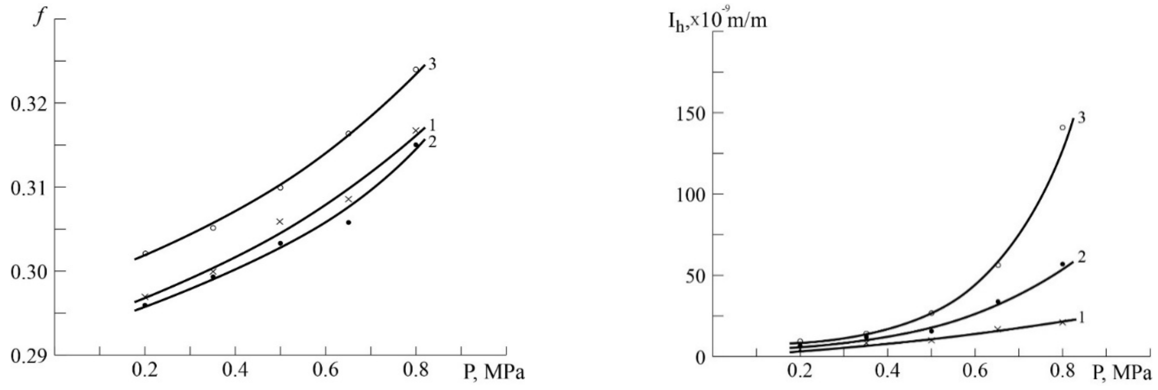
a b

Fig. 1. Dependences (a) of the coefficient of friction (f_{TR}) and (b) the linear wear intensity (I_h) for aromatic polyamide in the event of friction engagement with steel under the load (P) at sliding speeds: 1 – 1 m/s; 2 – 1.5 m/s; 3 – 2.0 m/s



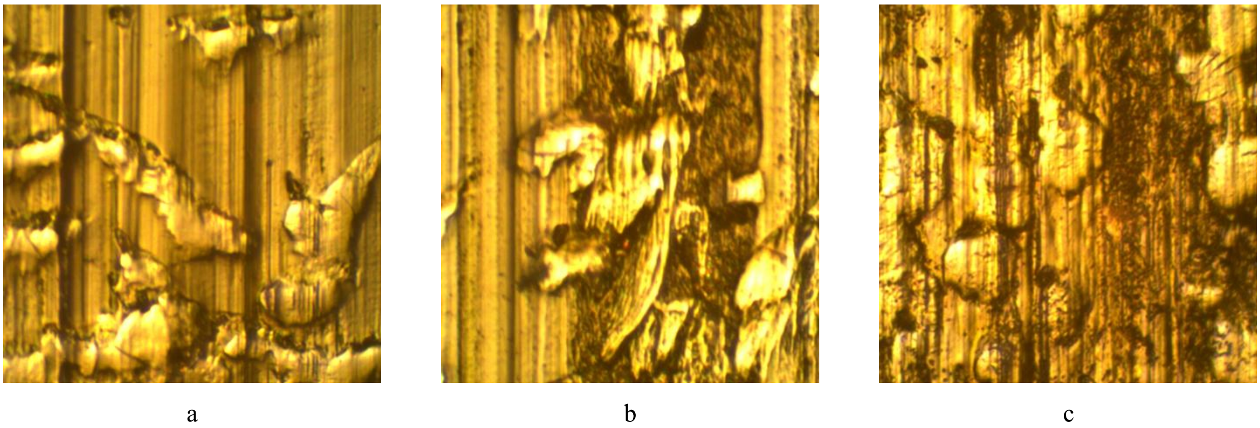
a b c

Fig. 2. Micrographs of the friction surfaces of the initial aromatic polyamide after friction engagement with steel at a steady load of 0.8 MPa and sliding speeds: a – 1.0; b – 1.5; c – 2.0 m/s (200-fold increase)



a b

Fig. 3. Dependences (a) of the coefficient of friction (f_{TR}) and (b) the linear wear intensity (I_h) for polymer composite 90 % aromatic polyamide + 15 % Terlon fiber during friction engagement with steel under the load (P) at sliding speeds: 1 – 1 m/s; 2 – 1.5 m/s; 3 – 2.0 m/s



a b c

Fig. 4. Micrographs of the friction surfaces of the polymer composite 90 % aromatic polyamide + 15 % terlon fiber after friction engagement with steel under the constant load of 0.8 MPa and sliding speeds: a – 1.0; b – 1.5; c – 2.0 m/s (180-fold increase)

The analysis of the obtained micrographs indicates that the friction surface obtained at a sliding speed of 1 m/s features apparent depressions and protrusions of roughness without traces of charring or sub-melting of the surface layer, which indicates the normal operation of this friction unit. In the case of increasing the sliding speed to 1.5 and 2.0 m/s, the areas of charring of the polymer appear on the polymer friction surface along with depressions and roughness protrusions. However, the amount and intensity thereof do not cause the sub-melting of its surface layer, which makes it possible to assert the possibility of reliable and long-lasting operation of the friction unit under the studied loads and sliding speeds.

According to the studies conducted, it is arguable that the studied polymer composite-steel friction unit operates in normal mode at loads from 0.2 to 0.8 MPa in the range of sliding speeds from 1 to 2 m/s.

It raises the interest to study the effect of terlon fiber on the physical and mechanical properties of the resulting polymer composite based on aromatic polyamide. The results of studies are presented in Table below.

Table. Values of physical and mechanical properties of the initial aromatic polyamide and polymer composite material 85 % phenylone + 15 % terlon

No.	Property factor	Factor value	
		Initial aromatic polyamide	composite material 85 % phenylone + 15 % terlon
1	Density ρ, kg/m^3	1325	1334
2	The stresses at the point of yield strength σ, MPa	213	176
3	Microhardness, MPa	165.8	152.45

According to the analysis of the physical and mechanical properties of the materials under study, a conclusion can be drawn that the densities of the initial aromatic polyamide and the composite based on it have fairly close values with a slight advantage of the latter. This is related to a certain structuring of polymer composites when fiber is introduced into their composition compared to the initial polymer. A note should be made of a slight decrease in the level of strength properties (stresses at the compression yield strength and microhardness) by 10–15 % in filled composites. This is a consequence of the presence of structural micro- and macrodefects therein, which occur due to an insufficient uniformity of fiber distribution in the polymer matrix. However, this reduction is not critical and does not limit the use of engineered composites in friction and sealing units.

4. Conclusions

As a result of the conducted studies, the influence of the load and sliding speed on the tribological properties of the initial aromatic polyamide and the polymer composite material based thereon, filled with terlon fiber, has been established. It was found that with an increase in the load from 0.2 to 0.8 MPa and the sliding speed from 1 to 2 m/s for all the materials under study, an increase in the coefficient of friction and the linear wear intensity by 9–10 % and 25–35 times, respectively, has been observed.

It is established that a polymer composite material, based on aromatic polyamide and terlon, features a better level of tribological properties, as compared with the initial polymer at the frictional engagement with steel. Thus, its coefficient of friction and the linear wear intensity feature 25 % and 20–40 % lower values respectively than for the initial polymer.

It has been found that the introduction of terlon fiber, based on aromatic polyamide, allows expanding the terminal conditions of friction and sealing pairs. Thus, friction pairs, wherein the aromatic polyamide is used, can work in normal operation mode and to maintain a high level of reliability and longevity over the entire studied load range at sliding speeds of no more than 1.5 m/s, and friction pairs with a composite of 85 % phenylon + 15 % terlon - at sliding speeds of up to 2.0 m/s.

The basic physical and mechanical properties of the materials under study have been determined, and it has been established that the introduction of fiber into the composition of phenylone reduces their level by 10–15 %, which is a consequence of the presence of structural micro- and macrodefects caused by an insufficient level of uniformity of fiber distribution in the polymer matrix.

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ПОЛІМЕРНІ КОМПОЗИТИ НА ОСНОВІ АРОМАТИЧНОГО ПОЛІАМІДУ Й АРАМІДНОГО ВОЛОКНА ДЛЯ ВАЖКОНАВАНТАЖЕНИХ ВУЗЛІВ ТЕРТЯ ТА ГЕРМЕТИЗАЦІЇ

Анотація. *У роботі обґрунтовано вибір полімеру та наповнювача на основі ароматичного поліаміду для створення матеріалу пар тертя, які працюють у важконавантажених вузлах тертя, і герметизації сучасних машин і механізмів. Досліджено вплив навантаження та швидкості ковзання на трибологічні властивості вихідного ароматичного поліаміду та наповненого 15 % арамідного волокна. У результаті трибологічних досліджень визначено граничні умови експлуатації виробів із вихідного ароматичного поліаміду та композиту на його основі, наповненого арамідним волокном. Визначено основні фізико-механічні властивості досліджених матеріалів і встановлено, що введення арамідного волокна в ароматичний поліамід сприяє 10–15 % зниженню рівня його властивостей через виникнення мікро- та макродефектів його структури, які є наслідком недостатньо якісного рівня розподілу волокна в полімерній матриці.*

Ключові слова: *ароматичний поліамід, арамідне волокно, триботехнічні властивості, коефіцієнт тертя, інтенсивність лінійного зношування, міцність.*