Secondary Thermoplastic Modified Wood-Polymer Composite with Increased Technological, Mechanical and Dielectric Properties

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Abstract: The article describes the development of new formulations of wood-polymer composites with a modified polymer matrix and the study of their technological, mechanical and dielectric properties. The research aim is to create a new composition of wood-polymer composites based on secondary raw materials with an improved set of technological, mechanical and dielectric properties, namely, the modification of the polymer matrix and the development of a new technology for its production. The optimal composition of the polymer matrix and modification influence of the wood-polymer composites polymer matrix on the physical, mechanical and dielectric properties of the samples are determined. Strength, impact resistance, abrasion, mechanical and dielectric properties are studied. A description of the climatic influences on the wood-polymer composites including the influence of moisture and temperature is given.

Keywords: Composite, wood, secondary raw material, polyethylene, thermoplastic polyurethane, modification, technological, mechanical, dielectric properties.

I. INTRODUCTION

Today, wood-polymer composites (WPC) are widespread in the USA, Canada, Singapore, China and are actively gaining popularity in Europe. The spectrum of their application is very diverse. Starting with the decoration of suburban real estate such as the interior design of houses, the construction of terraces, balconies, gazebos, park benches and ending with the decoration of floors and walls of office buildings and retail premises [1]. Productions that use in their technological process secondary raw materials that are difficult to dispose of or disposal is economically impractical are developing rapidly [2,3]. Interest in WPC is also due to the possibility of disposal of waste from the woodworking industry. WPC is a material that not only replaces wood, but also solves an environmental problem [4-6].

In the development of polymer composite materials (PCM), the properties of WPC are mainly improved regarding the change in the characteristics of the polymer and its adhesion to wood [7-9]. However, wood occupies at least 30% of the total weight of PCM, its properties cannot be neglected. Therefore, it is necessary to improve the properties of WPC also by modifying the wood filler. This leads to a significant decrease in the hygroscopicity and formability of wood materials and an increase in their biostability [10-12].

The use of secondary raw materials in the manufacture of composite materials is a trend throughout the world [13]. Cheap raw materials inspire manufacturers to expand the range of products. WPC, which contains difficult-to-process waste [14,15], is gaining more and more popularity. But the larger the list of materials used in the manufacture of WPC, the more difficult it is to investigate their effect on the composition, which should solve the problems and shortcomings of previously proposed products [16-18].

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The study of the destruction of products under the influence of light and weather factors and during operation forces manufacturers to modify composites. Considering the possible destruction of the product, its installation and processing are complicated reducing the service life.

In order to improve the physical, mechanical and operational characteristics of the material, a modification of the polymer matrix is proposed.

The main use of wood veneer is as a replacement for solid wood and conventional wood-based materials in the manufacturing of a broad range of industrial and household products. WPC products are not exposed to moisture, do not require additional painting and are processed by traditional methods of processing thermoplastic materials [17].

The use of wood-polymer composite products is widespread nowadays. The range of products and methods of their use is constantly growing. The main problem of wood-polymer composite products is water absorption. Seasonal changes in weather conditions, temperature fluctuations, natural precipitation – all this is the reason for the generation of moisture in the wood-polymer composite. The property of acquiring moisture is in the polyolefin matrix due to its porous structure, and accordingly, the wood filler also absorbs it [19,20]. Manufacturers try to solve this problem to a greater extent at the expense of either the shape of the product, or at the expense of tools and devices during installation, which leads to an increase in the cost of the finished product.

Another solution to this problem is the addition of various additives to the composition to modify both the wood filler and the polymer matrix in order to improve physical and mechanical properties [21-23].

A decrease in the price of products has a negative effect on the quality of the finished product [24]. Manufacturers use cheap secondary raw materials to reduce the cost of production without studying the impact of one or another impurity, both in the polymer matrix and in the filler.

The expediency of this direction is explained not only by the simplicity of solving the technical problem, but also by reducing the cost of production without significant loss of product quality. There are currently few studies regarding the protection of the wood filler due to the modification of the polymer matrix.

The purpose of the research presented in the article is to create a new composition of WPC with an improved set of properties, namely, the modification of the polymer matrix and the development of a new technology for its production.

To achieve the set goal, it was necessary:

1. To determine the optimal composition of the WPC polymer matrix with increased physical and mechanical properties.
2. Investigate the physical, mechanical and operational properties of polyolefin and modified polyolefin matrix.
3. Investigate the influence of the modification of the WPC polymer matrix on the physical and mechanical properties of the samples.
4. Investigate the operational properties of the WPC with a modified polymer matrix and compare it with industrial and previously proposed samples.

II. MATERIALS AND METHODS

A. Materials

The objects of research are: secondary polyethylene, secondary thermoplastic polyurethane, Styrofoam, petroleum-polymer resin, chipboard waste, microcalcite and gasoline.

B. Methods

1. Sample Preparation

Preparation of WPC is carried out as follows: a solvent (gasoline) is added to the petroleum-polymer resin and thoroughly mixed to a homogeneous solution (intermediate modifying mixture) for 10-15 minutes; the next stage is the addition of polystyrene foam waste, which is dissolved in the intermediate modifying mixture by stirring for 10 minutes with obtaining a modifying mixture. Wood filler for impregnation is added to the resulting modifying mixture and mixed for 5-10 minutes. until the mixture is completely homogenized. Microcalcite is added to the impregnated homogenized wood filler and mixed for 5 min. The homogenized modifying mixture is dried in a laboratory dryer for 1 hour at a temperature of 70 °C, or left for 12 hours at room temperature in the air.

During the stage of impregnation with the liquid phase of the wood filler and drying, 12-15% of the excess solvent evaporated. The dried filler is mixed with secondary polymer raw materials (polyethylene (PE) and polyethylene and thermoplastic polyurethane (PE +TPU)) and produced WPC by the extrusion method on an extruder with a ratio of length to diameter (L/D) of 40, a screw with a diameter of 30 mm at a speed rotation 90 rpm. During the entire period, the temperature is maintained in the range of 190-195 °C.

2. Characterization

Determination of the density of the samples ρ is carried out in accordance with ASTM-D-792. Determination of impact toughness (ISO 180:2000), ultimate bending stress σ (ISO 178:2010), Brinell hardness H_b (ISO 2039-2) and impact strength a (ISO 6272-1:2002) of the samples is carried out in accordance with international standards. Water absorption W is determined in accordance with ASTM-D-570 [25], wear resistance and shrinkage are determined in accordance with DIN 53 516, determination of changes in linear dimensions during heating in accordance with ISO 11501:1995.

The dielectric constant C is determined according to DIN 53458. The tangent of the dielectric loss angle $tg\delta$ is determined according to DIN 53458-1965-03. Electrical resistance is determined according to ISO 3915:2022. The retention of static charge is determined according to DSTU 12.1018-86.

For statistical analysis, data normality was assessed by Tukey's multiple comparison post hoc test using ANOVA. The parameters used in the ANOVA analysis had a significance value of $p < 0.05$ at the 95% confidence level.

III. RESULTS AND DISCUSSION

A study of the physical and mechanical properties of polymer matrix samples with different amounts of TPU modifying additive is carried out, the polymer matrix contains only PE in its composition. The amount of TPU varied from 3% to 20%. The results of physical and mechanical tests of the examined samples of the TPU polymer matrix are shown in Table **1**.

Figure **1** shows the dependence of impact toughness on the TPU content in the polymer matrix.

Figure **2** shows the dependence of static bending on the TPU content in the polymer matrix.

Table 1: Composition and Physical and Mechanical Parameters of the TPU Polymer Matrix

Composition	PE, %	TPU, %	a, kJ/cm^2	σ, MPa	Water absorption, %
1	100	-	11.57	5.54	7.75
2	97	3	10.33	6.98	0.43
3	95	5	11.8	8.84	3.15
4	93	7	12.36	9.61	2.9
5	90	10	13.86	10.79	2.45
6	85	15	10.42	9.83	2.45
7	80	20	8.73	6.62	2.52

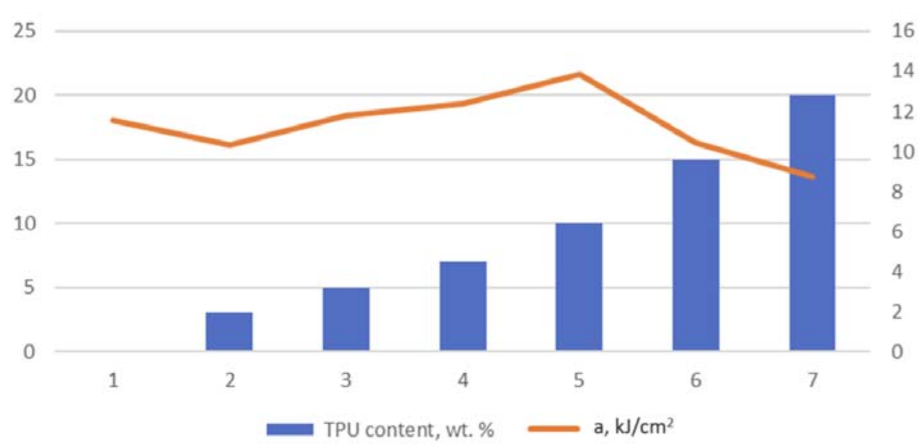

Figure 1: Dependence of impact toughness on the TPU content in the polymer matrix.

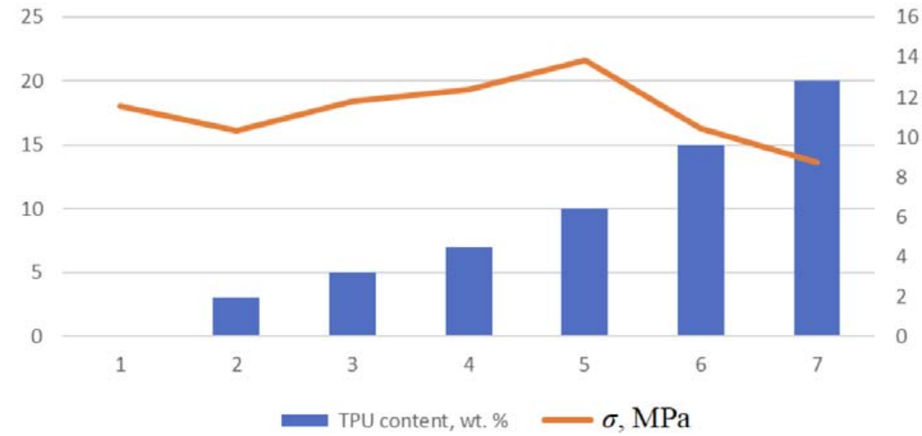

Figure 2: Dependence of static bending on the content of thermoplastic polyurethane in the polymer matrix.

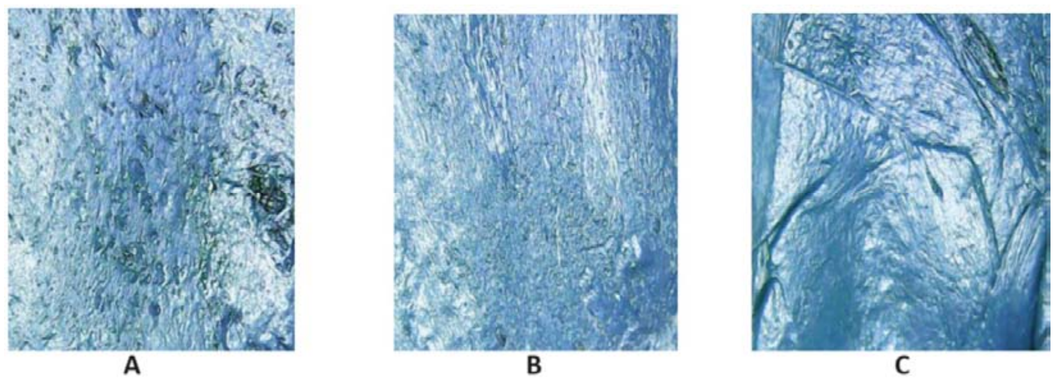

Figure 3: Photos of the surface of the polymer matrix in different compositions: **A** - the surface of the polymer matrix of composition 1; **B** - the surface of the polymer matrix of composition 3; **C** - the surface of the polymer matrix of composition 5.

Figures **1** and **2** show that the sample of the polymer matrix, which has 10% TPU in its composition, has the highest indicators of impact toughness and static bending.

Figure **3** shows photographs of the surface of the unmodified polymer matrix under a microscope in various compositions.

Photographs of the polymer matrix prove that the addition of TPU to the polyolefin polymer matrix modifies it by reducing the porosity of the composition.

On the basis of the conducted research, new compositions of DPC were developed, which are listed in Table **2**.

Tests of physical and mechanical properties of samples from new compositions of WPC with modified polymer matrices were carried out. The results of these tests are given in Table **3**.

As a result of the analysis of the data in the Table **3**, it can be seen that there is a parabolic dependence of the indicators of impact toughness and destructive

Table 2: Composition of WPC with Modified Polymer Matrices

Composition	Secon-dary PE, %	Secon-dary TPU, %	Petroleum polymer resin, g	Solvent gasoline B-70, g	Styrofoam waste, g	Furniture production waste, g	Micro-calcite, g
1	97	3	5	15	5	25	3
2	95	5	5	15	5	25	3
3	93	7	5	15	5	25	3
4	90	10	5	15	5	25	3
5	85	15	5	15	5	25	3
6	80	20	5	15	5	25	3

Table 3: Physical and Mechanical Properties of WPC Samples with a Modified Polymer Matrix

Composite	ρ, g/cm^3	a, kJ/cm^2	σ, MPa	H_b	Strength upon impact
1	1.079	9.03	9.44	27.3	h=25 a dent h=30 crushed
2	1.184	9.32	9.73	29.9	h=30 a dent h=35 crushed
3	1.188	9.68	10.2	32.6	h=30 a dent h=35 crushed
4	1.191	10.06	12.37	38.4	h=30 a dent h=35 crushed
5	1.22	8.76	10.26	39.1	h=30 a dent h=35 crushed
6	1.25	8.44	9.89	40.7	h=30 a dent h=35 crushed

stress during bending of the polymer matrix with an increase in the proportion of TPU, with a TPU waste content of more than 10%, a sharp decrease in these indicators is observed. All WPCs containing TPU waste showed a decrease in water absorption compared to the composite without TPU waste. The presence of TPU waste in the polymer matrix of WPC reduces water absorption by more than 2.5 times compared to WPC, the polymer matrix of which does not contain TPU. The effect of reducing water absorption is possibly related to the inclusion of the TPU polymer matrix, due to its better fluidity and compatibility.

When the content of TPU waste in the polymer matrix increases, the impact toughness, bending stress, and Brinell hardness of samples in the polymer matrix increase with the content of TPU waste from 3 to 20%.

Figure **4** shows the dependence of impact strength on the content of TPU in WPC.

It can be seen from the diagram that when the TPU content in the WPC is more than 10%, there is a sharp decrease in the impact strength indicators.

Figure **5** shows the dependence of static bending on the TPU content in WPC.

In Figure **5**, it can be seen that the highest indicators of static bending have a sample of WPC, which has 10% TPU in its composition.

The results of the operational tests of the tested samples of WPC are shown in Table **4**.

As a result of the data analysis in Table **4**, it can be seen that when the content of TPU waste in the polymer matrix increases from 3% to 10%, water

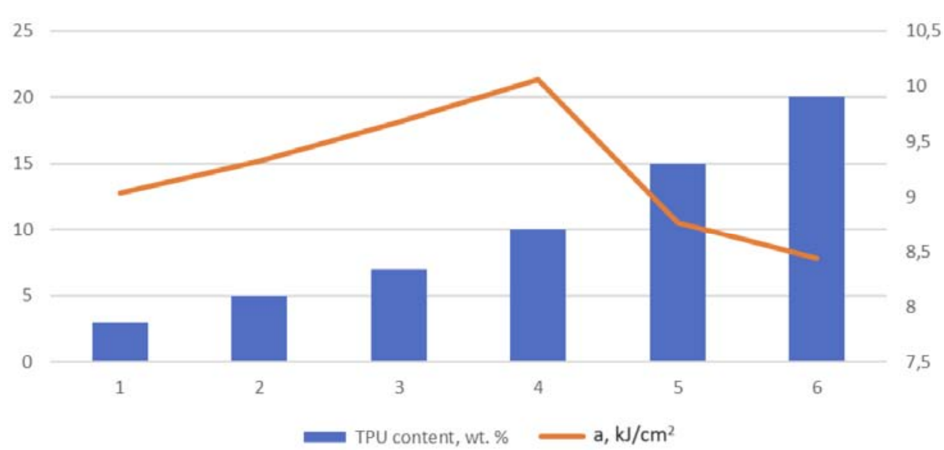

Figure 4: Dependence of impact strength on TPU content in WPC.

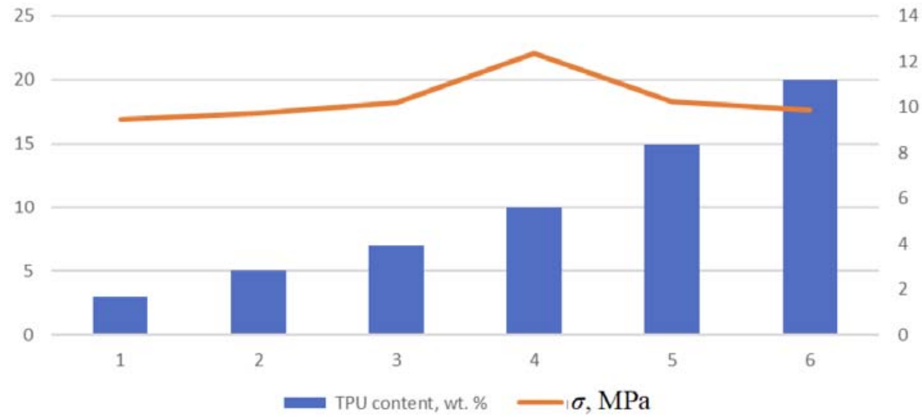

Figure 5: Dependence of static bending on TPU content in WPC.

Table 4: Operational Properties of Wood-Polymer Composites

Composite	Water absorption *W*, %	Wear resistance, m	Wear resistance, V	Resize in an interval, +30^{0}C;+80 ^{0}C	Resize -15 ^{0}C, %
1	+2,64	0,082	0,281	not detected	not detected
2	+2,51	0,078	0,277	not detected	not detected
3	+2,38	0,054	0,274	not detected	not detected
4	+2,2	0,0312	0,272	not detected	not detected
5	+3,6	0,049	0,29	not detected	not detected
6	+5,2	0,051	0,294	not detected	not detected

absorption decreases and wear resistance improves, but when the content of TPU waste exceeds 10% in the polymer matrix, the deterioration of these properties is revealed.

Figure **6** shows the dependence of water absorption on the TPU content in WPC.

Figure **6** shows that the lowest values of water absorption have a sample of WPC, which contains 10% TPU in the composition of the polymer matrix. At the same time, the effect of reducing the water absorption of WPC decreases in proportion to the proportion of TPU in the polymer matrix.

Figure **7** shows the dependence of wear resistance on the TPU content in the polymer matrix of the WPC.

Figure **7** shows that the best indicator of wear resistance has a sample of WPC, which also contains

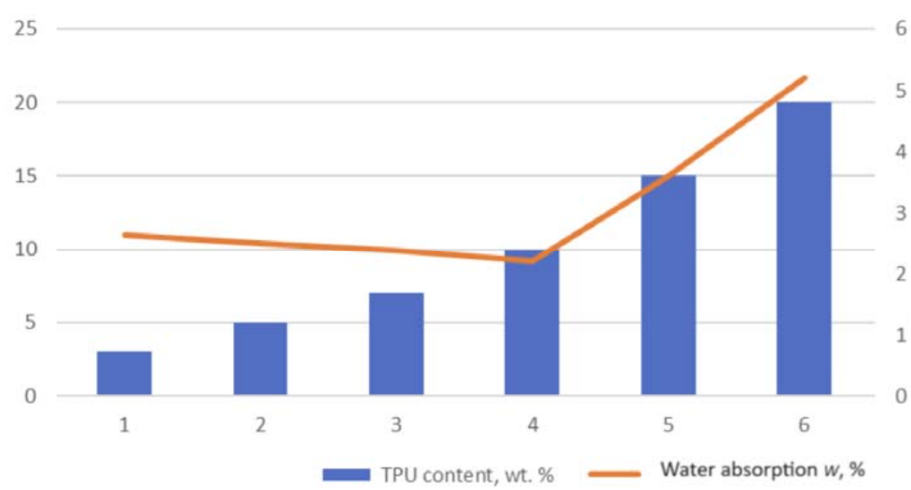

Figure 6: Dependence of water absorption on TPU content in WPC.

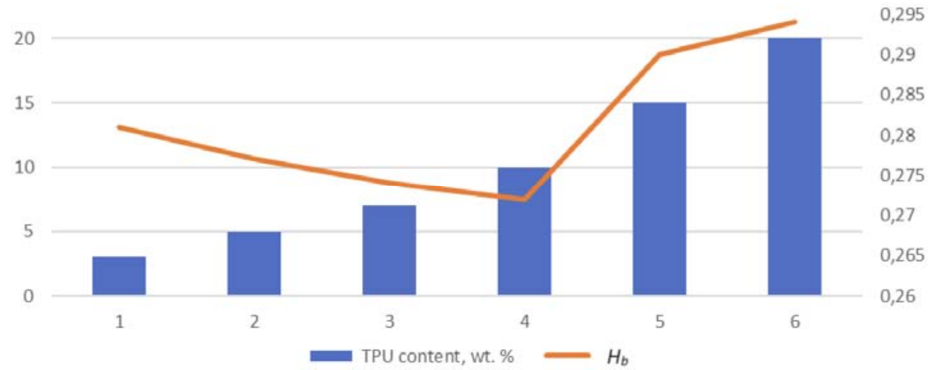

Figure 7: Dependence of wear resistance on the content of thermoplastic polyurethane in the polymer matrix of the WPC.

10% TPU in the composition of the polymer matrix. From above, an increase in the wear resistance of WPC with a TPU content of more than 10% is observed. Change in the dimensions of the WPC samples in the temperature range of +30 °C; +80 °C and -15 °C were not detected.

A comparative analysis of Figures **8** shows that the *tgδ* of the proposed samples of WPC, which is reduced by 17% with increasing temperature. The results of WPC samples tests for the ability to hold a static charge and dielectric constant *C* are shown in Table **5**.

Comparative operational tests of the developed WPC and industrial WPC were carried out. The results of the comparison of operational tests of the proposed and industrial samples of WPC are shown in Table **6**.

Figure **9** shows photographs of the surface of the WPC with a modified polymer matrix under a microscope in different compositions.

The analysis of the results of a complex of experimental studies showed that the investigated quality indicators of WPC samples have improved

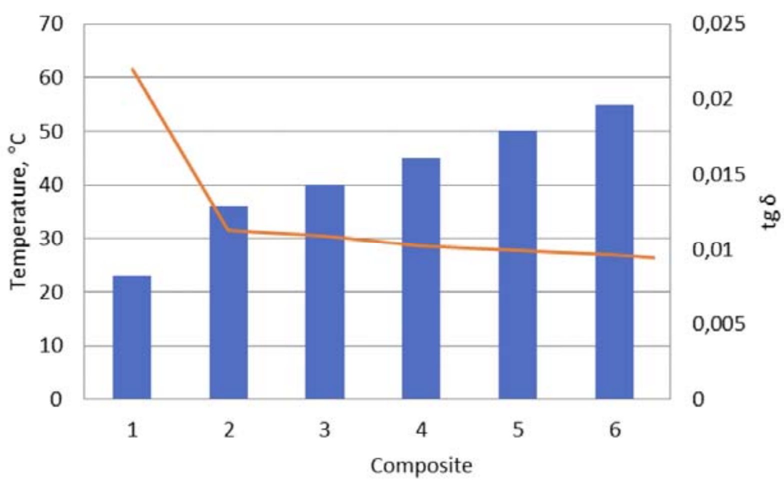

Figure 8: The influence of temperature on the dielectric loss angle tangent *tgδ* of the developed WPC samples.

Table 5: Dependencies of the Dielectric Properties on the Temperature of WPC Samples

Composite	Average value of static charge	Average value *C*, pF	Average value *tgδ*
1	Not found	1.53	0.0373
2	Not found	1.63	0.0435
3	Not found	1.63	0.0193
4	Not found	1.63	0.0172
5	Not found	1.63	0.0237
6	Not found	1.50	0.0195

Table 6: Comparative Characteristics of the Main Indicators of the Developed WPC and Industrial Samples of WPC

Composite	Water absorption *W*, %	Wear resistance, m	*a*, kJ/cm^2	*σ*, MPa	Resize in an interval, +30 °C;+80 °C	Strength upon impact
1	+2.64	0.082	9.03	9.44	not detected	h=25 dent h=30 crushed
2	+2.51	0.078	9.32	9.73	not detected	h=30 dent h=35 crushed
3	+2.38	0.054	9.68	10.2	not detected	h=30 dent h=35 crushed
4	+2.2	0.0312	10.06	12.37	not detected	h=30 dent h=35 crushed
5	+6.2	0.142	8.09	14.6	not detected	h=15 dent h=20 fault
6	+5.4	0.113	6.98	15.88	not detected	h=15 dent h=20 fault
Commercial sample of WPC (Ukraine)	+10.8	0.309	2.74	18.2	length +1.98 thickness +6.78	h=10 fault h=15 fault
Commercial sample of WPC (Czech Republic)	+8.6	0.409	7.55	9.14	length +2.98 thickness +6.78	h=12 fault h=18 fault
Sample OSB (Czech Republic)	+44.18	0.326	11.03	9.04	length +3.86 thickness +10.3 width +4.18	h=10 fault
Chipboard sample laminated (Czech Republic)	+42.15	0.312	11.52	14.7	length +2.28 thickness +7.7 width +6.29	h=10 fault

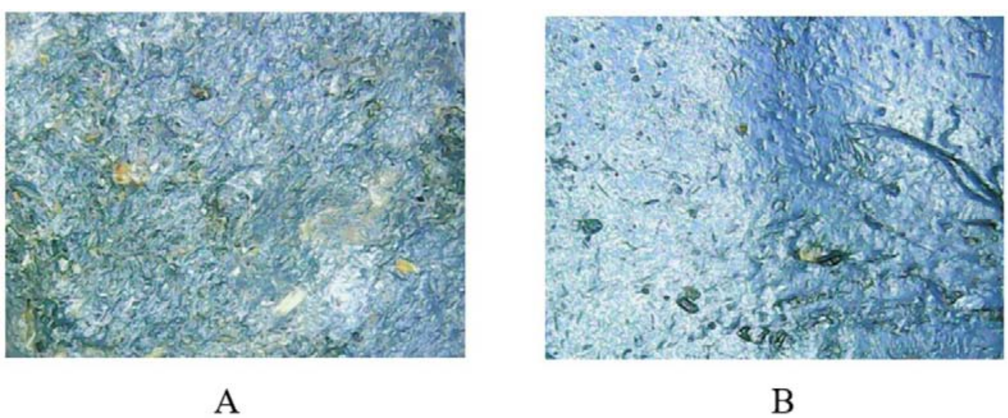

Figure 9: Photographs of the surface of WPC with different TPU content: **A** - the surface of the WPC with a TPU content of 5%; **B** - the surface of the WPC with a TPU content of 10 %.

water absorption compared to previously proposed samples by almost 50 %, wear resistance by 300 %, impact toughness by 5.4 %, impact strength by 29 %, but static bending worsened. This is due to the fact that thermoplastic polyurethane waste is added to the polyethylene polymer matrix. The porous structure of polyethylene becomes more filled and elastic thanks to the modification.

CONCLUSIONS

Thus, the analysis of the scientific and production experience accumulated so far indicates the advantages of the proposed composition of the modified polymer matrix in order to obtain products with increased technological, mechanical and dielectric properties.

The research and development of the processes for obtaining WPC, the polymer matrix of which is modified with TPU waste, are given. The influence of the TPU waste content on the physical, technological, mechanical and dielectric properties characteristics of the samples is investigated. The optimal composition of the polymer matrix and modification influence of the wood-polymer composites polymer matrix on the physical, mechanical and dielectric properties of the samples are determined: improved water absorption compared to previously proposed samples by almost 50%, wear resistance by 300%, impact toughness by 5.4%, impact strength by 29%, but static bending worsened. It can be seen that all the proposed WPC samples do not have the ability to accumulate static electric charge, which indicates the integrity of the material without changing the operational properties The parameters of production of products by the extrusion method are defined.

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