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Reuse of polyester-glass laminate waste in polymer composites

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ABSTRACT

Purpose: of this paper is to develop a new generation of polymer composite materials that would ensure the use of residual and serious environmental problems of polyester-glass laminate waste.

Design/methodology/approach: The glass reinforced polyester waste was ground and added to produce new composites. Thermoplastic - high impact polystyrene was selected for the composite matrix. Composites containing 10, 20, 30% by weight of the filler of polyester-glass laminate powder were made. The process of extrusion and subsequent injection was used to prepare the test samples. The influence of the filler on selected properties of composites was evaluated. The physical properties of the filler as well as the processing properties of the mixture as well as the mechanical properties - impact strength and tensile strength of the obtained composites were investigated.

Findings: A decrease in tensile strength and impact strength was observed along with an increase in the amount of filler.

Research limitations/implications: It would be interesting to carry out further analyzes, in particular with a higher volume fraction of the filler or with a different composite structure, e.g. using PVC as a matrix. The developed research topic is a good material for the preparation of publications of a practical and scientific nature, especially useful in the research and industrial environment.

Practical implications: The shredded glass-polyester waste can be used as a filler of polystyrene, however, the resulting composite could be used to produce parts with slightly less responsible functions such as artificial jewelery or toy elements.

Originality/value: Obtained results are a new solution a global waste management solution for glass reinforced polyester waste, which may contribute to the sustainable development of the composite materials industry through the partial utilization of waste composites with a duroplastic matrix.

Keywords: Mechanical properties, Polymeric composites materials, Plastic forming, Filler of polyester-glass laminate powder, High impact polystyrene

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PROPERTIES

1. Introduction

The ecological problem of environmental pollution with post-production waste containing polymer materials is still present [1-3], despite many attempts to manage waste from polyester-glass laminates and has not been solved so far [4-14]. Many methods of recycling fibrous polymer composites are developed, including thermal recycling, pyrolysis to obtain fuel, etc [15-17]. However, the most common method of waste management is still landfilling, the major disadvantage of which is the large area of waste disposal of polymeric materials and their composites [18]. The second frequently used method of disposing of waste is its incineration. Unfortunately, a large part of the material did not burn due to the large amount of fibrous reinforcement or mineral fillers [19]. Recently, an increasingly common solution is the reuse of laminate waste with a duroplastic matrix, i.e. material recycling, the main purpose of which is to transform waste material into a product of utility value. The shredded waste can be successfully used as a polymer matrix filler, replacing part of the fibrous or powder filler and improving the mechanical characteristics or other properties (e.g. wear resistance) of the matrix used. Waste processed in this way is used experimentally in the furniture industry (for the production of composite panels), as an additive to asphalt, concrete, polyester molding compounds, polymer concrete and in many other products, in which the addition of recyclate does not significantly reduce their functional properties [20-26]. Therefore, the attempt to search for a material that could be used as a matrix of composites with the addition of the above-mentioned particles of polyester-glass laminates seems to be right. Given the long-term trend of increasing polymer prices, much attention has been paid to adding fillers to reduce costs in the plastics industry [27-31]. These fillers include all kinds of fibers, such as glass, carbon and cellulose, which have the highest priority as biodegradable fillers for commercial thermoplastics. The choice of polystyrene for the matrix of composites was quite promising due to its current application and properties (HIPS is a versatile material with a huge number of applications. It is extremely popular in the construction industry, in the manufacturing industry. High-impact polystyrene panels are often used for interior design and kitchen finishes and bathrooms, as well as in the advertising and industrial industry for the production of packaging and pallets for food products and electronic components, and for the production of waste containers or covers for solarium beds) [32,33]. They can also be used as thermal insulation for the production of heat exchangers [34,35].

The inspiration for taking up a specific topic of work was to find a way to manage post-production waste for the INBEHA company from Mikołów. The INBEHA company is a distributor of plastic plates and profiles and deals with the production of structural elements, parts of machines and devices. During CNC machining of laminate boards, a by-product in the form of dust is created, which is a big problem for the company, as its annual disposal is a very high cost for the owner. It should also be noted that the waste is not pure laminate dust generated in the cutting process, but powder with an admixture of impurities in the production hall. An attempt was made to use the waste as a filler for a high-impact thermoplastic, so that the properties of the matrix still allow the resulting composite to be used for specific applications. The waste was added in various amounts to the polymer matrix, initially by mixing the granulate with the filler in an extrusion process, and then from the finished mixture by the injection process, samples in the shape of standard paddles were made. The samples prepared in this way were subjected to strength tests.

2. Experiments

2.1. Materials and methods

As part of the experimental work, a polymer composite blend was prepared by extrusion, and then test samples were made in the injection process. Tests were carried out to assess the quality of the recyclate used as a filler, among others The density, grain distribution and decomposition temperature were determined, and then the influence of the filler addition on the mechanical characteristics of the composites made was determined (the tensile strength and impact strength tests were carried out using the Charpy method).

The following were used to produce the test samples:

- for the matrix of composites: SYNTHOS PS HI 552M [36] granules,
- as a filler: waste in the form of dust resulting from cutting laminate (polyester-glass) boards at Inbeha.

As part of the preliminary tests related to the determination of the physical properties of the filler particles, the following were performed: density testing, shape analysis and particle size distribution. The density measurement was performed on an automatic gas pycnometer – Micromeritics Accupyc 1340. In order to determine the filler particle size distribution, tests were carried out on the ANALYSETTE 22 MicroTec Plus laser particle size meter, located in the laboratory of the Institute of Engineering and Biomedical Materials, Faculty of

Mechanical Engineering, Silesian University of Technology. The shape of the powder particles can be determined by descriptive and mathematical methods. The descriptive method used in this work is based on the Polish Standard PN-EN ISO 3252:2002. According to this standard, the selected filler was classified according to the shape of its particles based on the analysis of images obtained with the use of a scanning electron microscope (SEM). Before the filler material was introduced into the matrix in the extrusion process, it was subjected to thermogravimetric analysis (TGA) in order to determine the temperature of the onset of decomposition. The test was performed on a thermogravimeter by METTLER TOLEDO in the Łukasiewicz Research Network - Institute for Engineering of Polymer Materials and Dyes, Center of Paints and Plastics in Gliwice with the PN-EN ISO 1133: 2006 standard. The research was carried out using a Zwick/RoellMflow load plastometer with additional equipment and an AS 60/220.X2 analytical balance by Radwag, located in the laboratory of the Institute of Engineering Materials and Biomedical Materials at the Faculty of Mechanical Engineering, Silesian University of Technology. The mass flow rate (MFR) and volume flow rate (MVR) of the manufactured thermoplastic composites were determined. In the melt flow rate test procedure, three weights were prepared with a mass of 3-4 grams of each of the three types of plastics intended for injection. The test conditions were then determined, i.e. test temperature, nominal load, cut-off time interval, which was measured by the piston travel distance or the piston travel time at least three times.

2.2. Procedure for the preparation of research samples

The procedure of preparing the test samples first assumed the extrusion process, as a result of which granules were obtained for making ready-made samples (in this case, "paddles") in the injection process.

Samples of polymer blends granules were prepared using a Göttfert Feinwerktechnik GmBH twin-screw extruder (L:D=10:1). The samples were prepared by making three mixtures with PS HI 552M matrix, differing in the type of fillers (content 5-15% by volume). The most important parameters of the extrusion process for the selected PS are: max. temperature 265°C, screw speed 20-60 RPM. The extrusion process conditions were set so as not to exceed the temperature of the maximum melting peak obtained during the TGA measurements. Increasing the efficiency by increasing the rotational speed of the screw could have an impact on a sharp increase in friction causing an increase in temperature in the extruded mass, therefore the selection of appropriate extrusion parameters was a very important issue due to the need to moisten the fillers by the matrix material. Most of the relevant extrusion parameters are listed in Table 1. The extrudate, after exiting the extruder head, was cooled in air, and then ground on a granulator. The average diameter of a single granule was 4 mm, and the length was approx. 7 mm. The obtained granulate was observed on the SEM microscope.

Figure 1 shows the granulate with a filler in the amounts determined by weight, respectively, 10%, 20% and 30% after the extrusion process.

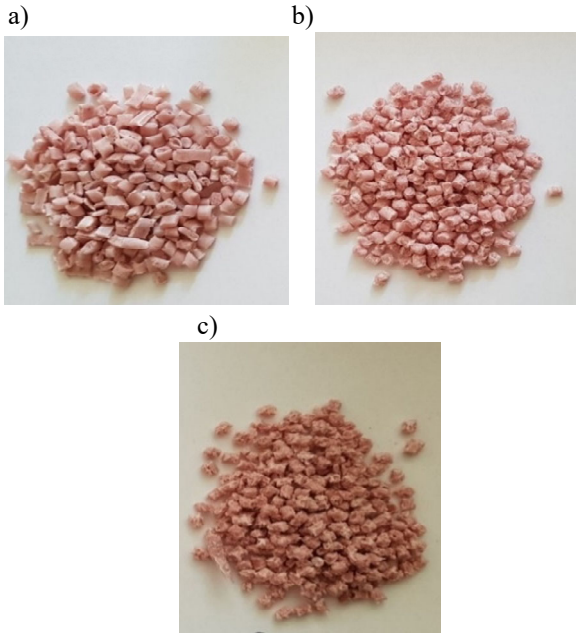

Fig. 1. Composite granulate based on PS: a) with 10% wt. filler, b) with 20% wt. filler, c) with 30% wt. filler

The prepared granules were used to make 10 oar-shaped samples and 20 bar-shaped samples of each mixture in the injection process on an injection molding machine with a set of standardized sample forms Arburg Allrounder 270-210-500 machine (ARBURG GmbH, Loßburg, Germany) at the Central Mining Institute in Katowice. Selected parameters of the injection process are presented in Table 2. The sample (Fig. 2), called a fitting in the standard, is flat and has the shape of a "paddle". The dimensions of a single sample were as follows: thickness 4.0±0.2 mm, measuring part width 100±2 mm, and total length greater than 150 mm. On the other hand, the bar-shaped samples had the following dimensions: thickness 4.0±0.2 mm, width 10±0.2 mm and length equal to 80 mm.

Table 1.
Parameters of the extrusion process

Temperature of individual zones of the screw 1/2/3, °C	Efficiency, $\frac{kg}{h}$	Torque, kpm	Screw rotation, rpm
240 / 230 / 220	3	5	20-60

Table 2.
Parameters of the injection process

Injection pressure, bar	Injection temperature, °C	Mold temperature, °C	Injection time, sec	Cooling time, sec	Pressing time, sec
240	210	40	6.5	25	7

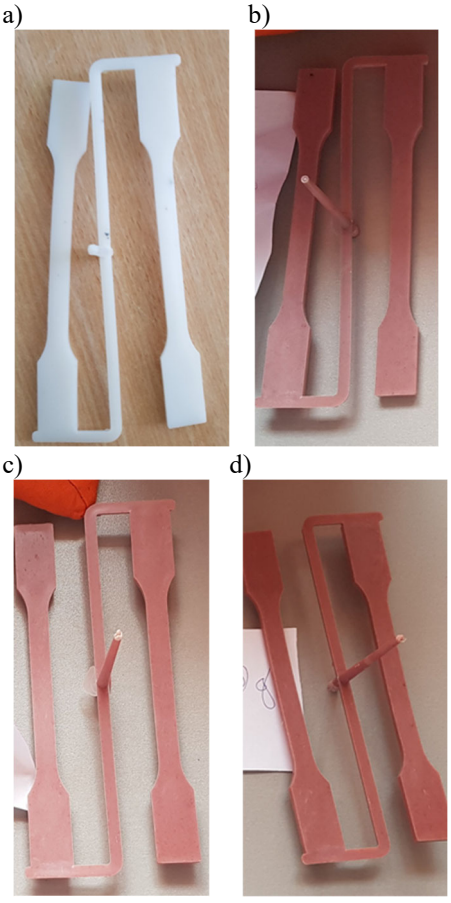

Fig. 2. Photos of samples prepared for tensile strength tests: a) unmodified reference material, b) composites with 10% wt. filler, c) composites with 20% wt. filler, d) composites with 30% wt. filler

The mechanical properties of the tested composites were determined on the ZWICK Z020 universal testing machine. The samples were prepared in accordance with the recommendations of ISO 527-2, the tests were carried out at a tensile speed of 20 mm / min, under normal temperature, humidity and pressure conditions. The impact tests were carried out on the Charpy Hammer by Zwick Roell HIT25P according to the PN-EN ISO 179-1:2010 standard. For testing composites (samples with filler), a pendulum hammer with an initial impact energy of 1 J was used, while for testing the unmodified material, a pendulum hammer with an initial impact energy of 5 J was used.

3. Results of own researches

3.1. Test results for physical properties of fillers

The average density of the powder from the laminate waste obtained on the basis of the tests is 2.12 g/cm^3. The results of the tests of the size of the filler particles (dust of polyester-glass laminate waste) are presented in the graph of the grain size distribution curve and the cumulative particle grain composition curve (Fig. 3) and in the Table 3.

Table 3.
Results of the analysis of the particle size distribution of the fillers

No. sample	D_m, µm	Z, µm	D(4,3), µm	$D_{50\%}$, µm	$D_{90\%}$, µm
1	218.33	3.48	120.2	80.1	287.9
2	162.99	4.58	81.5	43.3	204.3
3	157.78	4.56	75.1	40.4	190.2
4	147.86	4.48	69.3	37.7	175.4
5	138.56	4.38	64.2	35.9	162.8

D_m – modal value, grain diameter that occurs most often,
Z – span – is calculated from the formula Z = (D90%-D10%) / D50%,
D (4,3) – de Broucker mass or volume mean diameter,
D50% and D90% – give the particle size for which 50% and 90% of the particles by volume are below the sizes given, with D50% being the mean particle size.

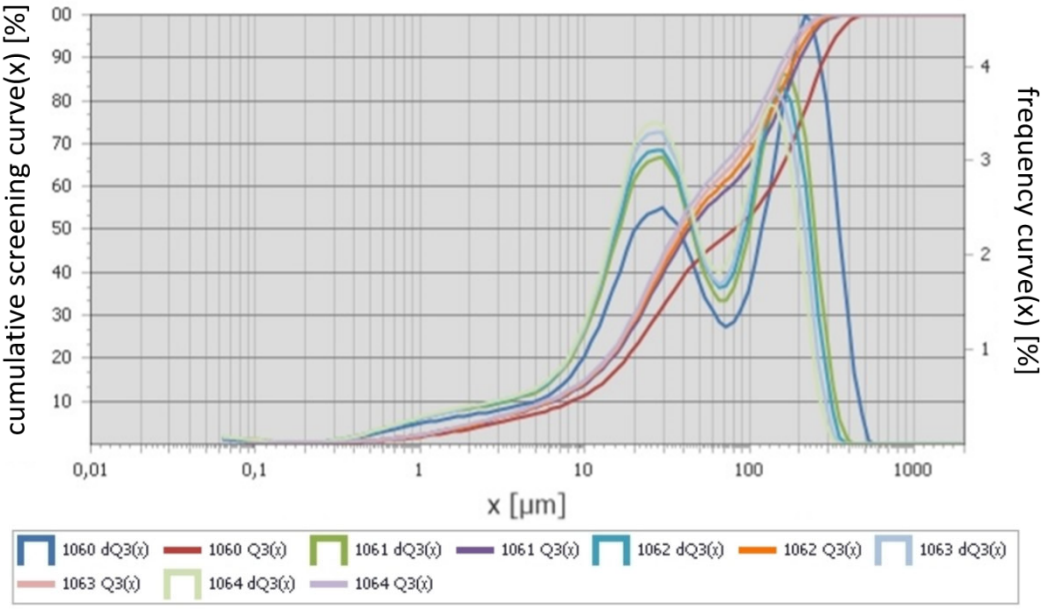

Fig. 3. Curve of particle size distribution and a cumulative curve of particle size distribution of the particles of waste glass-fiber composite

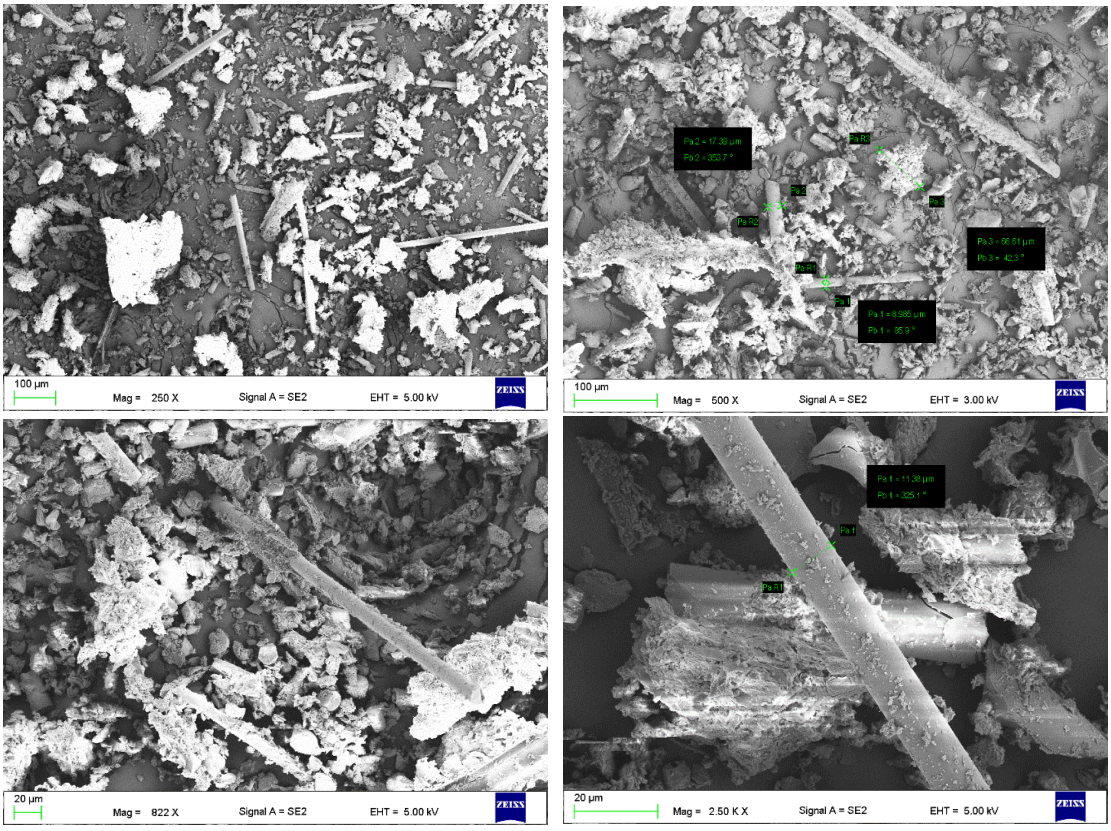

Fig. 4. The morphology of the composite powder filler

The microscopic photos made allowed to classify the selected filler in terms of the shape of its particles. The obtained microscopic images (Fig. 4) and the classification results are shown in the figure. The conducted analysis shows that the laminate powder has an irregular shape with visible elongated fibers characteristic of glass fillers.

Thermogravimetric analysis (testing the temperature of the onset of decomposition) gives the result in the form of a thermogravimetric graph (Fig. 5) showing the dependence of the sample mass as a function of temperature increasing at a constant speed. It can be read from the diagram that the temperature of the beginning of the decomposition of the tested mixture (ground plate of polyester resin with glass fiber) is about 260°C.

The results of the mass melt flow rate (MFR) tests of the produced mixture (granulate) are shown in Table 4.

3.2. Test results of selected mechanical properties of composites

The results of the tensile strength tests for the unmodified material are presented in the form of a line diagram for five tests in the collective diagram (Fig. 6). Similarly, the test results for samples with 10, 20 and 30% by weight of filler were presented in the form of collective line plots (Figs. 7-9). The comparison of the tensile strength at the yield point of the tested samples depending on the proportion of the filler is shown in the bar chart in Figure 10. However, Figure 11 shows a comparison of the strain at break of the tested materials and the weight fraction of the filler.

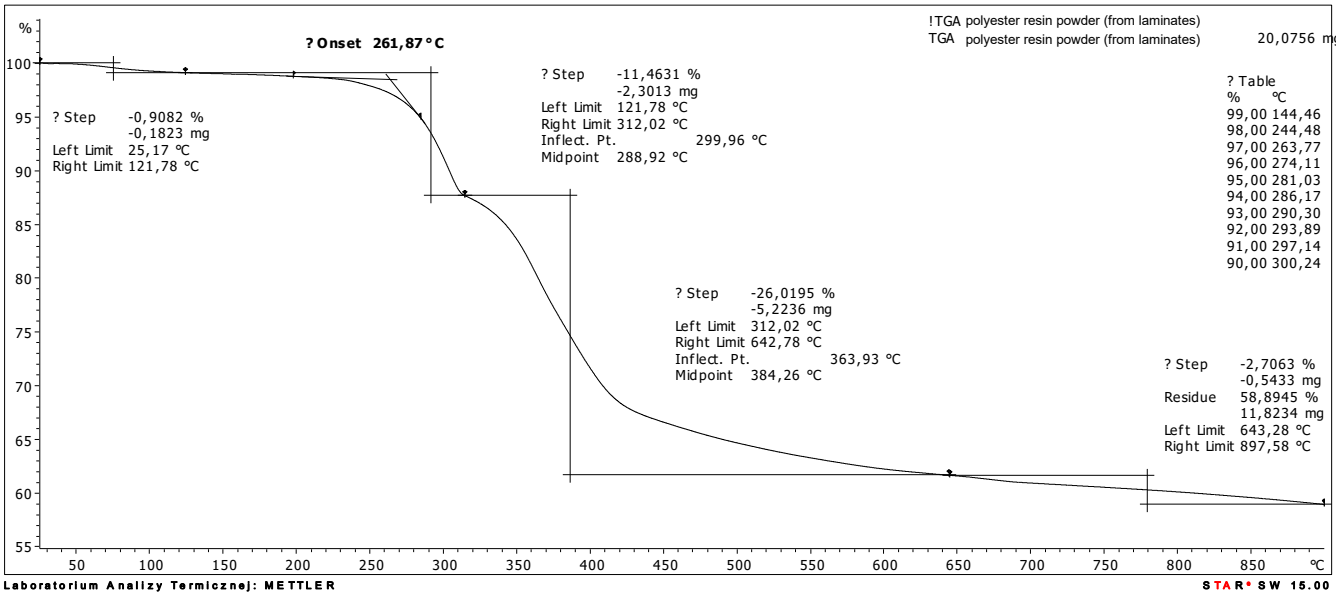

Fig. 5. Graph for Sample Mass vs. Temperature Test: Decomposition/ Thermal Stability

Table 4.
Melt flow rate of the prepared mixture

Material	Conditions		Results	
	Weight, kg	Temperature, °C	MVR, cc/10 min	MFR, g/10 min
PS unmodified	2.16	195	4.8	9-10
PS + 10% wt. of filler			1.30	1.17
PS + 20% wt. of filler			1.18	1.06
PS + 30% wt. of filler			0.88	0.79

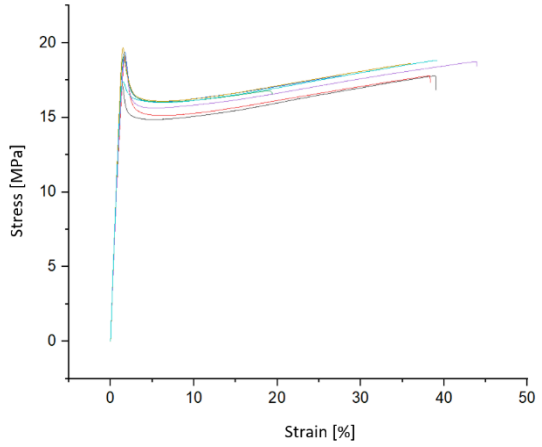

Fig. 6. Stress-strain curve for unmodified PS material

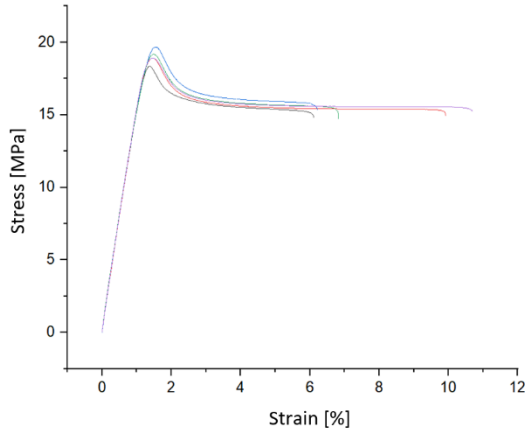

Fig. 7. Stress-strain curve for PS composites with 10% wt. of filler

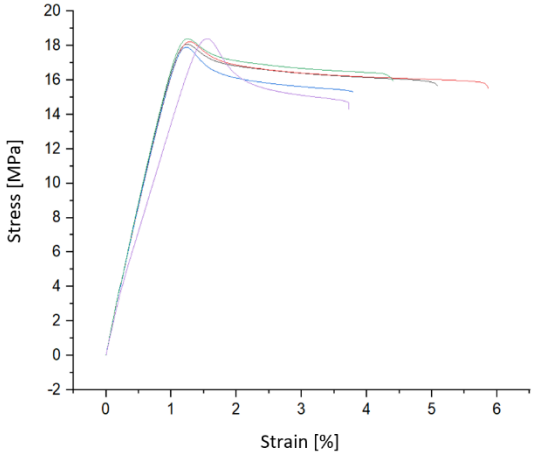

Fig. 8. Stress-strain curve for PS composites with 20% wt. of filler

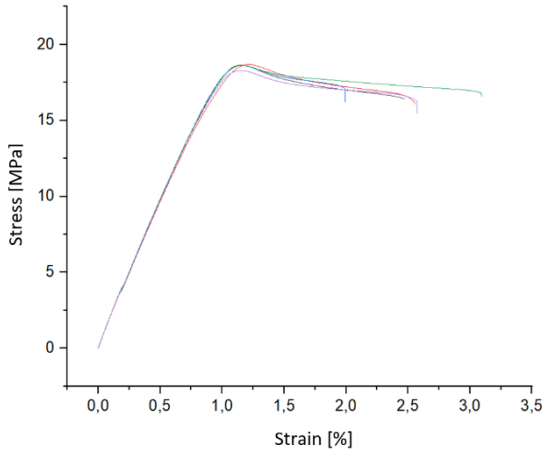

Fig. 9. Stress-strain curve for PS composites with 30% wt. of filler

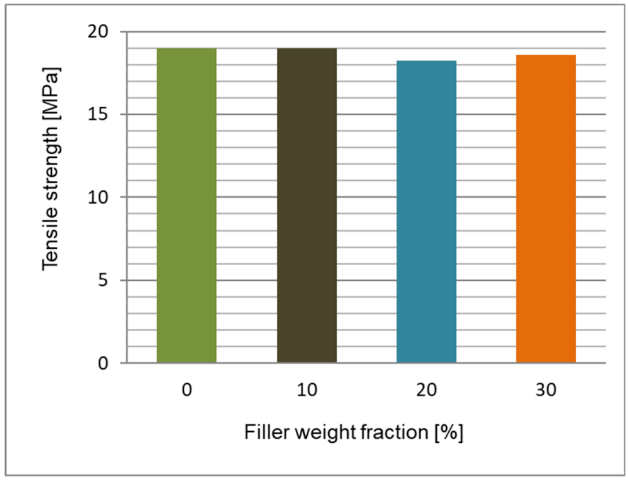

Fig. 10. Graph of the dependence of the tensile strength at the yield point on filler weight fraction

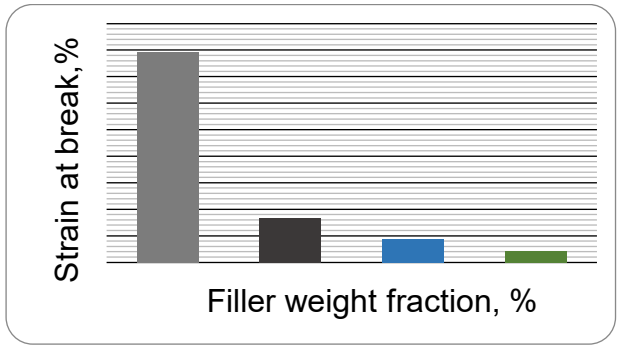

Fig. 11. Graph of the dependence of strain at break on filler weight fraction

The obtained results of strength tests clearly indicate that the stress limits of composite samples do not significantly deteriorate in relation to the original unmodified material (there are no significant differences in the results). Filler is 2.11% in relation to the initial value measured for pure material without fillers, which was 39.67%.

Graphical interpretation of the dependence of impact strength on the weight content. of the filler in the composite is shown in Figure 12.

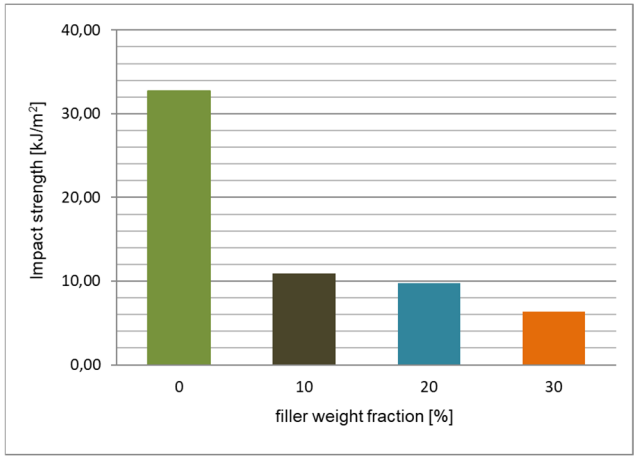

Fig. 12. Effect of filler weight fraction and type on the impact strength

Impact tests show that even the smallest amount of filler deteriorates the impact toughness by more than 3 times in relation to pure granulate. The introduction of the filler with the content of 10, 20 and 30% by weight, respectively, reduced the impact resistance and deformation with a slight change in the tensile strength of all produced samples, which means that the resulting polystyrene-based composite can still be used for the production of parts, however, with slightly less responsible functions, such as imitation jewelry or toy components. At the same time, pure polystyrene has a much lower chemical resistance than polyethylene and for this reason it is unsuitable for the production of packaging for food products containing fats, and perhaps, after comprehensive research, it would turn out that these properties have improved thanks to the introduced filler. The result of the work does not allow for an unequivocal decision to reject or accept the thesis about the possibility of using the tested waste as a filler for the indicated materials, which in turn would allow for partial utilization of composite waste with a duroplastic matrix.

4. Conclusions

The assumption of the work was to weaken pure polystyrene as little as possible by adding to it production waste in the form of contaminated laminate dust taken from the Inbeha company straight from the production hall, so that the waste was as close as possible in composition to the target collected filler. The assumption was that polystyrene with a filler in the form of waste from the laminate would be suitable for the production of food packaging, e.g. yoghurt containers, where there is no need to use an elastic material resistant to bending or stretching.

As can be seen already with the use of 10% by weight of the filler, the samples show lower susceptibility to longitudinal deformation in the tensile test, on average 5 times, which excludes the use of this material for the production of parts exposed to high tensile forces. On the other hand, the force that must be used to exceed the yield point of the material is minimally deteriorated and amounts to approximately 19 MPa. Even 30% of the filler content does not significantly reduce the stresses at the yield point, so the strength of the material does not deteriorate significantly, which has been verified by statistical analysis using the Bartlett and F-Snedecor tests, so the material can be used for the production of parts, however, one should take into account the fact that that destruction under high loads will occur much faster than with pure PS granules without fillers.

The results of the impact tests show that the material is not suitable for the production of dynamically working parts. As little as 10% of the filler content causes a significant, 6-fold reduction in impact resistance.

The conclusion drawn from the study are as follows:

A decrease in tensile strength and impact strength was observed along with an increase in the amount of filler.

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