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Waste from renewable sources as a polyethylene filler

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Abstract. The paper presents research on the impact of three organic fillers from renewable sources, i.e. ground wheat bran, pumpkin seed husks and peanut shells, constituting production waste of the food industry, on selected mechanical and thermal properties of composite injection moldings based on LDPE. The basic strength properties were tested in a static tensile test, hardness and heat deflection temperature, Vicat softening point, and the microscopic structure of the obtained polymer compositions was analysed. The tests were carried out with a different mass content of fillers of 0, 5, 10, 15 and 20% and variable size of filler grains. Samples for testing were obtained in the injection molding process. An increase in the Young's modulus and hardness as well as a decrease in the remaining tested strength properties along with an increase in the filler's share content were found.

1. Introduction

Promoting the use of natural waste in various industries, including polymer processing, is associated with the pressure of global organizations responsible for environment protection, on ecological issues, such as recycling, landfill and waste disposal. Interdisciplinary approach to the subject of polymer processing makes it possible to use materials that until now had no practical application. The increase in the use of natural waste in polymer processing can contribute to reducing the negative impact of this industry on the environment, by accelerating the degradation of polymer components and reducing the consumption of petrochemical polymers.

The properties of filled materials depend not only on the type and geometrical properties of the filler, processed polymer, the presence of additional components, the constructional solution of the plasticizing system, processing conditions [1], but also on the filler content and particle size [2, 3, 4]. For example, smaller particles bond more easily and form a more durable structure [5, 6]. The combination of wood fillers with a thermoplastic polymer gives a light material, having good mechanical strength and suitable for reprocessing [7, 8]. Authors [9] investigating the mechanical properties of a material based on a linear low density polyethylene filled with crumbled pumpkin seed husks found their good suitability as a filler, which may predispose such materials to various applications.

The aim of the study was to determine the effect of powdered fillers obtained from renewable resources, such as wheat bran, pumpkin seed husks and peanut shells on selected mechanical properties of low-density polyethylene (LDPE) filled with these natural fillers. In particular, the study examined the relationships between the basic strength and thermal properties of the produced injection moulded pieces and filler contents.

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2. Experimental

2.1. Material

Test specimens were made of powdered low-density polyethylene (LDPE) Dowlex 2631.10UE manufactured by the DOW Chemical Company (Schkopau, Germany). This polymer is used for producing thin-walled parts by rotational casting and high dimensional accuracy parts by injection molding. Table 1 lists the properties of the tested polymer after the specifications provided by the manufacturer.

The first filler used in the study was wheat bran, i.e. the coat of wheat grains, coming from a local mill. The second natural filler used in the research were pumpkin seed husks obtained from the plant dealing with the purification and sale of pumpkin seeds. The third natural filler used in the research were peanut shells obtained as a result of separating the edible part of the nut.

For a better mixing effect an adhesion promoter in the form of a carbofunctional silane called aminopropyltriethoxysilane was used.

Table 1. Selected properties of Dowlex 2631.10UE.

Property	Value
Density 23^{o}C, kg/m^3	935
Melt flow rate (230 ^{o}C; 2.16 kg), g/10 min	7
Tensile stress at yield, MPa	17.8
Tensile strain at yield, %	419
Shore hardness, ^{o}Sh D	56
HDT temperature, B (0.45 MPa) ^{o}C	52
Vicat softening temperature (A120 (120°C/h 10N), ^{o}C	115
Melting temperature, ^{o}C	124

2.2. Test stand

The tests were performed using the ARBURG ALLROUNDER 320C 500 – 170 single-screw injection molding machines (Loßburg, Germany), provided with a two-cavity mold for manufacturing standard specimens in compliance with ISO 527-1:2012. The injection molding machine has a cylindrical screw with a diameter of 30 mm and the length to diameter ratio of 20. The highest peripheral screw speed equals 42 m/min and its torque is up to 250 Nm. The composite polymer was injected into the mold at the pressure of 100 MPa and the holding pressure equaled 85 MPa.

2.3. Methods

The process of preparing organic fillers covered their fragmentation into a powder and separation of fractions of a given grain size using sieves with mesh sizes of 0.2mm, 0.8mm and 1.2mm. As a result, three fractions with grain sizes comprised in intervals (<0.4mm, 0.4-0.8mm, 0.8-1.2mm) were obtained.

Before starting the injection molding machine, mixtures of low density polyethylene were prepared with organic fillers from renewable sources, i.e. wheat bran, pumpkin seed husks and peanut shells with different mass content of fillers equaling 0, 5, 10, 15 and 20%.

The static tensile tests were performed on produced composite moldings using a standard testing machine, Z010 AllroundLine from Zwick Roell (Ulm, Germany). The Z010 testing machine has the maximum tensile force of up to 10kN and the tensile rate up to 2000mm/min. Static tensile tests were performed using the testing machine on 10 injection moldings in compliance with the ISO 527-1:2012 standard.

Hardness measurements were conducted by Shore method using the ART.13 hardness tester manufactured by Affri System Hardness Testers (Induno Olona, Italy). Hardness measurements were conducted in accordance with the procedure described in the ISO 48-4:2018 standard. Heat deflection temperature (HDT) and Vicat softening point were measured using the CEAST HV3 from INSTRON

CEAST Division. The temperature measurements were made in compliance with the procedures described by two standards: PN-EN ISO 75 (HDT) and PN-EN ISO 306 (Vicat).
The examination of morphology of the specimen cross sections was performed using the Nikon Eclipse LV100ND microscope (Warsaw, Poland), equipped with the DS-U3 camera and NIS-Elements AR 4.20.00 software.

3. Results and discussion

3.1. Strength tests of produced injection moldings

On the basis of the obtained results of strength measurements in the static stretching test, diagrams of relevant relationships were made. An example diagram showing the dependence of the Young's modulus from the content of natural fillers and the size of their grains is shown in Figure 1.
All polymer mixtures filled with organic fillers showed an increase in Young’s Modulus, the highest at 20% by weight of the filler. This shows that the stiffness of the polymer has increased, which is probably due to the limited mobility of polymer chains because of the presence of a natural filler [10]. An important role is also played by the size of grains at the phase boundaries, the LDPE polymer matrix - the surface of the filler grains [11]. The larger the size of the filler grains, the higher the value of the Young’s modulus.

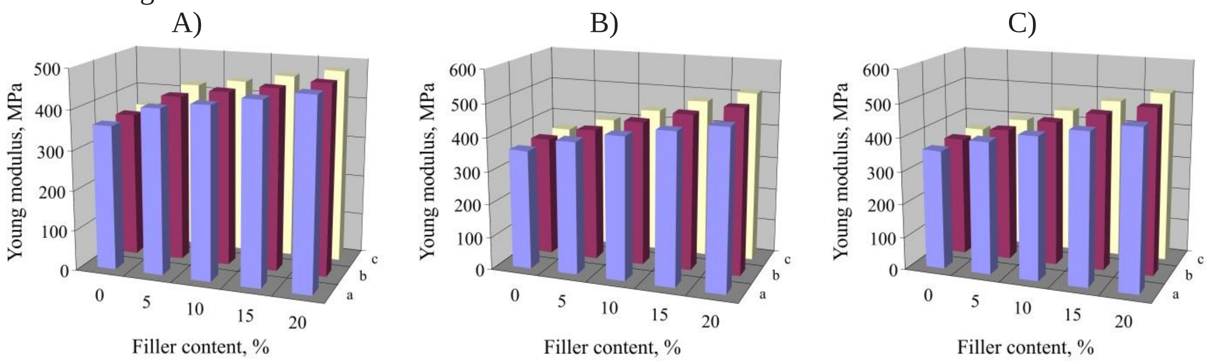

Figure 1. Graph of Young's modulus of low-density polyethylene filled with powdered organic fillers versus various contents and grain sizes: a) grain size from 0 to 0.4 mm, b) from 0.4 to 0.8 mm, c) from 0.8 to 1.2 mm; A) peanut shells, B) pumpkin seed husks, C) wheat bran.

It was also found that the addition of 20% filler causes a decrease in the value of the highest tensile stress (Fig. 2). This is probably due to the matrix being a polyolefin polymer which contributes to the weakening of the adhesion of the hydrophilic organic fillers to the hydrophobic polymer matrix. The structure of most natural fillers favors the increase of water absorption, which results in the variability of mechanical properties [12].

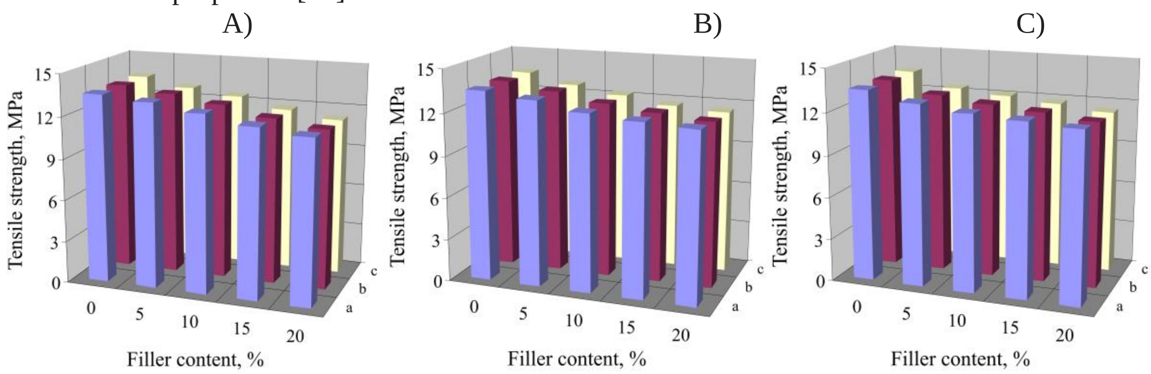

Figure 2. Graph of tensile strength of low-density polyethylene filled with powdered organic fillers versus various contents and grain sizes: markings as in Figure 1.

Analogous tendencies of change of the described property were obtained in other works, in which the effect of various natural fillers on the mechanical properties of polymer compositions such as rice bran, maize starch and potato starch [13] was investigated.

3.2. Hardness of injection moldings

For the purpose of investigations to determine the effect of mass content and various sizes of natural filler grains on the hardness of the obtained injection moldings, Shore D hardness measurements were made. The results of hardness tests are presented in Figure 3 in the form of dependence of hardness expressed in Shore D on mass content and various sizes of filler grains used for testing. When analyzing the graph, it should be noted that the grain sizes used and the mass content of natural fillers significantly affect the hardness of polymer mixtures.

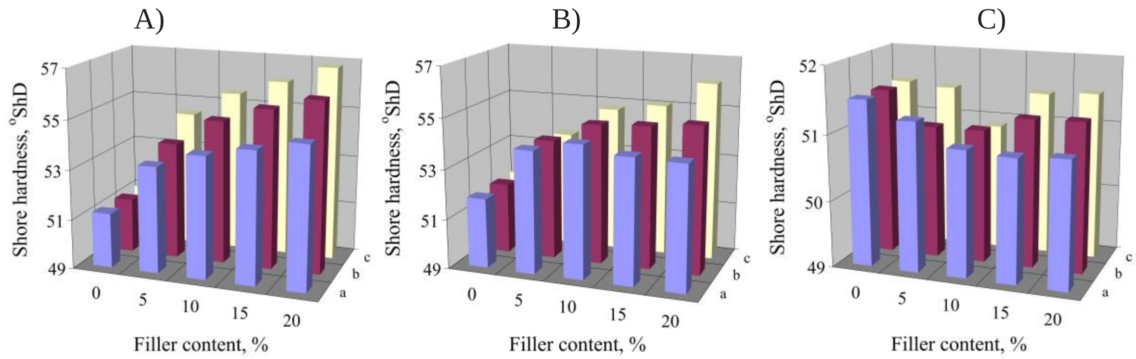

Figure 3. Dependence of hardness of low-density polyethylene filled with powdered natural fillers on their various contents and grain sizes: markings as in Figure 1.

3.3. Vicat softening point and Heat deflection temperature (HDT)

No rule was defined that would describe the effect of the presence of powder fillers on the Vicat softening point (Figure 4A). Temperature changes depend on the type of filler and its interaction with the polymer matrix. In the analyzed case, it is difficult to talk about the influence of the filler on the softening point due to the fact that the percentage values of temperature changes reach a maximum of 1% of the initial value.

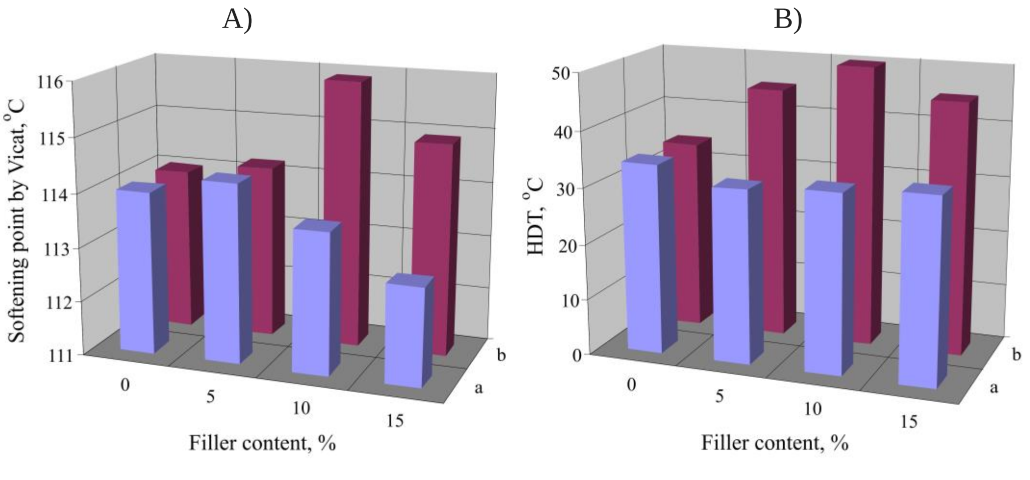

Figure 4. Dependence of: A) Vicat Softening Temperature, and B) Heat Deflection Temperature of LDPE filled with powdered natural fillers in the form of a) wheat bran and b) pumpkin seed husks on various contents.

The graphic dependence of the heat deflection temperature on the content and type of the filler is shown in Figure 4B. The heat deflection temperature of the injection moldings tested shows a decreasing tendency when increasing the content of the natural filler in the form of wheat bran. The addition of powder filler in the form of pumpkin seed husks to low density polyethylene results in an increase in the tested property and then a decrease. The probable cause of the decrease in the heat deflection temperature of the obtained samples with the addition of wheat bran may be the poor adhesion of the filler grain to the polymer matrix.

3.4. Morphology of produced polymer compositions

As a part of the conducted tests, microscopic tests of the cross-section of the obtained polymer mixtures containing 5, 10, 15 and 20% of natural fillers were carried out. Only the pictures of morphology of the injection moldings examined containing pumpkin seed husks were presented in this article (Fig. 5).

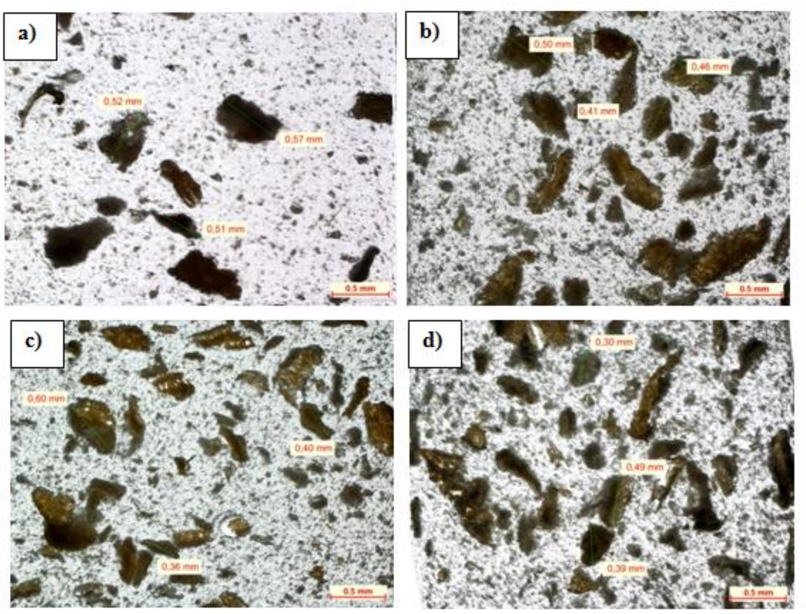

Figure 5. Microscopic photos of the cross-section of tested polymer mixtures filled with powdered pumpkin seed husks of the grain size 0.4-0.8 mm: a) LDPE + 5% natural filler; b) LDPE + 10% natural filler; c) LDPE + 15% natural filler; d) LDPE + 20% natural filler.

The analysis of the morphology showed an uneven distribution of the added natural filler. Particles of varying sizes and agglomerates can be observed. Micrometric pumpkin seed husks particles with probably higher surface energy than surface energy of low density polyethylene tend to agglomerate in such medium (Figures 5b, 5c and 5d), which is an unfavorable phenomenon, because it reduces the contact surface between the filler and the polymeric material, which results in reduction of adhesion.

4. Conclusions

Modification of polymer materials with various types of natural fillers brings with it many changes starting from the course of the processing process, mechanical and thermal properties, to the morphology of the obtained products. Analysis of changes in the properties of such modified polymers is important from the point of view of the applicability of the polymer composition.

On the basis of the research on the properties of a polymer mixture of low density polyethylene with natural fillers, it has been found that they are suitable for use as a filler for this material. In the case of Young's modulus tests, there was a significant increase in this value, and in the case of hardness tests, a slight increase in the tested property along with an increase in the content of fillers and grain size. VST and HDT do not show significant changes in the range of applied grain sizes and the tested mass filler content.

Acknowledgments
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