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The Effects of the Substitution of Wood Fiberwith Agro-based Fiber (Barley Straw) on the Properties of Natural Fiber/Polypropylene Composites

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Abstract. Ecological concerns and the impending depletion of fossil fuels are driving the development of new bio-based, green products. Natural fibers are used increasingly as a filler or reinforcement in commercial thermoplastics due to their low cost, high specific properties and renewable nature. Agricultural byproducts and wastes are excellent alternative materials to supplement or substitute wood material as a reinforcement in composites.This comparative study focuses on the effects of the substitution of wood fiber with agro-basedfiber (barley straw) on the mechanical and physical properties of natural fiber/polypropylene composites. The studied mechanical properties are flexural strength and modulus, Brinell hardness and Charpy impact strength. Water absorption and thickness swelling are studied as physical properties. Generally, the research resultsindicate that almost all the studied properties weakened significantlyas woodwas substituted with barley straw.Ofmechanical properties, the major decrease wasobserved in hardness.However, the use of barley straw slightlyimproved impact strength.The moisture-related properties, water absorption and thickness swelling,which have a great impact on the durability of a composite material, weakened significantly.

1 Introduction

Composites made from a blend of thermoplastic and natural fibers have become the subject of much research and attractive to manufacturers because of their many advantages, such as improved environmental performance, mainly due to the use of biodegradable materials, and reduction in the use of non-renewable (oil-based) resources throughout the whole life-cycle of the composite [1]. However, the main disadvantage of these natural fiber/polymer composites seems to be the incompatibility between the hydrophilic natural fibers and the hydrophobic thermoplastic matrix,which makes the use of compatibilizers or coupling agents necessary in order to improve the adhesion fiber/matrix [2].

In general, cellulosic materials can be characterized as ubiquitous, cheap, renewable, biodegradable and low-density materials with good mechanical properties, such as high strength and stiffness, making them suitable for reinforcing wood-plastic composite (WPC)or natural fiber composite (NFC) material [3, 4]. The potential of any natural fiber depends on its chemical structure, especially the cellulose content and mechanical properties of the fibers [5].

Due to the global demand for fibrous materials, worldwide shortage of trees in many areas, and increased environmental awareness, research on the development of composites prepared by using alternative low cost lignocellulosic sources and various waste materials, such as agricultural byproducts or agro-waste materials, is also pursued actively in order to decrease the overall manufacturing costs and improve the properties of the materials [6 7, 8]. Field crop residues and/or agricultural byproducts are excellent alternative waste materials to supplement or substitute wood because they are abundant, inexpensive, widespread, and easily available [9]. The use of cereal straw and other agricultural byproducts or agro-waste materials as fillersin the production of plastic composites alleviate the shortage of wood resources, and can have the potential to start a natural fiber industry in countries where there are little or no wood resources left [6, 7, 8].

Agro-based material, such as straws and stalks are also sources of cellulose-based fibers which are increasingly considered as low-cost potential alternatives to wood fibers,although they have lower cellulose content compared to wood [5].Fibers with higher cellulose content, i.e. higher degree of polymerization of cellulose and lower microfibrillar angle, render better mechanical properties. However, the quality of the fibers obtained will determine their end use, and not the cellulosic content. A major difference between straw and wood fibers is the ash content. Ash contains silica,which has many undesirable effects, e.g. it blunts the cutting machinery and makes combustion more difficult[10].

The traditional use of straws includes bedding for animals and livestock feeding. Some agro-based residues such as wheat straw, corn stalk and bagasse have already been successfully utilizedin manufacturing composites and panel boards [5]. Barley strawsareused as a construction

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material, e.g. in wall plaster and concrete, due to their good insulation properties [11, 12] but not reported to be exploited in the production of WPCs/NFCs. However, the utilizing of barley husksin the production of polypropylene composites has been studied, and improvements in mechanical properties have been observed [7].

The aim of this study is to investigate the potential of agro-based material as a substitute of wood material in natural fiber/polypropylene composites. The mechanical and physical properties of barley straw flour -filled polypropylene (PP)are evaluated and compared with that of wood flour-PP composites. The studied properties are flexural strength and modulus, Charpy impact strength andBrinell hardness as mechanical properties, and water absorption and thickness swelling as physical properties.

2 Materials and Methods

The wood fiber (WF) used in the study was spruce (*Piceaabies*) saw chips with the specific gravity of 1.58 g cm^{-3}. Sieve analysis revealed that the wood particles had a larger fraction of long particles (2.0 mm). Barley (*Hordeumvulgare*) straw flour (SF) was used as agro-based material.The barley straws, obtained from a local farmer, were ground with an Unthawood shredder, and then agglomerated by usinga PlasMecs.r.l mixing plant.

The thermoplastic matrix in the composite was commercially available polypropylene (PP) supplied by IneosPolyolefins (Eltex P HY001P). The polypropylene homopolymer had the density of 0.91 g cm^{-3} and melt flow rate of 45 g 10 min^{-1} (230 ^{o}C 2.16 kg^{-1}). The coupling agent was maleated polypropylene (MAPP), Orevac® CA 100 (Atofina, France). Struktol TPW 113 was used as the lubricating agent. The compositions of the WPCs are shown in Table 1.

Table 1. Composition of WPCs [wt%]

Composite	Fiber	PP	MAPP	Lubricant
WF-PP	64.0	30.0	3.0	3.0
SF-PP	64.0	30.0	3.0	3.0

The fiber material, polypropylene, and additives were compounded with a Weber CE7.2 conical twin-screw extruder. The screw had the L/D ratio of 17, and screw speed was 13 rpm. The barrel temperatures of the extruder were 160-190^{o}C. The pressure at the die varied between 3 to 4 MPa, depending on the material blend, and the material output was 25 kg h^{-1}. The hollow profiles were extruded through a rectangular die (Fig. 1).

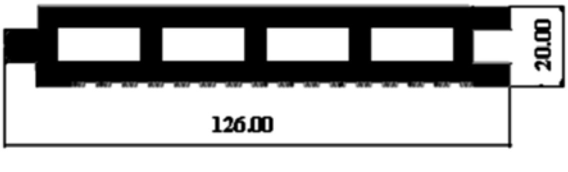

Fig.1. Hollow profile of extruded wood-polypropylene composites

The Charpy impact strength for unnotched samples tested in flatwise position was determined with a Zwick 5102 Model impact tester in accordance with the method ISO 179-1/1f U. The method is based on the recommendation given by TS 15534-1 Wood-plastic composites (WPC)-Part 1: Test methods for characterisation of WPC materials and products. The specimens were conditioned at 23 ± 1 °C and 50% relative humidity for at least 24 h before the Charpy impact test was performed.The Charpy impact strength was calculated according to the following equation:

$$a_{cU} = \frac{E_c}{h \times b} \times 10^3 \quad (1)$$

whereEc is the corrected energy, in joules, absorbed by breaking the test sample; h is the thickness, in millimeters, of the test sample; and b is the width, in millimeters, of the test sample.

For the bending test, rectangular test pieces with the following dimensions: width 50±1 mm, thickness 20±1 mm and length 450 ± 1 mm, were cut from the extruded panels. The flexural strengthand flexural modulus of the compositeswere measured with a standard material testing machine (Zwick GmbH & Co. KG) in accordance with EN 310 and EN ISO 527 standards.

Hardness (Brinell) was measured from 100 × 80 mm composite samples according to standard SFS-EN 1534.

The resistance of the composites to water absorption and thickness swelling was tested according to the procedure described in EN 317, which includes 28-day immersion of the composites in water.The tests were carried out with 20 sample replicates with the width 50 mm and length 50 mm for each type of WPC studied. The thickness of the samples was the nominal thickness of each panel. The sample replicates were weighed and their dimensions were measured before their immersion into water. Water absorption (WA) was calculated according to the following equation:

$$WA = \frac{(M_e - M_o)}{M_o} \times 100 \quad (2)$$

where M_e is the mass of the sample after immersion, g; and M_o is the mass of the sample before immersion, g.

Thickness swelling (TS) is an important property that indicates the stability performance of the WPC. TSwas calculated as follows:

$$TS = \frac{(T_e - T_o)}{T_o} \times 100 \quad (3)$$

whereT_e is the thickness of the sample after immersion, m; and T_o is the thickness of the sample before immersion, m.

3 Results and Discussion

The effects onthe mechanical properties of the composites are shown in Fig. 2.

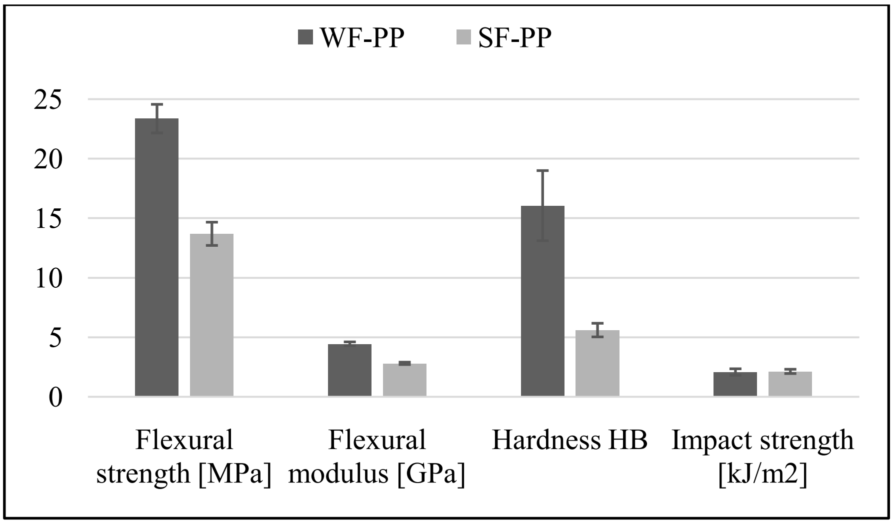

Fig.2. Mechanical properties of WPCs

The use of straw fiber flour as a reinforcement of the composite was found to cause a significant drop of all the studied mechanical properties, except for the impact strength. The most obvious drop wasdetected inhardness and flexuralproperties. The Brinellhardness of the SF-PP composite was 65 % lower thanthe hardness ofthe WF-PP composite. The flexuralproperties of the SF-PP composite werealso remarkably lower than the corresponding values of the WF-PP composite.The flexural strength of the SF-PP composite exhibited a decrease of 41 % from 23.4 to 13.7 N/mm^2. In the case offlexural modulus, the decrease was 37 %. It was also observed that the Charpy impact strength property increased slightly with the use of barley straw flour. The improvement was approximately 2.4 %.

The results deviate partly from the results obtained by Panthapulakkal and Sain[6],who compared different agro-residues as fillersin high-density polyethylene composites. With quite a similar filler loading as in the present study, they observed that the wheat straw flour -reinforced composite had slightly improved flexural and tensile properties compared to the wood flour -reinforced composite. Also, in theirstudy the change of impact strength was insignificant, with a decrease less than 1.5 %.Generally, the strength of fiber-reinforced composites depends on the properties of the constituents and the interface interaction [13].The mechanical properties of composites can be attributed to the quantity, strength and modulus of cellulosic fibers, as well as to interfacial adhesion between the matrix and the fiber. Compatibilizers, such as MAPP, are used to improve interfacial adhesion between hydrophilic cellulosic fibers and hydrophobic polypropylene [7, 8, 14, 15], and consequently, the mechanicalandmoisture-related properties [16] of composites are improved.Due to the differences in chemical compositions ofvarious cellulosic fibers, the effects of compatibilizerare not similar for all. Wood and barley straw contain the same principal components, that is, cellulose, hemicellulose, and lignin,but there are significant differences in the proportions of those components. Cellulose provides positive effect on mechanical and other properties of the composite material. Lignin is considered to be largely responsible for strength and durability of wood [17]. Therefore, e.g. in case of barley straw, different loadings and amounts of additives should be studied more to optimize the effect of compatibilizer.

Table 2 shows the water absorption and thickness swelling of the WPCs. Both composites showed a high uptake of water,which was attributed to the high content of lignocellulosic materials present in the composites. In general, the SF-PP composite had significantly higher water absorption and thickness swelling than the WF-PP composite. After 28 days of immersion, theSF-PPcomposite absorbed water by 32 %,which was about 70 % higher than the water absorption of the WF-PP composite.In thickness swelling the difference was smaller, approximately 20 %. The SF-PP composite absorbed watervery rapidly during the first 7 days of immersion. After that, the water absorption stabilized over the total immersion time. As for the WF-PP composite, the water absorption increased quite linearly over the total immersion time.The resultsof water absorption are in accordance with the results of Zabihzadeh [18], who also found that the water absorption of wheat straw composites was clearly higher than that of wood composites.

The hydrophilic nature of natural fibers is responsible for water absorption in composites, and therefore a higher loading of fibers leads to a higher amount of water absorbed [19]. The increased moisture content of composites is primarily responsible for the degradation of the fibers and the fiber-matrix interface, resulting in a loss of mechanical properties [2]. The water absorption of composites depends on the presence and amount of cavities/water pathways in their structure; thus it also related to the density of the composite [20], i.e. composites with high density have low moisture content, and composites with low density have high moisture content [21]. The amount of voids on the surface and at the interface of the composite can be affected by the use of a lubricant in the manufacturing process of WPCs.

Table 2. Water absorption and thickness swelling of WPCs

Composite	Water absorption [%] (STD)			
	24 h	7 days	14 days	28 days
WF-PP	3.35 (0.11)	9.13 (0.19)	14.86 (0.41)	19.18 (0.21)
SF-PP	13.39 (0.12)	26.97 (0.30)	30.77 (0.26)	32.27 (0.33)
Composite	**Thickness swelling [%] (STD)**			
	24 h	7 days	14 days	28 days
WF-PP	0.69 (0.13)	2.48 (0.21)	4.51 (0.22)	6.93 (0.20)
SF-PP	2.57 (0.70)	7.74 (0.64)	8.32 (0.67)	8.46 (0.83)

4 Summary

The feasibility of utilizing barley strawas an alternative filler for wood in natural fiber/polypropylene composites was studied.The mechanical and physical properties of these fillers were studied to compare their reinforcement ability.

In mechanical properties, the substitution of wood flour with barley straw flour was found to weaken the flexural and hardness properties. The most significantdecrease was observed in Brinellhardness. Also the flexural properties decreased significantly with the use of barley straw as the filler. The substitution of wood flour with barley straw flour had only a minor effect on the Charpy impact strength. The use of straw flour increased the impact strength slightly.

In physical properties, the substitution of wood flour with barley straw in natural fiber/polypropylene composites was found to increase water absorption and thickness swelling remarkably. The increased moisture content propagated to weakened properties of the composite.

According to the results, the reinforcement ability of barley straw flour was significantly lower than that of wood flour. Agro-based materials, such as barley straw, offer a huge low-cost potential alternative to wood fibers in composites. Therefore, they are suitable fillers for products where mechanical properties are not critical, andwhere they are not exposed to a high moisture content. Further research should be done to determine the favorable loading and advantageous particle size of barley straw when used as a filler in natural-fiber composites. Also the effect of different fillers, such as mineral fillers, should be studied in order to find out how they affect the mechanical and physical properties of natural fiber/polypropylene composites.

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