RECYCLING ABILITY OF THE POLYPROPYLENE FOR PRODUCTION OF THE NATURAL FIBER COMPOSITES

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SUMMARY

The focus in this work has been put on the possibility to use recycled polypropylene (PP) in production of composites based on renewable raw materials. The materials are kenaf fibers (KF) and rice straw (RS) as reinforcements and PP as matrix. The matrix was recycled once and two times and the fibers were compounded with the recycled matrix. Rice straw /Polypropylene/Coupling agent (RS/PP/CA) and Kenaf/Polypropylene/Coupling Agent (K/PP/CA) composites with fiber content of 30 wt. % and appropriate compatibilizing agent (CA), have been prepared by two steps procedure: extrusion and compression moulding. The processing and material properties have been studied on the composites with recycled matrix and compared to the composites with virgin matrix. The composite strength of composites with recycled PP is similar to that of composites with virgin PP. The thermal stability of recycled PP based composites is very similar to the thermal stability of the composites with virgin matrix. The interfacial adhesion was studied by SEM microphotographs. It shows that adhesion between fibers and matrix in all composites is very good. These composites based on recycled PP matrix show good applicable characteristics as construction materials for housing systems.

Key words: eco-composites, natural fibers, rice straw, kenaf, polypropylene

INTRODUCTION

The growing environmental awareness and new rules and regulations are forcing the industries to seek more ecologically friendly materials for their products. In recent years, the development of biocomposites from biodegradable polymers and natural fibers have attracted great interests in the composite science, because they could allow complete degradation in soil or by composting process and do not emit any toxic or noxious components. For example automotive applications based on natural fibers with polypropylene as matrix material are very common today. Biodegradable polymers from renewable resources are becoming more and more interesting in a society where a large part of the garbage and waste consist of polyolefins [1-4].

Natural fibers have many advantages compared to synthetic fibers, for example low weight, and they are recyclable and biogradable. They are also renewable and have relatively high strength and stiffness and cause no skin irritations. Many investigations have been made on the potential of natural fibers as reinforcements for composites and in several cases the results have shown that the natural fiber composites own good stiffness [1-6]. However the main drawback of natural fiber may be their hydrophilic nature, which decreases the compatibility with hydrophobic polymeric matrix. In these (natural fiber-reinforced) composite fields, therefore, most of the research has used different kinds of coupling agents for improving interfacial properties between the polymer matrices and natural fibers in order to enhance the physical and mechanical properties of the final products.

The purpose of this study was to investigate the recycling ability of the matrix PP and its applications for production of the kenaf fiber reinforced composites and rice straw reinforced composites. The matrix was recycled one and two times with a twin-screw extruder. The fibers were compounded with matrix and coupling agent by melt mixing and the compounds were compression molded. A maleic anhydride grafted PP (MAPP) was used as a coupling agent to improve the compatibility and adhesion between the fibers and matrix. The mechanical properties of the composites were studied according to the flexural testing and the thermal properties were studied with thermo gravimetric analysis (TGA).

MATERIAL AND METHODS

The materials which were used have been:
Matrix: Polypropylene PP. It is suitable matrix for extrusion of composite materials. In order to promote the matrix/fiber compatibilisation, different amount of MA grafted PP has been added during the blending (reactive blending).
Reinforcements: Kenaf fibers and Rice Straw. They were dried at 80^{O}C for 7-9h before they were mixed to produce composites.
First, the PP matrix was recycled one and two times (PPx1 and PPx2) by extrusion procedure with a twin-screw extruder. The obtained recycled matrices were cut in pellets form to perform the sheets.
The preparation of the composites have been preformed by melt mixing, in a Brabender-like apparatus with progressively increasing RPM (speed screw). During the blending the coupling agent MAPP has been added. The polymer and coupling agent were first mixed than have been added the fibers. The fiber content in all composites was 30 wt. %. The kneading temperatures were 185^{O}C. They have been mixed for 10 minutes with progressively increasing RPM. The obtained composites were cut in pellets form to perform the sheets.
The main goal is represented by the use of recycled polymeric matrix that can obtain the composites with good practical characteristics for using as building materials. For this purposes different composites have been prepared, as reported in table 1.

Table1. Prepared composites

Composites	Matrix	Fiber
PP/RS/CA*	PP	Rice Straw
PPx1/RS/CA	PP recycled one time	Rice Straw
PPx2/RS/CA	PP recycled two times	Rice Straw
PP/K/CA	PP	Kenaf fibers
PPx1/K/CA	PP recycled one time	Kenaf fibers

* CA = coupling agent

Test samples for mechanical testing were fabricated by hot-press forming with a compression moulding press. The pellets from the composites were put in moulding frame with desired dimensions and they have been molded by thermo compression at T= 185^{O}C for 10 minutes with increasing pressure up to 10 000 pounds. After expire of the heating time, the press was cooled by circulating cold water.
From the recycled matrices and from all composites the plates with thickness 3 mm were produced.
The flexural testing was performed according to ASTM standard for flexural testing on an Instron. The flexural tests were performed on unnotched (Young's Modulus) samples. At least 6 specimens were tested for every material.
Thermal degradation temperature was measured by a thermo gravimetric analyzer (TGA) Perkin Elmer in a nitrogen atmosphere. The samples of about 10 mg were heated from 50 to 600^{O}C at a rate of 20^{O}C/min under nitrogen flow (25ml/min).

RESULTS AND DISCUSSION

The mechanical properties of the recycled matrix were compared to neat matrix and the mechanical properties of the composites with recycled matrix were compared to the composites with neat matrix. Table 2 shows the summary of the results for the matrices. There aren't drastically differences between mechanical properties of the neat matrix and recycled matrices. The flexural strength for PP neat and recycled: PPx1 and PPx2 are very similar but the modulus for PPx1, and PPx2 are higher than PP neat. The flexural strengths for PP are decreased for a little (about 5 %) with increasing of the number of recycles of the PP but the flexural modulus are increased for about 25%.

Table 2. The flexural tests of neat PP and recycled PP

	Stress at peak, MPa	Standard Deviation	Modulus, MPa	Standard Deviation
PP neat	51,5	5,5	1081	120
PP x1	52,8	2,1	1305	64
PP x2	49,5	2,9	1341	101

Table 3 shows the summary of the flexural results for kenaf - composites with neat and recycled matrices and rice straw - composites with neat and recycled matrices.
The obtained values for PP recycled based composites for flexural strength and modulus are very similar with value for composite with neat PP. There are differences between flexural strength of the composites reinforced with RS and composites with Kenaf. Generally, the kenaf - composites have better mechanical properties then rice straw composites.
Fig. 2 a) shows the influence of the recycled matrices on the properties of the composites. As shown in Fig. 2, which presents the property retention of the composites based on neat PP, PPx1 and PPx2 (in dependence of the number of recycles of the matrices), the flexural properties for PP recycled based composites were held close to the flexural properties for composite based on neat PP.

Table 3. The flexural tests of composites with neat and recycled matrices

	Stress at peak, MPa	Standard Deviation	Modulus, GPa	Standard Deviation
PP / RS / CA 60/30/10 wt/wt	42,6	3,4	1,9	0,082
PPx1 / RS / CA 60/30/10 wt/wt	42,2	1,2	1,8	0,041
PPx2 / RS / CA 60/30/10 wt/wt	39,6	4,6	1,8	0,063
PP / Kenaf / CA 60/30/10 wt/wt	51,3	4,8	2,1	0,068
PPx1 / Kenaf / CA 60/30/10 wt/wt	51,1	3,0	2,3	0,204

A research group Oksman et al. [7] have studied the recycling properties of the PLA/kenaf composite. The physical properties and molecular weight were held close to 90% of that of the initial PLA/kenaf composites. The reason for the decrease in the physical properties decline was mainly the decline in the molecular weight of PLA and the kenaf fiber's length caused by repeated kneading. However, the physical properties of the PLA/kenaf composite probably can be kept constant by the adjustment of the ratio of the initial PLA/kenaf composite and the recycled PLA/kenaf composite.

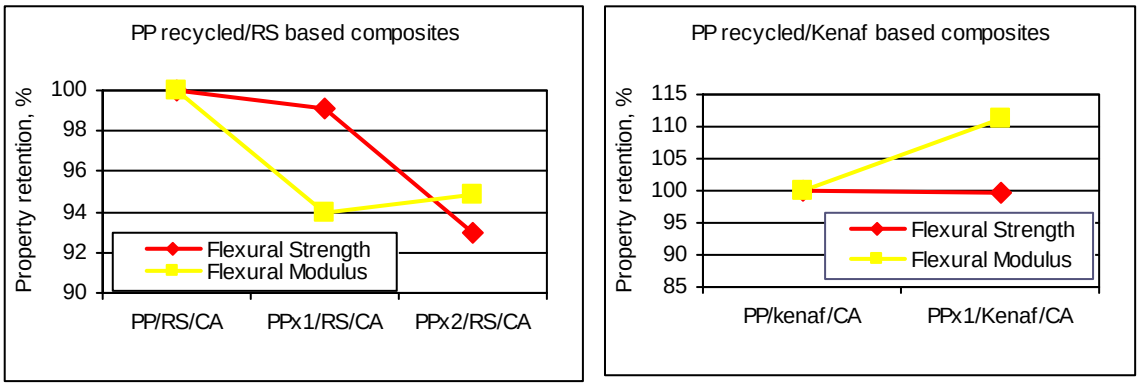

Fig. 2. Influence of the recycled PP on the properties of the PP/RS and PP/kenaf composites

Thermal degradation of PP and PPx1, PPx2 occurred at 443,9 ^{O}C, 432,9 ^{O}C and 422^{O}C respectively. The thermal stability of the recycled PPx1 and PPx2 are almost without differences but a little smaller than thermal stability of the PP neat (see Table 4). In the case of PP recycled based composites, the incorporation of the recycled matrix one and two times has a very small affected the thermal degradation temperature, (see Table 5). The composites with recycled matrices showed a lower degradation temperature (less than 40^{O}C) than a composite with neat matrix, and a two-stage loss of mass was mainly observed for all composites, see fig.5. These lower degradation temperatures may be attributed to the decrease of molecular weight of PP by more times kneading. It can be generally said that the increase of molecular weight by cross-linking reaction between matrix and fibers, or molecular chain – extension of the matrix itself, could increase the thermal degradation temperature [7-9].

Table 4. Weight residual for neat and recycled PP

	Td [oC] (weight residual) ~ 90 %	*Td* [oC] (weight residual) ~ 50 %	*Td* [oC] (weight residual) ~ 10 %
PP neat	377,60 (90,38 %)	429,32 (50,38 %)	449,79 (10,38 %)
PP x1	357,49 (90,89 %)	414,67 (50,87 %)	438,64 (10,89 %)
PP x2	354,87 (86,37 %)	403,64 (46,37 %)	427,15 (11,37 %)

Table 5. Weight residual for composites with recycled matrices

	Td [oC] (weight residual) ~ 90 %	Td [oC] (weight residual) ~ 50 %	Td [oC] (weight residual) ~ 10 %
PP / RS / CA	344,43 (89,74 %)	411,21 (49,74 %)	452,17 (9,74 %)
PP x1/RS/CA	309,09 (87,51 %)	385,22 (47,51 %)	458,82 (12,51 %)
PP x2/RS/CA	343,53 (86,82 %)	405,97 (46,82 %)	475,27 (11,82 %)
PP / Kenaf / CA	356,81 (85,94 %)	408,94 (50,94 %)	441,96 (10,94 %)
PP x1/ Kenaf / CA	356,92 (86,98 %)	412,35 (51,98 %)	443,77 (11,98 %)

Based on this work devoted to study the effect of recycled matrices PP in composites reinforced with rice straw and kenaf fibers, on the mechanical properties, the following conclusions can be drawn:
The flexural properties of the PP recycled based composites are very close to flexural properties of the composite with neat PP. In particular, PP-based composites with kenaf fibers have sown high mechanical properties.
The thermal stability of the PP recycled composites is slightly lower compared to composite with neat matrix.
In particular, recycled PP matrices are effective materials for production of the natural fiber composites with high mechanical properties, for building applications.

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