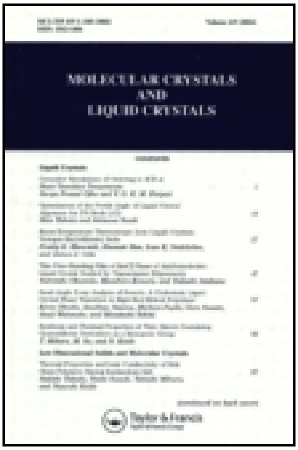

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Sławomir Borysiak [a] & Dominik Paukszta [a]
[a] Poznan University of Technology, Institute of Chemical Technology and Engineering, Poznan, Poland

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Mechanical Properties of Lignocellulosic/Polypropylene Composites

Sławomir Borysiak and Dominik Paukszta
Poznan University of Technology, Institute of Chemical Technology and Engineering, Poznan, Poland

This article discusses the mechanical properties of different composites based on pine, oak wood, rapeseed straw and isotactic polypropylene. The surface of the filler was modified with acetic anhydride to enhance the compatibility between hydrophilic lignocellulosic components and hydrophobic polypropylene matrice. After treatment, mixtures of PP with 30% of the natural components were extruded and injection molded. Tensile strength of composites was not improved by the rapeseed straw chemical modification. However, chemical treatment was successful in improving the strength of the wood in the PP matrice. The results concerning mechanical properties of composites, their aesthetic quality and possibility of their application in various fields indicate an opportunity of production of polypropylene/lignocellulosic composites. Moreover, rapeseed straw so far have been considered as waste materials in plastics industry.

Keywords: composites; mechanical properties; polypropylene; rapeseed straw; wood

INTRODUCTION

The composites of engineering polymers with natural lignocellulosic materials like flax, hemp, ramie, sisal fibres and wood have been recently intensively investigated due to interesting physical properties of such materials and their ecological aspects [1–5].

The processing of the composites a number of important problems to be dealt with. These include low compatibility between the wood and polymer matrice; instability of the lignocellulosic components of temperatures above 200 C; high moisture intake, and poor dispersion of

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Address correspondence to Sławomir Borysiak, Poznan University of Technology, Institute of Chemical Technology and Engineering, 60-965 Poznan, Poland. E-mail: Slawomir.Borysiak@put.poznan.pl

the filler in polymer melt. One of the essential problems in the technology of such kind of composites is adhesion between the components, because polymer matrices are hydrophobic whereas the surfaces of the lignocellulosic materials are hydrophilic [6–8]. Incompatibility between lignocellulosic particles and thermoplastic matrices can be improved by addition of chemical coupling agents [9–11]. Another method of improving the adhesion between wood and polypropylene matrice is chemical modification of natural materials and/or matrices [13–15]. Anhydrides such as maleic, acetic, succinic are popular modificators in wood/PP composites. Chemical tratment of lignocellulosic components is most important for mechanical properties of final materials. However, the relationship beetwen applied chemical modification and mechanical properties is very controversial. For example, J. Wu *et al.* [10], W. D. Ellis *et al.* [16] and R. Mahlberg *et al.* [12] showed significant improvement in the mechanical properties of the PP/wood composites as a result of the anhydride modifications. On the contrary, Nunez *et al.* [9] note that mechanical properties have not improved either by the wood chemical modification (esterification with maleic anhydride) or by the use of compatiblilizing agent.

The mechanical properties depends on many factors, for example: type of fillers, chemical treatment of components to improve adhesion, etc. Moreover, lignocellulosic materials can act also as a nucleant for thermoplastic polymer causing changes in morphology or even changes in the arrangement of chains in crystal lattice of matrix [17].

The present study focuse on new composites based on PP and rapeseed straw and also wood fillers.

EXPERIMENTAL DETAILS

Materials

The polypropylene Malen F-401 (Orlen Plock, Poland) was used as a matrice. It was characterized as follows: isotactic content – 95 %, melting point – 166 °C and melt flow index – 2.4–3.2 g/10 min.

In this work the following lignocelllulosic materials were used:

- Wood:
  - a) pine (*Pinus silvestris L.*)
  - b) oak (*Quercus pedunculata L.*)
- Rapeseed straw (*Brassica napus L.*)
  - a) Kaszub variety
  - b) Californium variety

TABLE 1 The Chemical Compositions and Moisture in Wood and Rapeseed Straw

	Pine wood	Oak wood	Kaszub rapeseed straw	Californium rapeseed straw
Cellulose, %	46.6	40.8	38.1	37.4
Lignin, %	28.0	26.5	18.8	21.0
Organic solvents soluble, %	4.6	6.6	2.8	5.1
Water soluble %	0.9	3.1	13.1	11.5
Hot water, %	3.7	10.9	21.1	18.9
Moisture, %	7.5	11.1	10.8	9.3

These lignocellulosic components were crumbled into fractions. To produce the composites we used 1–2 mm fraction. On the basis of the analytical results, it has been found the chemical compositions as well as moisture. Components means values of wood and rapeseed straw fillers are shown in Table 1.

Chemical Treatment of Wood and Rapeseed Straw

To improve the adhesion between components, a part of wood was mercerised by treatment with 17.5% solution of sodium hydroxide for 60 minutes. In case of rapeseed straw we applied other mercerization conditions: 20% solution of NaOH and treatment time of 120 min. The parameters of alkalization result from optimalization procedure of wood components.

In the next step, the lignocellulosic materials were chemically modified by reaction with the acetic anhydride. The lignocellulosic materials were immersed in acetic anhydride/xylene mixture. The content of acetic anhydride was 300% (wt) in proportion to the filler mass. The time of treatment was 120 minutes at temperature 120°C. After reaction the lignocellulosic materials were washed several times with ethanol and distilled water to remove unreacted acetic anhydride. Then the natural materials were dried for 48 h at room temperature until constant weight was achieved. The moisture content of the wood fillers before blended with PP was about 3.8%. The fact that the esterification process of wood occurred is confirmed by the weight percent gain (WPG). Values of the WPG index for the wood modified with the acetic anhydride are comparable and range from about 14 to 16%.

Preparation of the Composites

The composite with lignocellulosic materials was obtained by extrusion method using a “Fairex” (Mc Nell Akron Repiquetn, France)

TABLE 2 Values of Temperatures in Zones at Extrusion Method

Temperatures in zones at extrusion method [°C]			
I zone	II zone	III zone	head
140	185	195	200

single-screw extruder with a length-to-diameter ratio L/D of 25 and screw diameter of 25 mm. Extruding process parameters are presented in the Table 2. The extrusion speed was adjusted to 30 rpm. The extruded materials were water quenched, granulated and subsequently dryied.

Various composites with lignocellulosic components were obtained. The composite materials contain 30% of the lignocellulosic materials. Below are shown the abbreviations of all the materials: PP with unmodified pine wood (PP-PW), PP with modified pine wood (PP-MPW), PP with unmodified oak wood (PP-OW), PP with modified oak wood (PP-MOW), PP with unmodified Kaszub rapeseed straw (PP-RK), PP with modified Kaszub rapeseed straw (PP-MRK), PP with unmodified Californium rapeseed straw (PP-RC), PP with modified Californium rapeseed straw (PP-MRC).

The samples for the mechanical tests were produced by injection moulding method. The parameters of this processing technique are presented in Table 3. Moreover, an injection pressure of 60 bar, head temperature of 30°C, and cooling time of samples in injection mould of 30 sec were applied. After molding, the specimens were stored over desiccant at room temperature before mechanical testing.

MECHANICAL INVESTIGATIONS

This work analyses selected mechanical properties of blends: tensile strength, Young's modulus at tensile, elongation, impact resistance and hardness. Mechanical tests on composites have been carried out at room temperature with the use of Instron machine Model 4481 at a constant speed of 10 mm/min, according to standard PN-81/C-89034.

TABLE 3 Values of Temperatures in Zones at Injection Technique

Temperatures in zones at injection technique [°C]			
I zone	II zone	III zone	head
200	200	190	185

RESULTS AND DISCUSSION

Figures 1–3 show the results of the tensile mechanical investigations. The tensile strength of wood/PP and rapeseed straw/PP composites as well as polypropylene matrice is shown in Figure 1.

As regards composite samples with wood it was observed that addition of unmodified pine or oak wood particles causes decrease in tensile strength in comparison with polypropylene matrice. On the other hand, chemical modification of wood causes increase in strength. Values of tensile stress for these composites are higher than unfilled polypropylene. It is interesting that tensile strength for samples with two different species of wood is similar.

The characteristics of tensile strength for materials containing rapeseed straw prove to be completely different. The use of unmodified fillers from these lignocellulosic materials causes worsening tensile strength in comparison with polypropylene. The decrease of this parameter is more than that for analogical samples with wood particles. Chemical modification with the use of acetic anhydride is responsible for increasing value of tensile stress for composites with Kaszub variety only. In case of variety Californium chemical treatment didn't improve this parameter. It is worth noting that both composites containing rapeseed straw are characterised by lower values in comparison with polypropylene matrice. The Young's modulus of the composites are shown in Figure 2.

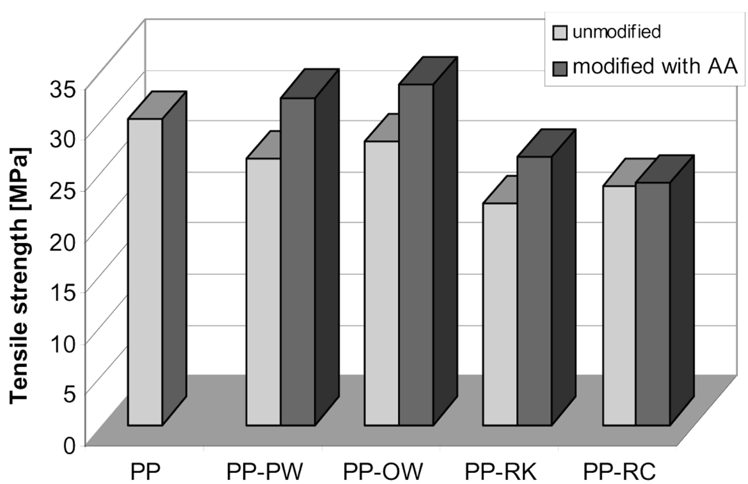

FIGURE 1 The tensile strength of wood/PP and rapeseed straw/PP composites.

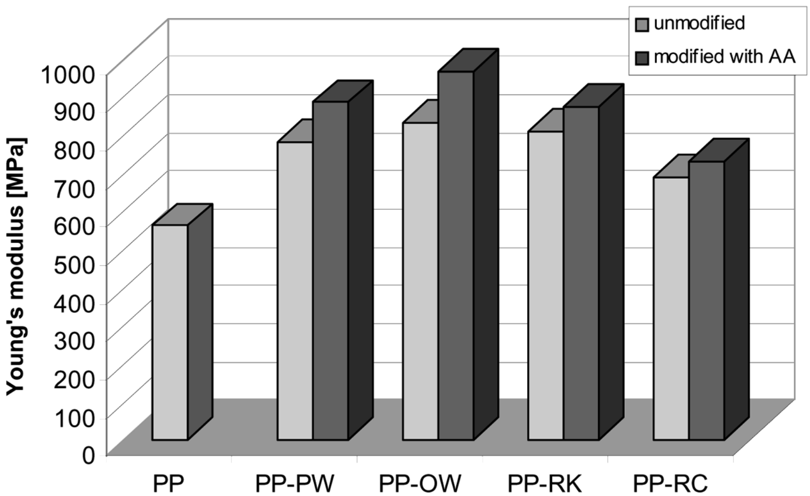

FIGURE 2 The Young's modulus of wood/PP and rapeseed straw/PP composites.

The characteristics of all composites filled by lignocellulosic materials are increasing Young's modulus in compare to thermoplastic polymer matrice. This phenomenon is also confirmed for samples investigated in this work. For polypropylene composites containing

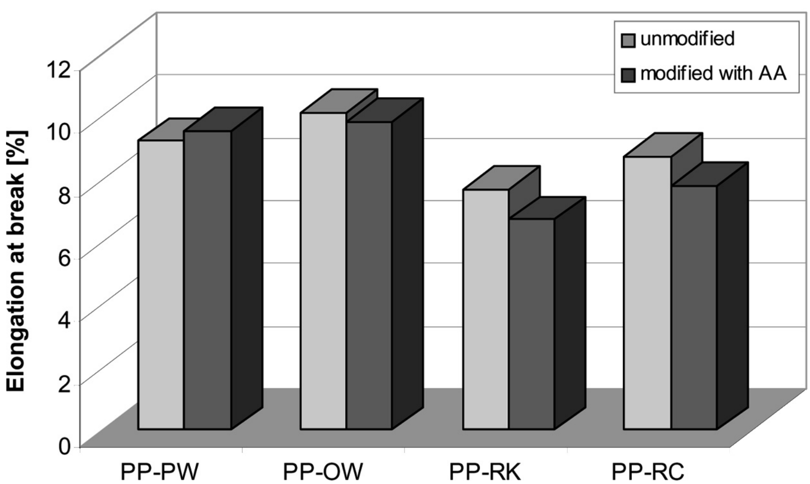

FIGURE 3 The elongation at break of wood/PP and rapeseed straw/PP composites.

unmodified fillers the growth of modulus values ca. 20–30% was noted. The using of chemical modification influences on increasing of elasticity, for example for PP/oak wood composites growth 40% was noted. It was found that the type of filler used has a significant impact on on elasticity of composite materials. The highest value of Young's modulus was identified for polypropylene composite with oak wood particles, on the contrary the lowest value of this parameter for composite with rapeseed straw variety Californium was noted. It is very interesting that composites with pine wood and rapeseed straw variety Kaszub (also unmodified and modified) display a high similarity in vales of elastic strain. The elongation at break of composites materials are shown in Figure 3.

All the investigated composites are characterised by very similar values (7–10%) of elongation at break. The chemical modification does not have an impact on the elasticity of composite materials. In addition, the composites are characterized by clearly lower values of elongation in comparison with polypropylene matrice. The elongation measured for polypropylene exceeds 300%. Moreover, the impact resistance and the hardness measurement was done. The impact resistance of composites is shown in Figure 4.

In all the cases, the impact resistance increases with lignocellulosic materials content, but increment is larger for wood filler. The impact

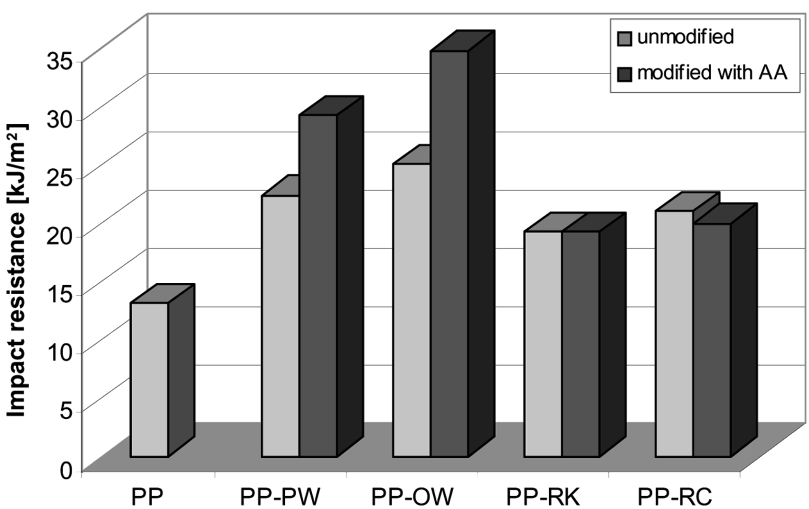

FIGURE 4 The impact resistance of wood/PP and rapeseed straw/PP composites.

resistance of unmodified wood/PP composites varies between 22 and 24 kJ/m^2. The application of chemical modification of wood causes a dramatic increase in impact resistance up to 29–35 kJ/m^2. A completely different effect has been observed for rapeseed straw/PP composites. The impact resistance of these composites is significantly higher than that of the matrix, but smaller in comparison with wood/PP composites. In contrast to wood/polypropylene composites, the chemical modification of rapeseed straw does not influence the impact resistance of composites. The values of this parameter are within the range of 19–21 kJ/m^2.

The hardness of composite materials is shown in Figure 5.

In all the composites, the hardness values were quite comparable. However, in case of wood/PP materials an insignificantly higher hardness was noted. Esterification with acetic anhydride improves the compatibility between the rapeseed straw and the matrices but does not increase the hardness of composites.

In conclusion, the results indicate that the tensile strength of composites can not always be improved through chemical modification of lignocellulosic filler with acetic anhydride. The increment is important in the tensile strength if treated wood is used. However, the chemical modification of rapeseed straw has no effect on the tensile strength, which is always lower than that of the polypropylene matrices. The improvement of mechanical properties is most probably the result of the type of fillers used.

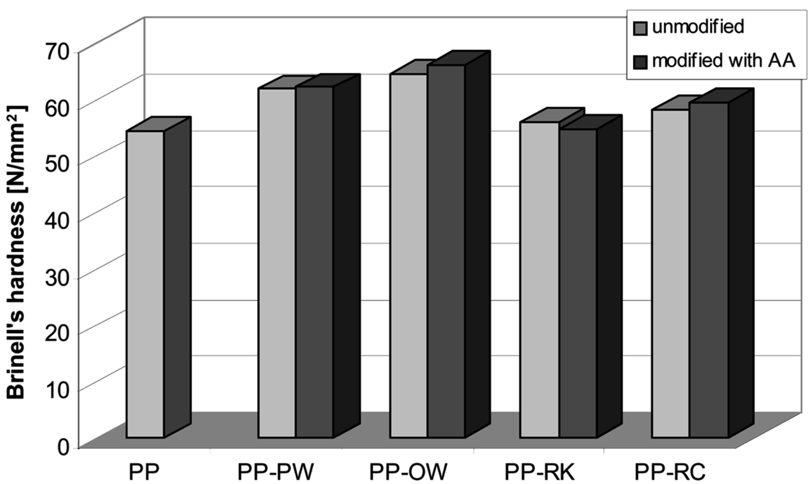

FIGURE 5 The Brinell's hardness of wood/PP and rapeseed straw/PP composites.

These observations correspond to the literature which mentions many variations in values of mechanical properties these divergences may be caused by many factors such as difference in molecular and anatomical structure as well as chemical composition of lignocellulosic materials. For example, the content of cellulose is 45–54% and 37.4–38.1% for wood and rapeseed straw, respectively. Moreover, other components like lignin have shown variation: 28–31% for wood and 19–21% for rapeseed straw [18,19].

The results concerning mechanical properties of composites indicate on possibility of production of these materials.

CONCLUSIONS

The following conclusions can be drawn from the above results.

The values of tensile strength depend on the type of lignocellulosic material as well as chemical modification used. The composites of polypropylene with modified wood particles are characterized by higher tensile stress than that of polymer matrice. The Young's modulus and impact resistance of all the investigated composites have significantly higher values than polypropylene. It is interesting to observe the effect of chemical treatment of wood fillers on important increase in the impact resistance of composites. However, the chemical modification of rapeseed straw does not change the value of this parameter.

The composites with wood or rapeseed straw are characterized by very similar values of elongation at break and hardness. The type of lignocellulosic components and chemical treatment used don't have an effect on on the above mentioned mechanical parametres.

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