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Mechanical Behaviour of Silane-Treated Wood Flour Polypropylene Composites Under Hydrothermal Conditions

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Abstract

Polypropylene wood-plastic composites were prepared with untreated and silane-treated wood flour. They were immersed in water at two different temperatures to investigate the effect of temperature on the water absorption behaviour of selected samples, with 30 wt% untreated and treated wood flour. It was found that the moisture equilibrium for materials containing 30 mass% untreated as well as those with the same content silane-treated wood flour was not reached even in 360 hours at 25°C. The maximum water absorption at 90°C for compositions based on polypropylene and 30 mass% of untreated and treated fillers was 5.07 and 6.75%, respectively. Tensile tests were conducted on the composite samples before and after immersion and the results were compared with their corresponding dry specimens. After immersion in water treated filler showed an increase in tensile strength and modulus of elasticity, while the untreated wood flour samples, higher relative elongation at break.

Keywords: polypropylene; wood flour; surface treatment; water absorption; temperature; mechanical properties.

1. Introduction

In the preparation of wood-polymer materials, it is important to analyze moisture absorption, its negative impact and ways to reduce it during the operation of the product from them. Very often, due to the hydrophilic nature of the wood fillers/polymeric matrix used and due to water absorption and hydrothermal degradation, the qualities of the composite materials deteriorate. There are several factors affecting moisture absorption in wood-polymer composites: the degree of filling, the nature of the cellulosic filler and the compatibility between the filler and the matrix [1, 2].

Various approaches have been tried to improve the compatibility between wood flour (WF) and polymers and to reduce the hydrophilicity of WF [1]. Of these, surface modification using coupling agents (silanes) as modifiers for WF is one of the most effective approaches [3–8]. Moreover, the combination of pretreatment with NaOH followed by modification with silanes, further helps to increase the strength of the bond between the polymer matrix and the wood [9]. This, in turn, makes it possible to obtain composites with improved mechanical properties.

A number of authors have reported that with an increase in the amount of wood fillers in polypropylene (PP) composites the degree of water uptake (absorption) increased and mechanical properties decreased [10–12]. The tensile properties decreased only for composites with wood flour (WF) placed in a water bath [11], compared to dry ones [10]. In ref. [12], the authors found out that the optimal mechanical and water absorption properties were achieved at 40% content of wood flakes in a PP matrix. At contents up to 40% wood flakes the tensile strength of PP composites improves, and above this content the mechanical properties deteriorate.

Wood-plastic composites made of isotactic polypropylene and different types of WF filler were obtained by Kim et al. [13, 14]. The characteristics of moisture sorption, resistance to fungal decay and mold resistance are determined. The color change of the composites after outdoor exposure was compared [13].

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The resulting materials of isotactic polypropylene and extracted and non-extracted flours from different wood species were also determined and mechanical properties, diffusion coefficient, maximum thickness swelling and thickness swelling rates [14].

Water soaking experiments of PP composites reinforced with wood powder were carried out by immersing samples at temperatures of 23, 60 and 80°C [10]. From tensile and flexural tests conducted on dry and wet composite samples, the authors concluded that the tensile strength and modulus of the composites, reinforced with 15, 30, 45 wt. % wood powder, have decreased, compared to their corresponding dry specimens. The negative effect of water molecules on the tensile properties of the composites was also confirmed by the dynamic mechanical analysis.

Wood flour filled polypropylene composites, moisture absorption and their mechanical properties were studied in a hydrothermal environment at 23, 60 and 100°C, Lin et al. [15]. It was found that modification of the matrix, content, size and surface treatment of the wood flours affected the degree of moisture absorption. For all of the composites with wood flour the tensile strength increased after immersion in water at different temperatures. The flexural strength and modulus followed a similar trend when immersed in water at 23°C. For materials immersed in water at 60 and 100°C, the opposite effect was observed.

The available data in the scientific literature on water absorption and the influence of moisture and temperature on the mechanical properties of PP composites containing WF are scarce [10, 13–15]. Considering that lignocellulosic polymer composites may be exposed to moisture or even immersed in water during their use, studies on water absorption, especially when heated at elevated temperatures, are relevant to their practical application. The study and analysis of these materials is complicated by the fact that the parameters in different investigations are variable (e.g. type and content of filler, temperature and time of immersion in water), which further complicates the comparison of the obtained results.

Thus it is important to investigate the behavior of polypropylene composite materials containing wood flour by water uptake at different temperatures and its effect on their mechanical properties. On the other hand, the use of a combination of pretreatment with NaOH followed by functionalization with vinyl-trimethoxysilane on the surface of wood flour in the preparation of polypropylene-based samples has not been reported by other authors.

For the purpose of the present work, wood flour-polypropylene composites with untreated and silane-treated wood flour were obtained. In order to investigate their behavior and the effect of water absorbed in the samples on the mechanical properties of the composites, they were immersed in water at temperatures of 25 and 90°C. The obtained results were analyzed and compared with those of the same samples before immersion in water.

2. Experimental

2.1. Materials and Methods

Previously prepared polypropylene composites with 30 mass% content of treated and untreated wood flour were used [16, 17]. PP composites with untreated WF were designated as WF30-PP, and those with treated (with NaOH and vinyltrimethoxysilane) wood flour as Na-sWF30-PP. The number 30, corresponds to 30 mass% content of filler in the polypropylene matrix.

The effect of absorbed water on the chemical structures of selected 30 mass% untreated (WF) and silane-treated (Na-sWF) samples was studied by FT-IR analysis. To determine the effect of temperature on the water absorption behavior of the samples, they were placed in distilled water at 25 and 90°C for 360 and 24 hrs, respectively. The water absorption values (*WA*, %) of the PP composites were calculated according to the equation in Ref. [18]. The tensile strength, elongation at break and Young's modulus of the materials were measured before and after immersion at the indicated temperatures, as described in our works [16, 17].

3. Results and discussion

The water absorption behavior of materials is related to the temperature of the immersion medium. Therefore, to investigate the effect of temperature on the water absorption behavior of the samples, specimens with 30 mass% content untreated (WF) and silane-treated (Na-sWF) wood flour selected in this work were immersed in distilled water at 25 and 90°C. Fig. 1 and 2 show the dependences of the amount of absorbed water, for compositions based on WF30-PP and Na-sWF30-PP, on the immersion time at a temperature 25 and 90°C, respectively.

The analysis of the experimental data shows that at room temperature, regardless of filler used, the difference in the absorption capacity and saturation time of the composites is insignificant – Fig. 1. For WF30-PP at 25°C the maximum water absorbtion (*WA*, %) is 5.13% (Fig. 1) and at 90°C it is 5.07% (Fig. 2). Values of the order of those determined by us for the moisture content for composited based on polypropylene at 30 mass% content of wood powders

(5.12%) was also confirmed by Wang et al. [10]. The lower moisture absorption of the PP composites with Na-sWF, compared to untreated WF (Fig. 1) is consistent with results reported in ref. [15]. The authors also confirm that water uptake in materials decreased upon functionalization with silanes (A-174 and A-1100) on the surface of the filler in the following order: untreated wood flour, silane-treated wood flour composites [15].

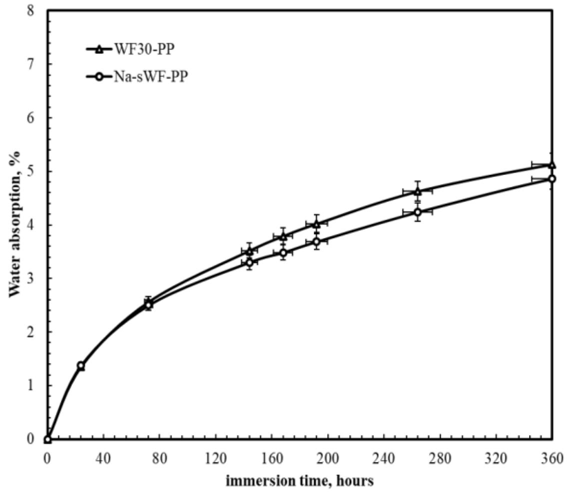

Fig. 1. Water absorption curves of WF30-PP and Na-sWF30-PP composites at 25°C

Water absorption in WF30-PP reached its saturation level, where no further water absorption was observed, for 14 hrs of immersion in water at 90°C – Fig. 2. For the specified materials, moisture equilibrium was not achieved even for 360 hrs at a temperature of 25°C – Fig. 1. Wang et al. [10] found that at a content of 45 wt. % wood powder in composite specimens and temperatures of immersion 23, 60 and 80°C, the effective moisture equilibrium takes about 1600, 1200 and nearly 400 h, respectively. The increase in water absorption can be explained by the hydrophilic nature of wood flour, as well as the possibility of forming hydrogen bonds between water molecules and –OH on the surface of WF [12, 19–21]. However, the resulting materials show significant levels of resistance to aqueous environments at 25°C [22].

For Na-sWF30-PP at 25°C the maximum water absorption (*WA*, %) is 4.86% (Fig. 1) and 6.75% at 90°C (Fig. 2). *WA* curves at 90°C are characterized by a plateau indicating the reaching of the equilibrium state – Fig. 2. Absorption of water molecules for the Na-sWF30-PP composites increased by 1.3 times compared to untreated wood flour materials. The difference in *WA*, as was established in our previous work [16] may be due to the fact that the alkaline pretreatment of wood flour creates a porous and coarse surface, that can easily absorb water. This means that

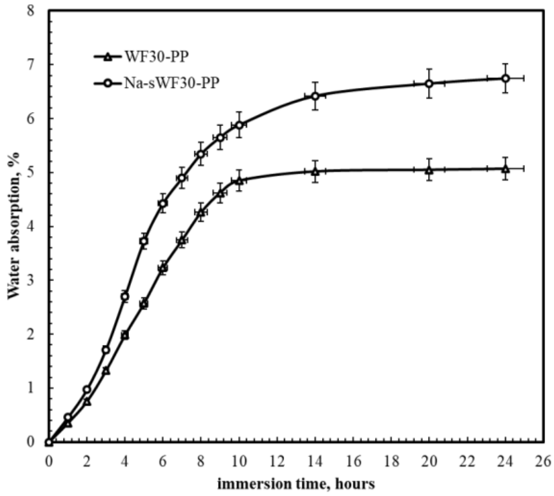

Fig. 2. Water absorption curves of WF30-PP and Na-sWF30-PP composites at 90°C

at elevated temperatures, moisture absorption is more strongly influenced by the type of filler. Effective moisture equilibrium in Na-sWF30-PP is reached after 20–22 hrs at 90°C. This indicates that increasing the ambient temperature can accelerate the diffusion rate of water molecules and water absorption. The higher temperatures, the shorter time required to reach the water diffusion equilibrium of the composites [10, 15]. Similar to the materials with 30 mass% untreated WF, for those containing the same amount of Na-sWF moisture equilibrium was not reached even in 360 hrs at a temperature of 25°C – Fig. 1.

Tables 1 and 2 summarize characteristic peaks observed in the FT–IR spectra of WF and Na-sWF, and those for polypropylene matrix. Due to the partial delignification of the WF surface as a result of the performed alkali treatment the intensities of all the registered peaks decreased and the peak at 1730 cm^{-1} is not present in the spectrum of Na-sWF [16].

Fig. 3 a and b presents the spectra of the studied WF30-PP (a) and Na-sWF30-PP (b) composites before and after immersion in water, registered in the region 4000 – 400 cm–1, in the form of foils. As can be seen from the figure, all the spectra are identical. They include both the absorption bands of the initial polypropylene (Table 2) and of the filler used (Table 1). This suggests that immersion in water and temperature do not affect the structure of the samples based on polypropylene. The bands in the region 3500 – 3000 cm–1 characteristic of valence vibrations of the –OH groups are observed for all compositions with 30 mass% WF and Na-sWF. The reason for this is the hydrophilic nature of the filler used.

Tensile tests were performed on WF30-PP and Na-sWF30-PP samples before and after immersion in distilled water at temperatures of 25 and 90°C for different periods of time. The tensile strength (σ, MPa) of samples containing 30 mass% WF was found to in-

Table 1. Characteristic peaks observed in the FT–IR spectra of WF and Na-sWF [16]

Wave length of the peaks registered, cm^{-1}	Assignment	Filler	
		WF	Na-sWF
604, 1640, 3383	off-plane vibrations, bending vibrations and stretching vibrations of the O–H fragments present in the structure of the wood flour	✓	✓
1056, 1270	C–O stretching vibrations in the structures of cellulose and lignin	✓	✓
1106, 1153	bending vibrations of C-O-C in lignin composition	✓	✓
1373, 1420	off-plane vibrations, bending vibrations of the C–H bonds in the structure of methyl ($–CH_3$) and methylene ($–CH_2$) fragments	✓	✓
1510, 1592	in-plane vibrations of C=C in the structure of aromatic rings present in the cellulose and lignin compositions	✓	✓
1730	C=O stretching vibrations of carbonyl groups taking part in lignin composition	✓	
2898	C–H stretching vibrations taking part in cellulose composition	✓	✓

Table 2. Characteristic peaks observed in the FT–IR spectra of polypropylene

Wave length, cm^{-1}	Assignment
974, 998, 1166	rocking vibrations of $–CH_3$
1375	symmetric bending vibrations of $–CH_3$
1434	symmetric bending vibrations of $–CH_2$
2843	symmetric stretching vibrations of $–CH_2$
2920	asymmetric stretching vibrations of $–CH_2$
2956	asymmetric stretching vibrations of $–CH_3$

increase from 18.53 MPa to 21.73 MPa after immersion in water for 360 hrs at 25°C – Fig. 4. At a temperature of 90°C and an immersion time of 24 hrs, the indicated parameter is lower and close in value to that of WF30-PP samples before immersion in water – 18.65 MPa. A similar tendency to decrease the tensile strength of the samples after immersion in water at elevated temperature has been reported in ref. [10]. The authors found that the tensile strength of the composites reinforced with 30 wt. % wood powder after immersion in distilled water at 60°C decreased by 17.2%, compared to their corresponding dry specimens [10].

It is known from the literature that the surface treatment of the filler or matrix [10, 22, 23] can weaken the influence of moisture and improve some of the mechanical properties of the composites [24, 25]. From the same Fig. 4 it can be seen that the tensile strength of the samples containing 30 mass% treated Na-sWF increased after immersion in water. The strength for Na-sWF30-PP before immersion is 16.86 MPa. After 360 hrs at 25°C, its value is 21.42 MPa, and after 24 hrs at 90°C the specified parameter is 19.64 MPa. The increase of the tensile strength can be

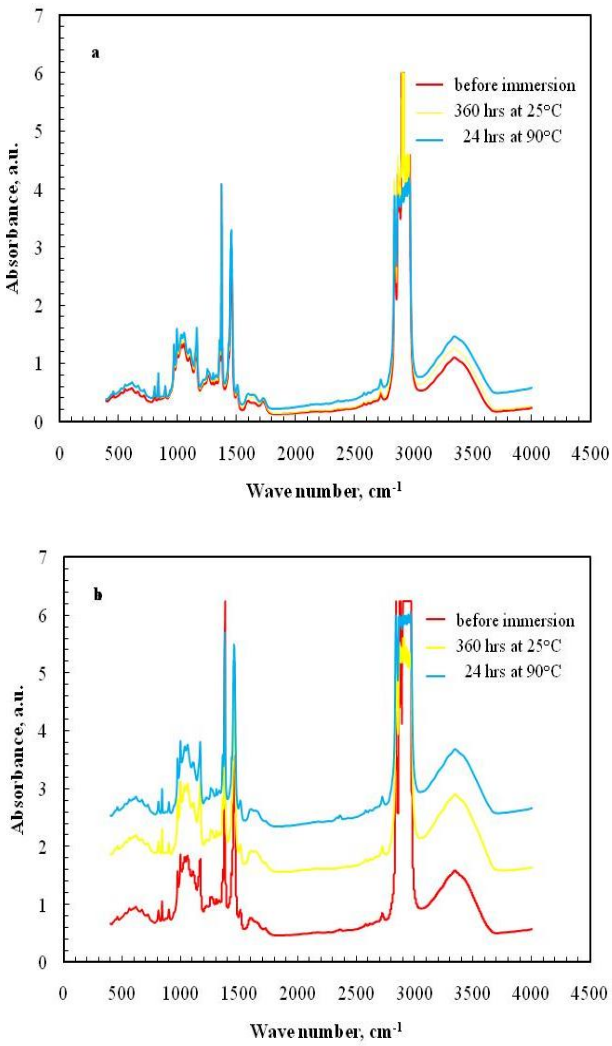

Fig. 3. FT–IR spectra of WF30-PP (a) and Na-sWF30-PP (b) composites before and after immersion in water

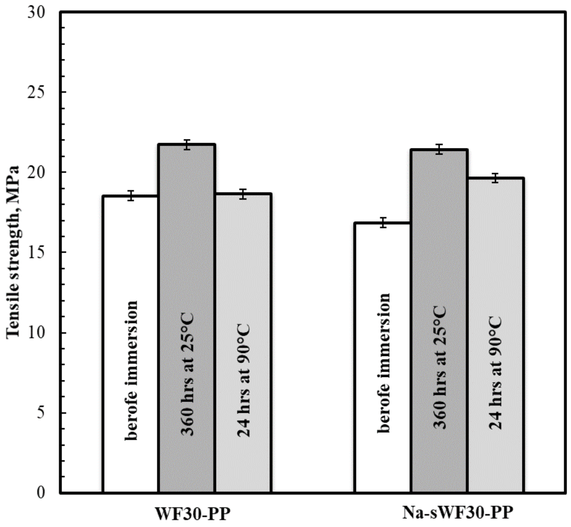

Fig. 4. Dependence of the tensile strength of WF30-PP and Na-sWF30-PP composites before and after immersion in water

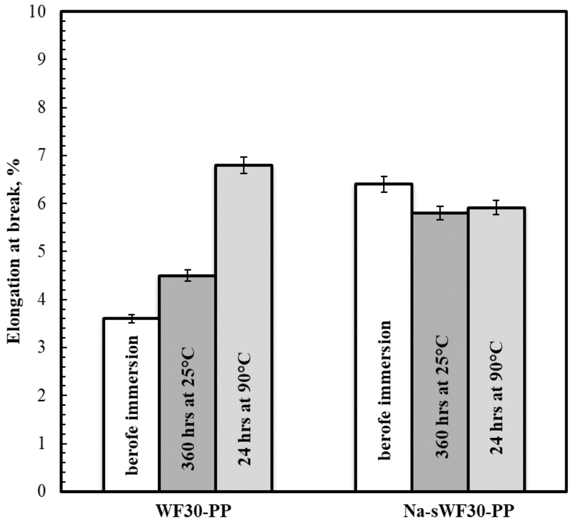

Fig. 5. Dependence of the elongation at break of WF30-PP and Na-sWF30-PP composites before and after immersion in water

attributed to the absorption of water molecules which leads to stronger filler-matrix intermolecular interactions accompanied by the formation of cross-links [26]. The observed "strengthening" effect of Na-sWF-PP - based composites at both water immersion temperatures is consistent with the results reported by Lin et al. [15], who also found that after immersion in water at different temperatures, the tensile strength of PP-wood flour composites increased, to a greater or lesser extent. Such improvement in tensile strength of all PP composites with untreated and silane-treated wood flours after immersion in water baths of different temperatures [15] as well as at a content of wood sawdust up to 40% was also found by Ferede [12]. After this content the mechanical properties of the composite are reduced.

The WF30-PP materials showed a higher elongation at break (Fig. 5) after immersion in water (24 hrs, 90°C and 360 hrs, 25°C), compared to the untreated samples (3.6%). The increase in elongation at break of the polymer composites filled with hydrophilic filler after immersion in water from 4.5% (360 hrs, 25°C) to 6.8% (24 hrs, 90°C), can be explained by the action of water molecules as a plasticizer. The claim that water can have a plasticizing effect on the polymer matrix has also been reported by other authors [10, 27, 28].

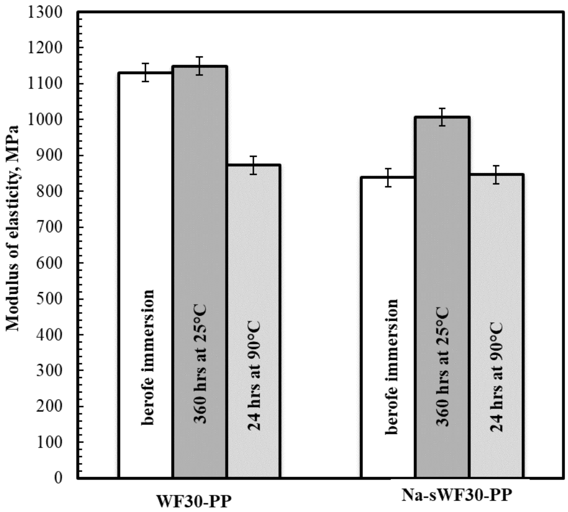

Fig. 6. Dependence of the modulus of elasticity of WF30-PP and Na-sWF30-PP composites before and after immersion in water

The introduction of Na-sWF30 into the PP matrix resulted in a decrease in the elongation at break after immersion in water at both investigated temperatures. From the results of Fig. 5, it can be seen that the sharpest decrease in elongation occurs in the Na-sWF30-PP composites after immersion in water at a temperature of 25°C – from 42.8 % (for PP matrix) to 5.8 %. After immersion for 24 hrs at 90°C, the Na-sWF30-PP composites showed slightly higher values of elongation at break.

The modulus of elasticity of all investigated composites before immersion in water increased to 1131 and 838 MPa, respectively, with the addition of WF and Na-sWF to the PP matrix – Fig. 6. For comparison, the modulus of PP under the same conditions is 756 MPa. The larger modulus can be explained by the limited mobility and deformation of the matrix with the introduction of secondary phase particles. The latter play the role of a mechanical limiter. Simultaneously, the higher modulus of WF compared to that of PP may also contribute to the observed increase [29–31].

Immersion of the composites in water further increases the modulus of elasticity. This increase is insignificant (18 MPa) for PP composites with 30 mass% WF, while for materials containing Na-sWF the increase is with 169 MPa. The modulus increased followed a similar trend with immersion in water at ambient temperature for all composites with wood flours of different contents, mesh sizes, and surface treatments as was established by other authors [15]. The reason may be that the wet wood dust can still play a role in the distribution of stresses in the specimens – from the PP matrix to the wood flour.

As the temperature increases (90°C and 24 hrs immersion in water), when water molecules penetrate into the composite material, the wood flour expands, the structure of the filler is damaged, and the modulus decreases. Despite the influence of temperature and duration of water immersion, the modulus of WF30-PP and Na-sWF30-PP composites is maintained (about 860 MPa), still greater than that of pure polypropylene.

4. Conclusions

The analysis of the experimental data showed that, at room temperature, the difference in the absorption capacity and saturation time of the composites, regardless of the filler used, was insignificant. However, the resulting materials show significant levels of resistance in an aqueous environment at a temperature of 25°C. As the temperature increases, the moisture absorption of the polypropylene-based samples is affected by the surface treatment of the filler. Water absorption for composites with 30 mass% silane-treated wood flour increased 1.3 times, compared to materials with untreated filler. Tensile tests of the compositions were performed before and after 24 hrs (at 90°C) and 360 hrs (at 25°C) of immersion in water. The obtained results were analyzed and compared with those of the same samples before immersion in water.

5. Formatting of funding sources

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6. Conflict of interest

There are no conflicts to declare.

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