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Influence of Water on Tribological Properties of Wood-Polymer Composites

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ABSTRACT

Utilization of ecological materials for appliances and products is one of the ways to achieve the goal of sustainability. Wood-polymer composites as a cheap, lightweight, durable and esthetic material has gained attention of scientists, engineers and consumers alike. Different kinds of polymeric matrices, plants used as the fillers, chemical of physical modifiers and processing technologies have already been widely studied. Nonetheless, surprisingly few information on Wood-Polymer Composites' tribology can be found. This paper is an attempt to fill this gap. Polypropylene- and poly(lactic acid)-based composites with varying wood flour content have been analyzed. The Brinell's hardness and coefficient of friction of the samples have been determined. In order to evaluate the influence of the moisture content on the tribological and mechanical properties of the composites, the samples have also been aged in water. The investigation revealed that polymeric composites filled with wood flour can present favorable coefficient of friction, compared to the neat resins. The results of our study can establish a good starting point for further investigation.

1. INTRODUCTION

Contrary to human needs, earth's resources are limited. In the era of rapidly growing population and global warming, environmental issues cannot be neglected in almost all the fields of engineering. Both the industry and researchers try to utilize renewable resources in more and more products and processes. The idea of the replacement of synthetic fibers with natural-based fillers in polymer composites has been incorporated since 1970s and has recently gained popularity [1-4]. So-called WPCs (wood-polymer composites), comprising of natural filler embedded in polymeric matrix, are widely used by automotive, construction or marine industry, electronics, packaging and even medicine [1, 3, 5]. The resulting composites are lightweight, durable, esthetic, cheap and can be processed with virtually all of the known technologies [3, 4, 6]. What's more, the research has shown [2], that they are usually preferred by environmentally-attituded consumers and their manufacturing can contribute to sustainable development of impoverished regions [7].

The WPCs have been comprehensively researched. Fillers derived from different plants [5] and various polymeric resins have been combined [1,3]. Nevertheless, a vast majority of papers focus on mechanical properties or the interactions between the fibers and the matrix [6, 8-11]. Surprisingly, the tribology of the WPCs is described to a much lesser extent. Few papers describe friction and wear of thermosetting-based resins [12-15]. Even less articles focus on tribology of thermoplastic polymers and their composites with natural fillers, such as polyoxymethylene [16,17] polypropylene [18,19] or polyethylene [20]. Nevertheless, the results are promising - apparently, an addition of wood flour can decrease the coefficient of friction in comparison to neat resin [19]. This subject needs to be fully described. If natural fibers decrease the coefficient of friction, the WPCs can be potentially used as sliding or frictional materials. This is particularly interesting, because both wood and polymers are often used in production of bearings.

High water absorption is one of the main drawbacks of natural fillers and the composites based on them [6, 11, 21, 22]. The presence of water in the material decreases its

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mechanical properties [6] and causes swelling of the specimens [21]. The influence of the moisture on the tribology of the composites should also be taken into consideration, particularly since it can act as a lubricant and decrease the coefficient of friction [15, 23].

The presented paper is a study on tribology of thermoplastic wood polymer composites. Tests of wood flour-reinforced polypropylene and poly(lactic acid) were performed. Hardness, coefficient of friction and moisture absorption of the specimens, as well as the relationship between these properties were analyzed and discussed.

2. SAMPLE PREPARATION

Two types of composites, based on Natureworks 2500 HP poly(lactic acid) (PLA) and Moplen HP 500 N isotactic polypropylene (PP) were prepared. Basic characteristic of these resins is given in the technical data sheets [24, 25]. Both polymers were filled respectively with 10, 20 and 30 wt% of Lignocell 120C wood flour (WF) (J. Rettenmaier & Söhne). Samples of neat resins were also prepared. The composites were manufactured using the melt-mixing method. Dried resins and fillers (70°C, 24h) were mixed and extruded in single screw (in case of the PP) or co-rotating twin screw (in case of PLA) extruder. The temperature of the die was set to 180°C. The extrudates were granulated using a Shini pelletizer. The PLA+WF composites were then injection molded. The runners and sprues were collected and regranulated and then processed along with the PP. Resulting granulates were dried and formed by means of compression molding into 4mm-thick plates using a laboratory hydraulic press (5 min, 180 MPa, 190°C for PLA and 200°C for PP).

3. METHODOLOGY

3.1. Water absorption

To evaluate the water absorption, the samples (two of each kind) were kept in a steel container with water for about 500h. The weight of the samples was measured before and after the immersion. The moisture content W was evaluated according to formula 1.

$$W = \frac{m_2 - m_1}{m_1} \cdot 100\% \quad (1)$$

m_1 - a sample's mass before the immersion, m_2 - a sample's mass after the immersion.

3.2. Hardness

The hardness measurements (Brinell's method) were performed according to the PN-EN ISO 2039-1 norm using the KB Prüftechnik apparatus. The load was being applied to the sample for 4 s and then maintained for 30s. The hardness of the samples was determined before and after the water absorption test, where 5 measurements were performed on each side of a sample. Mean value and standard deviation were evaluated.

3.3. Friction measurements

The coefficient of friction was measured using a ZwickRoell Z020NT testing machine. The "horizontal plane method" was

implemented. A specimen was attached to the sled, which was drawn on a glass surface. Both static and dynamic coefficients of friction were evaluated according to formulas 2 and 3.

$$\mu_s = \frac{F_s}{Q} \quad (2)$$

$$\mu_d = \frac{F_d}{Q} \quad (3)$$

where: μ_s - static coefficient of friction, F_s - the force needed to initialize the movement, μ_d - dynamic coefficient of friction, F_d - mean value of the frictional force during the movement, Q - weight of the sledge and the specimen.

The drawing speed was established to 100 mm/min and the distance was equal to 60 mm. Two samples of each kind were examined, one measurement on each side. The coefficient of friction of all the samples was determined twice before and after the immersion in water.

3.4. Microscopic observations

Surface observations were performed using an optical microscope PZO MSt 130 with a Bresser lens. The obtained pictures were digitally edited and registered using dedicated software.

4. RESULTS AND DISCUSSION

4.1. Water absorption

The moisture content values, evaluated according to the method described in the paragraph 3.1. are given in table 1.

Table 1. Moisture content W values of samples (immersion time: 500h).

Sample	PP (W values in wt%)	PLA (W values in wt%)
Neat resin	≤0.07	≤0.43
Composites	≤1.33	≤ 6.27

Even though both polypropylene and poly(lactic acid) are hydrophobic polymers [26], the moisture content of neat PP is much lower in comparison with PLA. The difference becomes more evident in case of composites. As wood particles are hydrophilic [3, 11, 26], their presence increases overall moisture content of a material. In most cases, positive relationship between the wood flour content and moisture absorption is observed [6], but a correlation between moisture content and fiber dispersion can also be found [27]. It has been suggested, that water can be transferred into a composite by means of diffusion through microvoids in the polymeric phase or through spaces on the interface between the filler and the matrix, which could act as capillaries [6, 27, 28]. An increase in water absorption can be observed even if the fibers are not highly hydrophilic [28]. In case of a lignocellulosic filler, the composite absorbs even more moisture, as it was found in our research. Small cracks or porosities caused by processing of a composite may also promote water sorption [6]. In our case, porosities, small cracks, scratches, particles of the filler and other flaws could be observed on the surfaces, which is shown in figure 1.

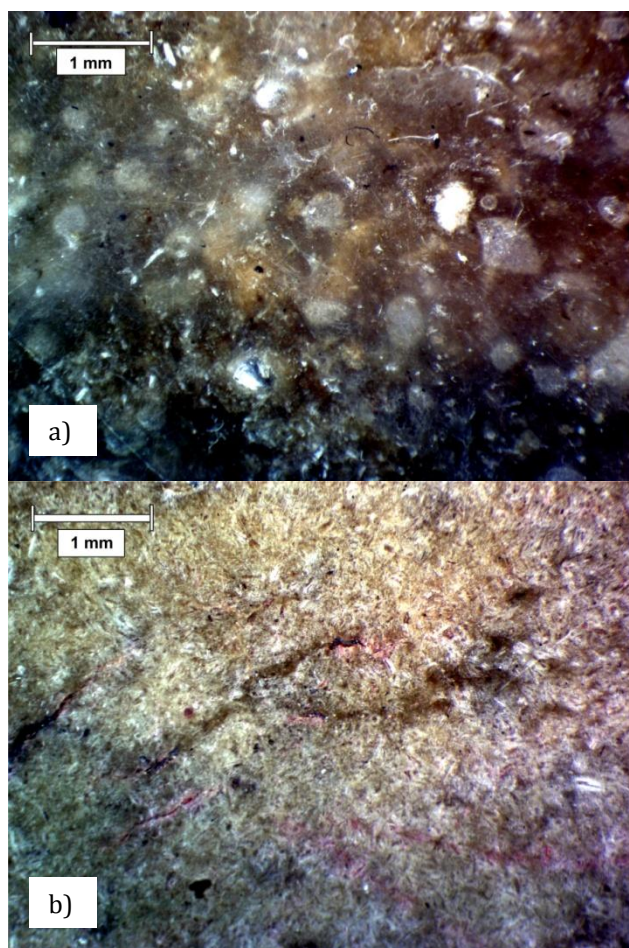


Fig. 1. Microscopic pictures of the surfaces of a) PP-based sample, b) PLA-based sample

4.2. Hardness

Hardness values plotted against the wood flour content are shown in figure 2.

It can be noticed that an addition of wood flour enhances hardness of both PP and PLA (Fig. 2). Their change is especially remarkable in case of polypropylene; the sample containing 10% of wood flour reveals a 50% higher HB, compared to the neat resin. Hardness of poly(lactic acid) also grows in function of the filler content. In some papers [10, 29], it has been stated that the addition of the lignocellulosic fibers increases hardness of polymeric composites, since they are tougher than the matrix [3, 10, 30]. On the other hand, hardness of the PP composites reinforced with 20 and 30% WF is slightly lower than that of the sample containing 10% of the filler. What's more, standard deviation values are significantly high. This effect may be explained by poor dispersion of the filler and uneven surfaces of the samples.

Hardness of all of the samples has changed due to water immersion, yet the pattern of the changes is not clear. In case of polypropylene, it can be assumed that the changes are negligible, taken into consideration measurement uncertainties. Drastic drop of the hardness of the sample containing 30% WF can be attributed to rinsing some of the filler out of the surface. Poly(lactic acid) and its composites are markedly softened by water. This behavior can be caused by the hydrolytic degradation of this polymer [31-33]. On the

other hand, time of the immersion was relatively short and it is not clear if the hydrolysis virtually occurred. Another explanation can be plasticization of the PLA by water [32]. As wood flour increases moisture absorption and promotes its diffusion into the composites, there is also a correlation between natural filler content and PLA softening. What's more, wood itself undergoes degradation in presence of water, which also has an effect on the overall strength of samples [6].

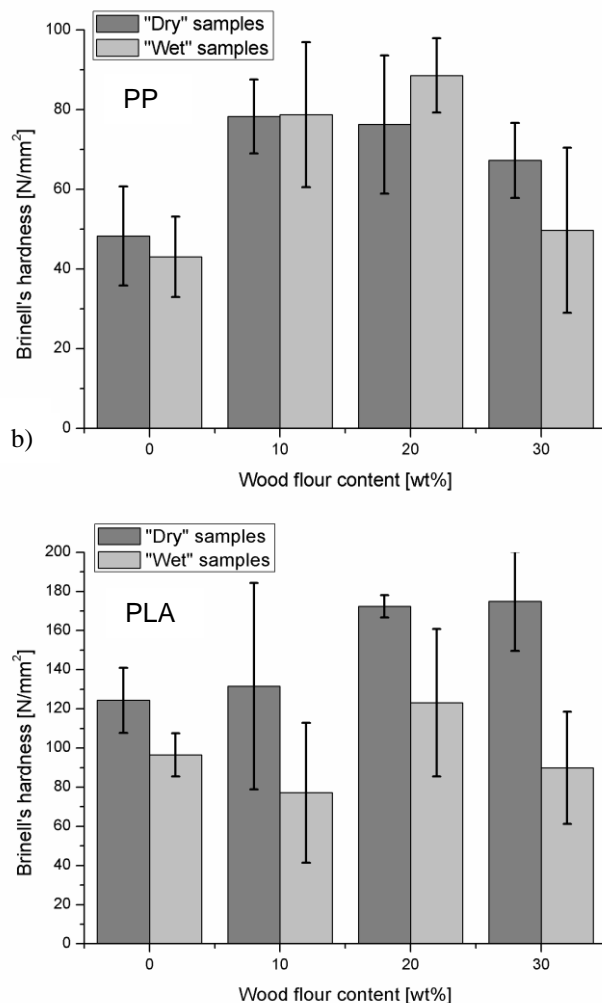


Fig. 2. Hardness values of samples with different wood flour content, a) PP-based samples, b) PLA-based samples

4.3. Friction measurements

The values of the coefficient of friction of different materials are presented in figures 3 and 4.

The small-scale friction test has proven that both neat resins and composites exhibit relatively low coefficients of friction. Static coefficient of friction of PLA (Fig. 4a) is much lower than the value of 0,32-0,37 reported by Auras, Harte and Selke in [33]. Of course, friction of polymeric composites is affected by numerous factors including load, surface geometry, sliding speed, temperature [34-36], to name a few. Because of this, values of the coefficient of friction determined in different conditions cannot be compared with each other [37]. However, it is possible to evaluate the

influence of wood flour content and moisture absorption on frictional properties of the samples.

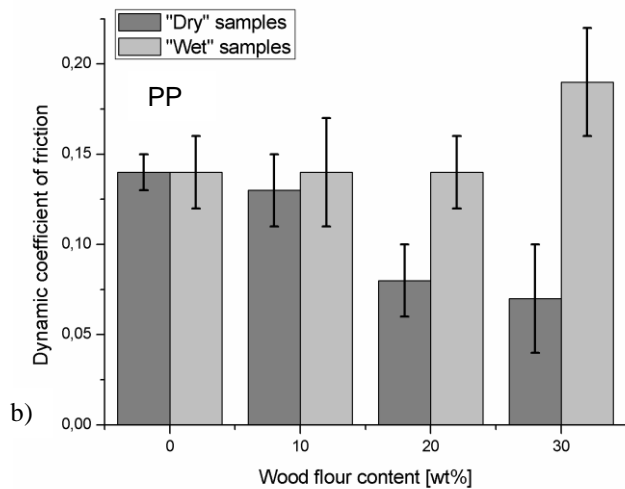
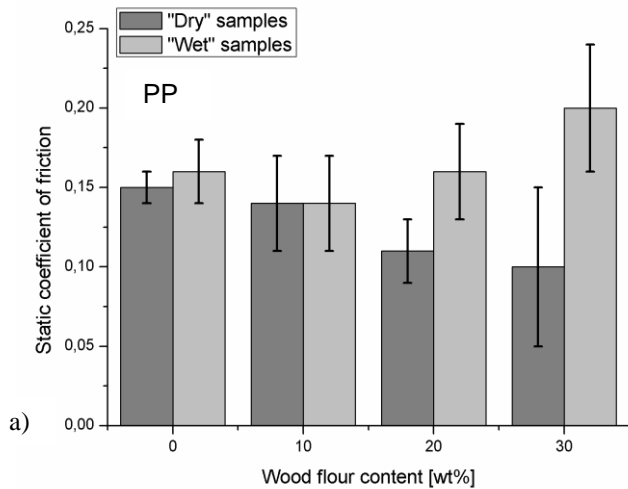


Fig. 3. Values of the coefficient of friction, a) μ_s for PP, b) μ_d for PP

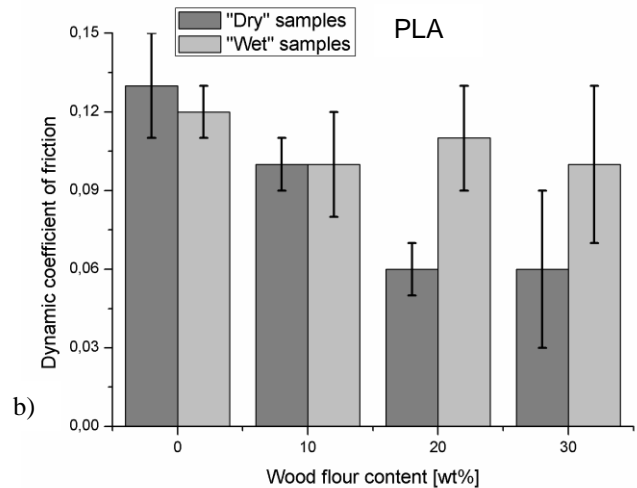
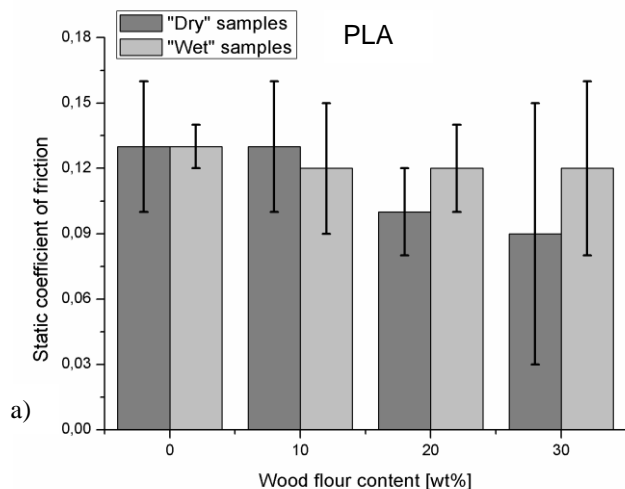


Fig. 4. Values of the coefficient of friction, a) μ_s for PLA, b) μ_d for PLA

It can be observed (Figs. 3 and 4), that both dynamic and static coefficients of friction of the samples decrease with increasing filler content. This phenomenon can be caused by different factors. Hardening of the polymeric composites by wood flour (as described in paragraph 4.2) can be one of them. It is known that hardness and friction of polymers are closely related. The harder the sample, the lower the deformation during sliding. What's more, the adhesion component of the frictional force is in inverse proportion with the strength of a body [19]. Nevertheless, the relationship between filler content (either a synthetic or an organic one) and friction of a composite is not fully understood. An addition of fibers can decrease as well as increase the coefficient of friction [20, 28, 38] and the resulting effect is believed to be determined with the interactions between the phases [38]. It is considered that poor interaction between filler and matrix leads to an increase of the coefficient of friction [28, 38]. Contrarily, wood flour-filled PP and PLA samples show lower values of μ even though adhesion of the polymeric matrix and the natural fiber is supposedly very low. Apparently, different factors need to be taken into consideration, for example differences in surface geometry or heat dissipation during the test.

Coefficient of friction changes under the influence of water immersion. In most of the cases μ values of the "wet" samples are higher than the ones of the "dry" samples. Papers indicate [15, 23, 28] that water acts as a lubricant and can drastically lower the coefficient of friction. Surprisingly, these findings does not disagree with the results of the test. Actually, the samples were not literally wet during the test, as the absorbed moisture amount did not exceed 6,3%. The change of the coefficient of friction is probably caused by structural changes of composites influenced by moisture. Both filler and matrix can be modified by aging in water. For example, wood particles usually swell [6, 21] and polymer can be plasticized by moisture, especially in case of poly(lactic acid) [6, 32]. These structural changes weaken the interaction between the phases [6], which affects the frictional behavior. For example fibers can be easily debonded during sliding process [15, 28]. An increase of the coefficient of friction caused by aging in water was observed

in case of oil palm fiber and epoxy resin composites [39] or carbon fibers and PTFE [28]. Apparently, such behavior can occur when natural as well as synthetic filler is incorporated in different matrices.

Relative change of the dynamic coefficient of friction ($\Delta\mu_D$) was evaluated using formula 4.

$$\Delta\mu_D = \frac{\mu_{i,w} - \mu_{i,d}}{\mu_{i,d}} \cdot 100\% \quad (4)$$

where: $\mu_{i,d}$ - coefficient of friction of the i -th sample, before the immersion, $\mu_{i,w}$ - coefficient of friction of the i -th sample, after the immersion.

The relationship between the relative changes of the coefficient of friction and wood flour content is shown in figure 5.

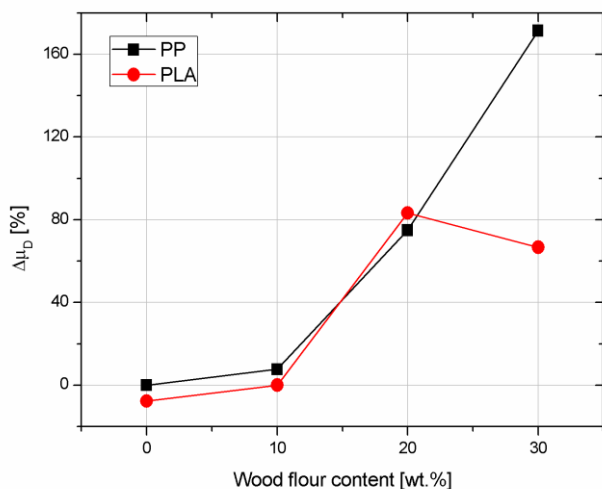


Fig. 5. The relative value of dynamic coefficient of friction as a function of wood flour content for the samples immersed in water for 500h (solid lines are drawn to guide the eyes only)

It can be noticed (Fig. 5) that the relative change of dynamic coefficient of friction due to the immersion in water % in case of PP-based samples exceeds 160. Values of $\Delta\mu_D$ for the composites containing PLA are as high as 80%. This observation proves the disadvantageous influence of water on tribological properties of WPCs.

As it follows from fig. 5, for the PP-based composites the relative change of dynamic coefficient of friction grows with increasing filler content. The lignocellulosic particles easily absorb water (as it is described in chapter 4.1.) therefore increasing amount of wood flour leads to explicit changes in tribological properties of the composites. Interestingly, this relationship does not fully apply to the PLA-based specimens. The $\Delta\mu_D$ value for the sample containing 30 wt% of the filler is smaller than in case of the composite filled with 20 wt% of WF. Apparently, the relationship between the wood flour content and the relative change of the dynamic coefficient of friction is linked to different factors, including filler distribution, residual moisture, presence of cracks or porosities on the surface of a sample and its roughness. Nevertheless, the phenomenon behind this relationship will be the topic of further investigations.

5. CONCLUSIONS

Surface properties of simple WPCs were tested. The results indicate that lignocellulosic filler hardens the polypropylene as well as poly(lactic acid)-based composites and lowers the coefficient of friction. Addition of wood flour to a polymeric resin can be highly beneficial in terms of tribology. Resulting composites can be potentially utilized as a bearing material. The results of the test are very promising, bigger scale tests should be performed to provide better insight into WPC's tribology.

Even a short-time and low-temperature immersion in water drastically changes both hardness and coefficient of friction of WPCs. The presence of the natural filler promotes moisture absorption and leads to further deterioration of mechanical properties. The increase of the coefficient of friction and decrease of hardness can be explained by a change of interaction between the filler and the matrix induced by sorption of water.

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