

WOOD-FILLED THERMOPLASTIC COMPOSITES¹

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Different physical properties of wood-filled thermoplastic materials produced by a special mixing and extrusion process are examined. The results show that the wood content and the kind of plastics are the main parameters that control the physical properties of composites. In general, wood-filled thermoplastic materials exhibit mechanical properties comparable to those of customary wood fiber products, i.e., medium density fiberboard (MDF); however, they show distinctly better behavior than the MDF and natural wood after exposure to moisture.

Introduction

Thermoplastics reinforced with special wood fillers are enjoying rapid growth due to their advantages, such as light weight, reasonable strength, and stiffness. Their processing is flexible, economical, and ecological. While there is a long history of the use of thermosetting polymeric materials, like phenol or urea formaldehyde in the production of wood composites (e.g., MDF), the use of thermoplastics as a matrix for wood particles is more recent. The selected thermoplastics must melt at or below (around 200–220°C) the degradation temperature of the lignocellulosic component [1]. This group of polymers includes polystyrene (PS), polyvinylchloride (PVC), polypropylene (PP), and high/low-density polyethylene (PE). The use of postconsumer thermoplastics, such as PP from automobile bumpers or PE from films, will help solve the pressing environmental and recycling problems.

Wood is a natural three-dimensional polymeric composite and primarily consists of cellulose, hemicelluloses, and lignin. These polymers make up cell walls of the wood and is responsible for most of its properties, even undesired ones, such as their hydrophilic nature and inherent polarity. The properties of wood, including its physical structure, mechanical properties, density, and aspect ratio (average length over diameter of the fibers) change from species to species [2].

The basic wood fillers used for the production of wood-polymer composites are wood particles and flour, as well as several types of wood pulp. To produce and separate the wood fibers, there exist different pulping processes. Chemical pulping such as the Kraft process separates the fibers by dissolving the bonding lignin, and very strong pulp, almost pure cellulose, is produced [2]. By mechanical pulping and with heat or chemical additives, wood chips are physically defibrillated. Thermomechanical pulp is of this type, where the refiner is maintained at a temperature near 120°C [13]. Wood fibers produced from mechanical pulping still retain most of its lignin and natural waxes — materials which aid the fiber dispersion in nonpolar hydrocarbon polymers [15].

The physical properties of wood-filled plastics are generally affected by the properties of their components, wood and thermoplastics, and their ratio, as well as interfacial bonding between the fiber and the matrix. The wood component influences the properties of the composite not only by the kind of wood species used, but also the kind of geometry, their aspect ratio, and the pulping process.

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Kokta et al. [3] studied the mechanical properties of wood flour-filled PP, by using aspen, spruce, and birch wood flours at different weight percentages. They found a decrease in tensile and impact strength with increasing wood content, while the Young's modulus linearly increased. The Young's modulus of composites filled by 30 wt.% of wood flour ranged from 1.7 GPa for birch wood flour to 1.9 GPa for aspen. Experiments performed by varying the average fiber length from 0.5 to 0.9 mm showed an increase in the Young's modulus with increasing fiber length, while the tensile strength was not significantly affected.

Wood fibers as reinforcing agents have also been studied in [7-14]. It was found that the mechanical properties of wood fiber-filled composites depended on the pulping process and the fiber length. Short and tiny fibers (average particle size 0.24-0.5 mm) should be preferred. They provide a higher specific surface area [4-7] and the fibers are distributed more homogeneously as compared to composites with long fibers, which improves the compatibility of the fiber and the matrix.

In [16], HDPE composites filled with wood flour, mechanical pulp, and cellulose pulp were compared. The highest strength values were observed for wood flour-filled composites, as a result of better dispersion and adhesion to the matrix of wood flour due to its higher lignin content, while the HDPE-cellulose fiber composites showed the lowest strength.

The interface between fiber and matrix considerably influences the mechanical properties of reinforced composites — it determines the ability of transferring the stresses from the matrix to the fiber and finally the strength of the compound. In most cases of nontreated filler composites, the tensile strength and elongation are lower as for unfilled composites [4-9]. The characteristic values decrease with increasing filler content (elongation is significant). Some tests [13, 15, 17] show that up to a critical content of the filler (20-40 for PP, 30-40 for PE, and 30% for PS), the strength of the composite increases, compared with polymers without fillers. Such a discrepancy may result from different aspect ratios of the wood fillers used.

Processing

A wood-filled thermoplastic was produced according to a special processing system (US Patent 4,505,869 by S. Nishibori, March 19, 1985), which allowed for the production of composites with up to 75 wt.% of the wood content. A wood powder was made by pulverizing the waste lumber of different species. The matrices were postconsumer plastics, such as automobile bumpers and LDPE or PVC films of agricultural applications. The processing comprised drying of the wood powder, mixing, and finally extrusion molding.

First, the wood powder was dried to a moisture content below 0.1 wt.%. Simultaneously, color and flame retardant pigments were mixed with the wood powder. After drying, plastics were added and melted in a mixer; the plastics wet well the exceptionally dry wood powder. Then, the mixture was extruded to the mold in a highly viscous state by a special screw designed to prevent heat generation. Brake rolls at the end of the mold pressed and solidified the highly viscous melt under high pressure. Up to 12 mm thick and 600 mm wide samples could be produced. In our case, samples of 4 mm thickness were produced, which were cut parallel to the extrusion direction, with dimensions according to DIN standards.

Experimental

The mechanical and other physical properties of variously formed wood-filled thermoplastic composites, differing in their wood content as well as in the kind of thermoplastics, were characterized and compared with those of Ponderosa pinewood and MDF. An MDF with a resin content of about 10 wt.% was used. The mechanical and physical properties were investigated at room temperature (23°C) at a relative humidity of 50%. In each case, 8 to 10 specimens were tested. Tensile and flexural tests for wood-filled thermoplastic composites were carried out with a test speed of 2 mm/min according to DIN 53455 and 53452, respectively. For the pinewood and MDF, the tests were carried out according to DIN 52377 and 52362, respectively. The Charpy impact energy was measured according to DIN 53453. Standard deviations for all materials and test methods were less than 10% of the mean value. To determine the influence of moisture on the mechanical

TABLE 1. Mechanical Properties of Wood-Filled Thermoplastic Composites, Natural Wood (Ponderosa Pine), and Medium Density Fiberboard (MDF)

Material, wt. %	Flexural modulus, GPa	Flexural strength, MPa	Young's modulus, GPa	Tensile strength, MPa
Wood (55), PP (45)	2.3	41.2	2.6	21.4
Wood (55), PE (45)	2.3	28.4	2.4	16.2
Wood (50), PVC-soft (50)	0.5	14.5	0.5	7.1
Wood (50), PVC-soft (25), -rigid (25)	2.7	40.0	2.6	21.9
Natural wood (Ponderosa pine)	9.4	87.9	10.1	43.6
Medium density fiberboard (MDF)	3.9	36.7	3.1	17.0

TABLE 2. Effect of Wood Flour Content on the Mechanical Properties of Wood-Filled PVC-Soft

Wood flour content	35 wt. %	50 wt. %
Young's modulus, MPa	190	520
Tensile strength, MPa	5.4	7.1
Flexural modulus, MPa	250	554
Flexural strength, MPa	8.7	14.5
Charpy impact energy, mJ/mm ²	10.3	3.7

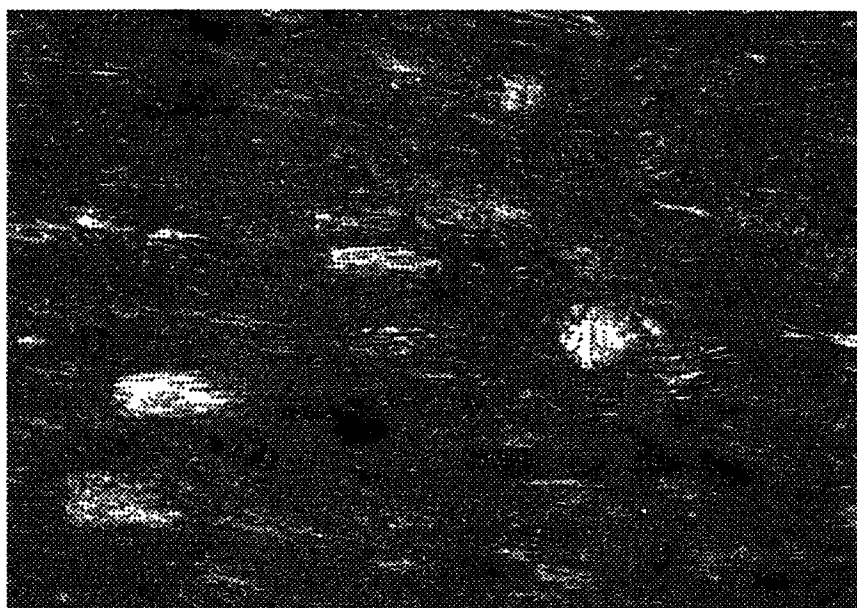


Fig. 1. Grinding of a wood-filled PVC-soft sample (wood content of 50 wt.%) parallel to the extrusion direction.

properties, samples were stored in distilled water for 28 days and afterwards tested in the tensile and flexural mode. For the thickness swelling and water absorption measurements according to DIN 52351, the specimens were immersed in distilled water in a horizontal position for 28 days at 23°C.

Results and Discussion

The mechanical properties of wood-filled thermoplastic composites depend on the kind of thermoplastics and wood used, their ratio, as well as interaction between the filler and the matrix. In Table 1, some mechanical properties of different thermoplastics filled by 50 and 55 wt.% are listed. It can be seen that the

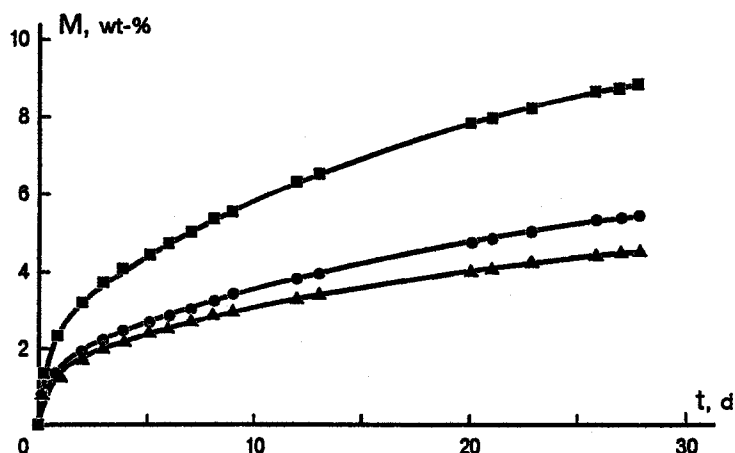


Fig. 2. Moisture absorption M versus time t for different wood-filled PVC samples: 50 wood and 50 wt.% PVC-soft (■); 50 wood, 25 PVC-soft, and 25 wt.% PVC-rigid (●); 35 wood and 65 wt.% PVC-rigid (▲).

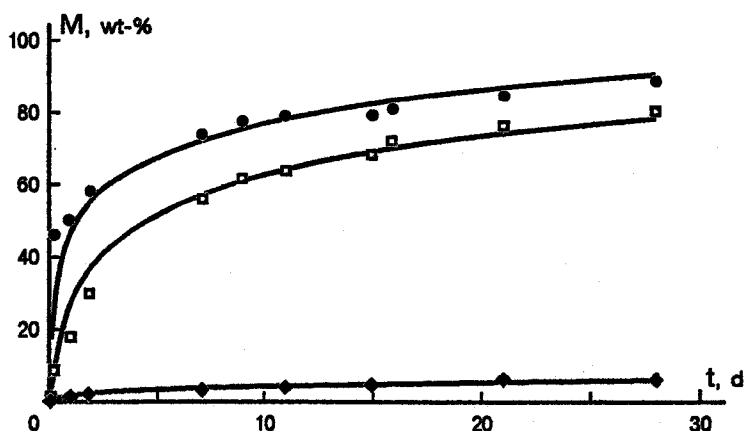


Fig. 3. Moisture absorption M versus time t for wood-filled PVC (◆) during 28 days storage in water, compared with pinewood (●) and MDF (□).

highest strength and stiffness values are reached with wood-filled PP, while PVC-soft shows the lowest values. Replacing half of the PVC-soft content by PVC-rigid, the Young's modulus increases fivefold and the tensile strength threefold (Table 1). The mechanical behavior of wood-filled thermoplastic composites also depends on the wood content. Table 2 shows that by increasing the wood content from 35 to 50 wt.%, the tensile strength and Young's modulus can be increased about 50 and 150%, respectively. The Charpy impact strength, however, decreases significantly with increasing wood content.

During extrusion, with the wood fillers oriented parallel to the extrusion direction, as shown in Fig. 1, the wood-filled plastic composites exhibited anisotropic behavior. Tensile tests were carried out parallel and transverse to the extrusion direction. The anisotropy coefficients ranged from 0.6 to 0.8, depending on the kind of material used.

To compare the mechanical properties of different wood-filled thermoplastic composites with other standard wood materials, Ponderosa pinewood and medium density fiberboards were used. The comparison of the results (Table 1) shows that wood-filled thermoplastic composites with a wood content of 50 to 55 wt.% exhibit approximately similar mechanical properties as the customary MDF. However, the mechanical properties of natural wood, which result from its fiber structure, cannot be attained with these kinds of composites.

The hygroscopic behavior of wood-filled thermoplastics is distinctly influenced by their wood content and the kind of thermoplastics. In Fig. 2, the moisture absorption of PVC-soft filled by two different wood contents

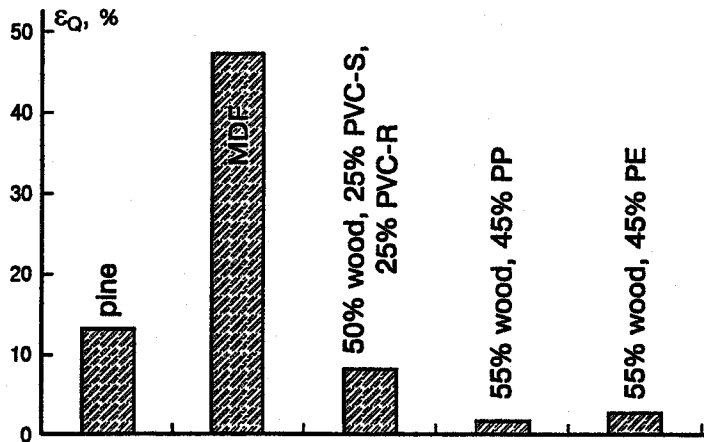


Fig. 4. Swelling due to moisture ϵ_0 of wood-filled plastics after 28 days storage in water, compared with pinewood and MDF.

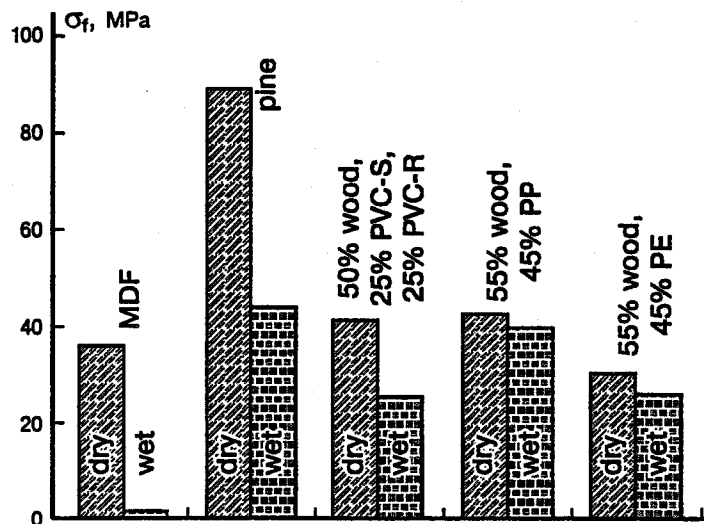


Fig. 5. Influence of moisture on the flexural strength σ_f of wood-filled plastics, compared with pinewood and MDF.

is depicted. It can be seen that the composite containing 35 wt.% wood powder absorbs 50% less moisture than the composite filled by 50 wt.% powder. Concerning the influence of the kind of thermoplastics on the moisture absorption, the investigations with different wood-filled thermoplastics with a wood content of 50 wt.% show that the moisture absorption of the wood-filled plastic composites ranges from 3, for wood-filled PP, to 9 wt.% for wood-filled PVC-soft, after storing the samples for 28 days in distilled water. Replacing half of the PVC-soft content by PVC-rigid, the moisture absorption can be decreased by 40%, as shown in Fig. 2.

Compared to pinewood and MDF, wood-filled thermoplastic composites are distinguished by their better hygroscopic behavior. While the moisture absorption of thermoplastics filled by 50% of wood powder ranged from 3 (PP) to 9 wt.% (PVC-soft), the moisture content of the MDF and pine wood amounted to 80 and 90 wt.%, respectively, after 28-day storage (Fig. 3). The small moisture absorption of wood-filled thermoplastic composites is associated with extremely low swelling, due to moisture, which amounts to only 2% for wood-filled PP after 28-day storage (Fig. 4). Furthermore, the mechanical properties of all the materials are affected by moisture, as shown in Fig. 5, in the case of flexural strength. While the flexural strength of the MDF and pine wood is reduced by 95 and 50%, respectively, for wood-filled thermoplastic composites it decreased only by 8 (PP) to 35% (PVC) during a 28-day storage in water.

Conclusions

The physical properties of wood-filled thermoplastic materials produced by a special mixing and extrusion process have been examined. The results show that the wood content and the kind of plastics are the main parameters controlling the physical properties of composites. By increasing the wood content of wood flour-filled PVC-soft from 35 to 50 wt.%, the tensile strength and Young's modulus can be increased about 50 and 150%, respectively. The Charpy impact strength, however, significantly decreases with increasing wood content.

A comparison of the experiments of the Ponderosa pine and MDF showed that wood-filled thermoplastic materials exhibited mechanical properties comparable to those of the customary wood fiber product — MDF. However, they showed distinctly a better behavior than the MDF and natural wood after exposition to moisture. After storing the samples for 28 days in water, the moisture absorption of wood-filled plastics ranged from 3 to 9 wt.%, depending on the kind of thermoplastics, whereas the pinewood and MDF absorbed 80 and 90 wt.%, respectively. The mechanical properties of the wood-filled thermoplastic samples stored in water decreased by only 8 to 35%, while the characteristic values of the MDF and pinewood reduced by 95 and 50%, respectively.

REFERENCES

1. R. W. Rowell, *Paper and Composites from Agro-Based Resources*, Lewis Publishers, Boca Raton (1997).
2. A. K. Bledzki, S. Reihmane, and J. Gassan, "Thermoplastics reinforced with wood fillers. A literature review," *Polym.-Plast. Technol. Eng.* (in press).
3. B. V. Kokta, R. G. Raj, and C. Daneault, "Use of wood flour as a filler in polypropylene: Studies on mechanical properties," *Polym.-Plast. Technol. Eng.*, **28**, No. 3, 247-259 (1989).
4. D. Maldas, B. V. Kokta, and C. Daneault, "Composites of polyvinyl chloride-wood fibers. IV. Effect of the nature of fibers," *J. Vinyl Technol.*, **11**, No. 2, 90-99 (1989).
5. S. N. Maiti and K. Singh, "Influence of wood flour on the mechanical properties of polyethylene," *J. Appl. Polym. Sci.*, **32**, 4285-4289 (1986).
6. K. L. Yam, B. K. Gogoi, C. C. Lai, and S. E. Selke, "Composites from compounding wood fibers with recycled high density polyethylene," *Polym. Eng. Sci.*, **30**, No. 11, 693-699 (1990).
7. M. M. Sain, C. Imbert, and B. V. Kokta, "Composites of surface treated wood fiber and recycled polypropylene," *Angew. Makromol. Chem.*, **210**, 33-46 (1993).
8. R. G. Raj, B. V. Kokta, and C. A. Daneault, "A comparative study on the effect of aging on mechanical properties of LDPE-glass fiber, mica, and wood fiber composites," *J. Appl. Polym. Sci.*, **40**, 645-655 (1990).
9. D. Maldas and B. V. Kokta, "Performance of hybrid reinforcements in PVC composites. Pt. I. Use of surface-modified mica and wood pulp as reinforcements," *J. Test. Eval.*, **2**, 68-72 (1993).
10. R. G. Raj, B. V. Kokta, D. Maldas, and C. Daneault, "Use of wood fibers in thermoplastics. VII. The effect of coupling agents in polyethylene-wood fiber composites," *J. Appl. Polym. Sci.*, **37**, 1089-1103 (1989).
11. M. M. Sain and B. V. Kokta, "Polyolefin-wood filler composite. I. Performance of m-phenylene bismaleimide-modified wood fiber in polypropylene composites," *J. Appl. Polym. Sci.*, **54**, 1545-1559 (1994).
12. P. Hedenberg and P. Gatenholm, "Conversion of plastic/cellulose waste into composites. I. Model of the interface," *J. Appl. Polym. Sci.*, **56**, 641-651 (1995).
13. R. T. Woodhams, G. Thomas, and D. K. Rodgers, "Wood fibers as reinforcing fillers for polyolefines," *Polym. Eng. Sci.*, **24**, No. 15, 1166-1171 (1984).
14. D. Maldas, B. V. Kokta, and C. Daneault, "Influence of coupling agents and treatments on the mechanical properties of cellulose fibre-polystyrene composites," *J. Appl. Polym. Sci.*, **37**, 751-775 (1989).
15. R. Simpson and S. Selke, "Composite materials from recycled multi-layer polypropylene bottles and wood fibers," *Polymer Prepr., Amer. Chem. Soc., Div. Polym. Chem.*, **32**, 148-149 (1991).
16. R. G. Raj, B. V. Kokta, G. Grouleau, and C. Daneault, "The influence of coupling agents on mechanical properties of composites containing cellulosic fillers," *Polym.-Plast. Technol. Eng.*, **29**, 339-353 (1990).
17. B. V. Kokta, R. Chen, C. Daneault, and J. L. Valade, "Use of wood fibers in thermoplastic composites," *Polym Compos.*, **4**, No. 4, 229-232 (1983).