EFFECTS OF WOOD FIBER CHARACTERISTICS ON MECHANICAL PROPERTIES OF WOOD/POLYPROPYLENE COMPOSITES

Nicole M. Stark

Chemical Engineer
U.S. Department of Agriculture
Forest Service
Forest Products Laboratory[1]
Madison. WI 53705-2398

and

Robert E. Rowlands

Professor
Mechanical Engineering Department
3348 Engineering Hall
University of Wisconsin
Madison, WI 53706

(Received March 2002)

ABSTRACT

Commercial wood flour, the most common wood-derived filler for thermoplastics, is produced in a mixture of particle sizes and generally has a lower aspect ratio than wood and other natural fibers. To understand how wood flour and fiber characteristics influence the mechanical properties of polypropylene composites, we first investigated the effect of different sizes of wood flour particles on the mechanical properties of wood-flour-filled polypropylene composites. We then compared the properties of wood-flour-filled composites to those of composites reinforced with refined wood fiber. We also studied the effect of a maleated polypropylene coupling agent on composite properties. Wood flour particles (35, 70, 120, and 235 mesh) were compounded at 40% by weight with polypropylene. Increases in tensile and flexural strength and modulus of the wood flour composites were found to correspond with increases in aspect ratio. Notched impact energy increased with increasing particle size, whereas unnotched impact energy decreased with increasing particle size. Refined wood fiber and 40-mesh wood flour was compounded at 20% and 40% by weight with polypropylene. Wood fiber resulted in higher strengths at both filler levels and higher moduli at the 40% level compared to the strength properties of wood flour composites. The higher aspect ratio of the wood fiber had little effect on impact energy. The maleated polypropylene coupling agent caused greater strength increases in wood fiber composites than in wood flour composites. The coupling agent did not significantly1 affect tensile or flexural moduli. Our results clearly support the use of higher aspect ratio wood fibers and coupling agents for increasing the strength of wood/plastic composites.

Keywords: Wood flour, wood fiber, polypropylene, mechanical properties, aspect ratio, coupling agent.

INTRODUCTION

The use of natural fibers as additives for plastics has been rapidly expanding. A 50% growth in the use of natural fibers in the plastics industry is forecast for 2000-2005 (Eckert 2000). Natural fibers generally refer to ligno-

[1] The Forest Products Laboratory is maintained in cooperation with the University of Wisconsin. This article was written and prepared by U.S. Government employees on official time and is therefore in the public domain and not subject to copyright. The use of trade or firm names in this publication is for reader information and does not imply endorsement by the U.S. Department of Agriculture of any product or service.

cellulosic materials derived from wood or agricultural materials such as kenaf, jute, hemp, flax, or other natural resources. Natural fibers are available in many different forms and produce different properties when added to thermoplastics (Sanadi et al. 1995; Zaini et al. 1996). Natural fibers may be used in the form of particles, fiber bundles, or single fibers, and may act as a filler or reinforcement for plastics (Osswald 1999).

One of the most common natural fibers used in the thermoplastics industry is wood flour, which is produced commercially from post-industrial sources such as planer shavings and sawdust. The scrap wood is sourced for species purity and then ground to specific particle size distributions. One variable used to characterize wood flour is mesh size, the size of the screen used to differentiate between particle sizes. The specific but broad particle size distribution in commercial wood flour varies among manufacturers. In addition, particle sizes of wood flour from a single commercial supplier may overlap between available mesh sizes. Extra screening of wood flour, for example screening out fines to narrow the particle size distribution, raises the cost of the flour. Therefore, typical commercial grades include a mixture of particle sizes. Because of this size overlap, particularly at larger mesh sizes (smaller particle sizes), it is difficult to characterize the properties of commercial wood flour/plastic composites on the basis of specific particle sizes.

In general, wood flour is used as a filler for plastic, which tends to increase the stiffness of the composite but does not improve its strength. Natural fibers can be used to reinforce rather than fill plastics, which increases strength as well as stiffness. Wood and other lignocellulosic fibers typically have higher aspect ratios than that of wood flour. Furthermore, the aspect ratio of the fiber can be increased. At a critical fiber length, stress is transferred from the matrix to the fiber, resulting in a stronger composite. Stress is efficiently transferred only if the bond between the matrix and fiber is good (Osswald 1995, 1999; Osswald and Menges 1995). It is often necessary to include a coupling agent in the composite to ensure a strong bond. A coupling agent improves the bond between the plastic matrix and natural fiber by chemical and physical means (Gauthier et al. 1999).

TABLE 1. *Standard mesh sizes used to classify wood flours.*

Standard mesh size[a]	Mesh size range		Screen hole size (mm)		Particle size (mm)
	Minimum	Maximum	Minimum	Maximum	
35	40	30	0.425	0.600	0.513
70	80	60	0.180	0.250	0.215
120	140	100	0.106	0.150	0.128
235	270	200	0.053	0.075	0.064

[a] Average of corresponding mesh size range

The objectives of this study were: (1) to investigate the influence of wood flour particle size on mechanical properties of wood flour/polypropylene (PP) composites, and (2) to compare mechanical properties of wood-flour-filled and wood-fiber-reinforced PP composites in relationship to aspect ratio and coupling treatment.

EXPERIMENTAL METHODS

Wood flour/PP composites

Ponderosa pine wood flour was screened by American Wood Fibers (Schofield, Wisconsin) using designated mesh sizes to produce four distinct wood flour particle size samples. The standard mesh notations and corresponding screen sizes are shown in Table 1. For example, a designated 35-mesh wood flour corresponds to particles that pass through a 30-mesh screen, but not a 40-mesh screen. The PP was Fortilene 3907 (Solvay Polymers, Inc., Deer Park, Texas), an injection molding grade homopolymer with a melt index of 36.5 g/10 min. The screened ponderosa pine wood flour was compounded with PP at 40% by weight.

After the wood flour was dried, it was dry-blended with PP and compounded using a 32-mm Davis Standard (Pawcatuck, Connecticut) co-rotating, intermeshing twin-screw extruder. The melt temperature was kept below 190°C

TABLE 2. *Summary of mechanical properties of polypropylene composites filled with 40% wood flour.*

	Tensile properties			Flexural properties		Izod impact energy	
Mesh size	Strength (MPa)	Modulus (GPa)	Elongation (%)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
35	21.7	3.20	2.27	38.7	2.69	22.3	54
70	25.5	3.61	2.27	42.6	3.15	19.7	79
120	24.9	3.47	2.29	42.9	3.00	18.7	84
235	24.3	3.46	2.1 1	41.4	2.89	16.0	91
COV (%)[b]	1	5	7	1	2	7	11

[b] Average COV for one standard deviation.

to prevent degradation of the wood flour. The extrudate, in the form of strands, was cooled in a water trough and pelletized. The resulting pellets were dried at 105°C for 24 h before being injection molded into ASTM test specimens. All materials were injection molded using a 33-ton Cincinnati Milacron (Batavia, Ohio).

Wood fiber/PP composites

Wood fiber was derived from recycled pallets as hardwood chips (Woodcycle, Inc., Sullivan, Wisconsin), ranging from approximately 2.5 to 12.5 cm long and 1.3 to 3.8 cm wide. The chips were reduced in size by a hammermill using a 3.175-cm screen. The resulting chips were reduced to fibers using a laboratory-scale 300-mm single-disk Sprout–Bauer (Andritz, Inc., Springfield, Ohio) pressurized refiner. The incoming steam pressure was 414 kPa, and the plate gap width was maintained at 0.127 mm. The fibers were then hammermilled through a 0.432-mm screen, to correspond to the size of commercial 40-mesh wood flour. American Wood Fibers supplied 40-mesh ponderosa pine wood flour for comparing properties of wood-flour-filled composites and wood-fiber-reinforced PP composites. Both the 40-mesh wood flour and screened wood fiber contained fines. The PP for the wood fiber composites was the same as that used for the mesh size studies. A coupling agent, maleated polypropylene (MAPP) (Unite MP 880, Aristech, Pittsburgh, Pennsylvania), was added to each composite material to improve interfacial adhesion and to allow for comparison between composites with and without a coupling agent.

The composite materials were dry-blended and compounded at 20% or 40% by weight wood fiber. The composites were made with or without MAPP at 3% by weight of the total composite. This resulted in a blend of 20% or 40% wood fiber, 0 or 3% MAPP, and the remainder PI? The composite materials were compounded and molded using the same procedures used for the wood flour composites.

Testing

Tensile tests were conducted according to ASTM D638 (ASTM 2000a), flexural tests according to ASTM D790 (ASTM 2000b), and notched and unnotched Izod impact strength tests according to ASTM D256 (ASTM 2000c). Five replicates were run for each test. An optical microscope equipped with a camera was used to examine wood flour and wood fiber characteristics. A scanning electron microscope was used to examine the impact fracture surfaces, coated with gold, of selected specimens.

RESULTS AND DISCUSSION

Wood flour/PP composites

In the following discussion, wood flours are referred to by their mesh size (see Table 1). Results of tensile, flexural, and impact tests on composites made with different sizes of wood particles are presented in Table 2. Table 3 shows the results of Tukey's pairwise comparison, at a = 0.05, of various properties of

TABLE 3. *Results of Tukey pairwise comparison at* α = *0.05 for polypropylene composites filled with 40% wood flour of different mesh sizes.* [1]

Property	Wood flour mesh size			
	35	70	120	235
Tensile strength (MPa)	—	—	—	—
Tensile modulus (GPa)	—	----------	----------	----------
Tensile elongation (%)	----------	----------	----------	----------
Flexural strength (MPa)	—	----------	----------	—
Flexural modulus (GPa)	—	—	—	—
Notched impact (J/m)	—	----------	----------	—
Unnotched impact energy (J/m)	—	----------	----------	----------

[a] A continuous line indicates no statistical difference between properties of a particular mesh size composite and those around it

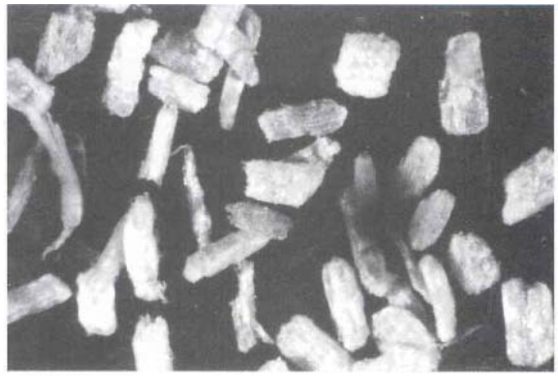

FIG. 1. Screened 35-mesh wood flour.

wood flour composites. A continuous line indicates that there is no statistical difference between the properties of a particular mesh size composite and those around it. For example, the tensile modulus reported for the 70-mesh composite is statistically different from that of the 35-mesh composite, but not from that of the 120-mesh composite.

The highest tensile strength was observed for composites containing 70-mesh wood flour and the lowest tensile strength for those with 35-mesh wood flour. Tensile modulus was lowest in composites with the largest particles (35 mesh). However, there is no statistical significance in tensile modulus between the other particle sizes. Flexural strength reached a maximum in the 70- and 120-mesh wood flour composites, but the difference between these composites was not statistically significant. Flexural modulus also reached a maximum in the 70-mesh wood flour composites. Notched impact energy generally increased with smaller mesh size (i.e., larger particle size), although the difference between the 70- and 120-mesh wood flour composites was not statistically significant. In contrast, unnotched impact energy appeared to increase with larger mesh sizes (i.e., smaller particle sizes). Indeed, the lowest unnotched impact energy occurred with the smallest mesh size. However, there was no statistical difference in unnotched impact energy between wood flour composites with 70-, 120-, and 235-mesh wood flour.

The nonlinear trends observed for the mesh size study can be explained by observing geometric characteristics of the wood flour. Figures 1 and 2 show 35- and 70-mesh wood flour, respectively, at the same magnification. The aspect ratio of the 70-mesh wood flour was notably higher than that of the 35-mesh wood flour. The particles associated with both of these meshes had roughly the same length, but the 70-mesh particles were much smaller in diameter. Table 4 shows 'average aspect ratios for commercial ponderosa pine flour. The aspect ratio of this flour ranges from 3.3 to 4.5; the highest aspect ratios are for 80- to 120-mesh flours. These data are reflected in our study, where maximum strength and modulus occurred in composites filled with 70-mesh wood flour.

The energy needed to cause dynamic failure of wood flour composites was measured using

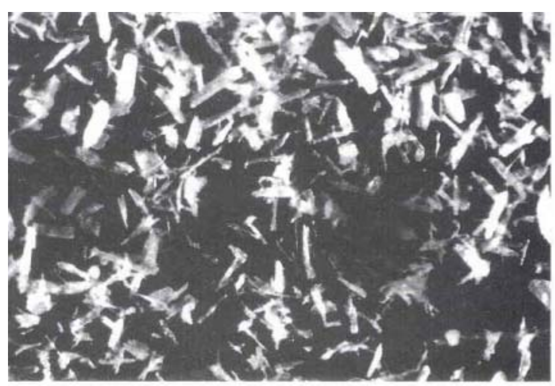

FIG. 2. Screened 70-mesh wood flour.

TABLE 4. *Aspect ratio (length to diameter ratio) of commercial ponderosa pine wood flour.*

Mesh size	Aspect ratio
20	3.53
40	3.35
60	4.23
80	4.52
100	4.53
120	4.11
140	3.73

Aspect ratio data courtesy of American Wood Fibers, Schofield, WI.

Izod impact tests. The energy required for crack propagation was measured with a notched Izod specimen, and the energy required for crack initiation was measured with an unnotched Izod specimen. Crack propagation occurred at the PP/wood flour interface as a result of the poor interface between the hydrophilic wood flour and the hydrophobic PP. Consequently, composites made with larger wood flour particles had higher notched impact capacity (i.e., higher critical crack propagation energy) as a result of the increase in fracture surface area. Conversely, unnotched impact energy (minimum energy needed to initiate a crack) decreased with increasing particle size. The wood flour in the PP matrix provided stress concentrations, therefore providing sites for crack initiation. The larger the wood flour particle, the larger the stress concentrations along the naturally weak interface of the hydrophilic wood flour and hydrophobic PP, and the lower the unnotched impact energy.

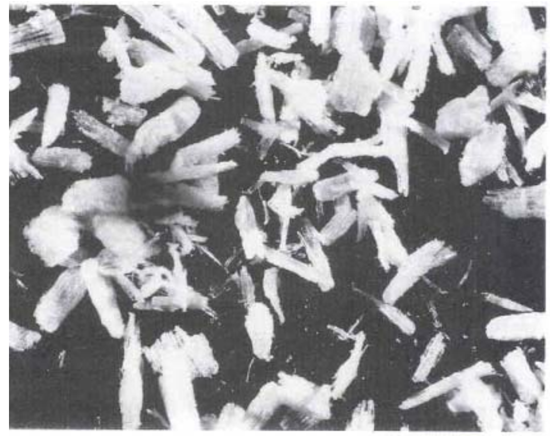

FIG. 3. Commercial 40-mesh wood flour.

COMPARISON OF WOOD FLOUR/PP AND WOOD FIBER/PP COMPOSITES

Figure 3 shows 40-mesh commercial wood flour produced by grinding post-industrial waste wood, primarily ponderosa pine. To produce wood fiber, hardwood chips were refined under pressure and screened through a 40-mesh screen, resulting in the fibrous-looking raw material shown in Fig. 4.

The wood flour studies led us to conclude that slightly higher aspect ratios yield composites with slightly higher strength and stiffness. Therefore, further increasing the aspect ratio should result in further property improvements. Wood fiber lengths were analyzed using the optical Kajaani method. Before processing, average fiber length was 0.66 mm. During processing, the fiber was broken down to an average length of 0.36 mm. Knowing that hardwood diameter ranges from 0.014 to 0.0222 mm, the aspect ratio in the composite after processing could be calculated as 16.26. Compare this to the average aspect ratio of the 40-mesh wood flour of approximately 3.3. The higher aspect ratio of the wood fiber (5 to 8 times higher than that of the wood flour) should lead to increases in strength, as en-

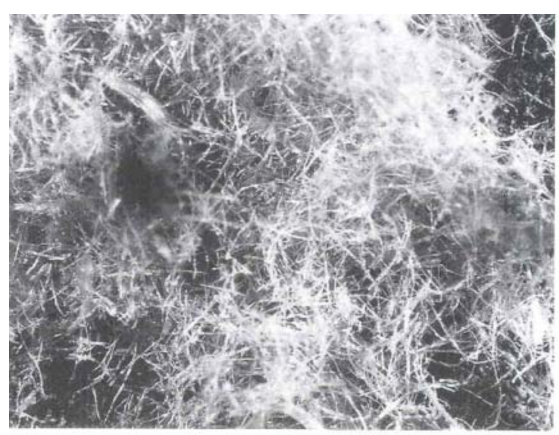

FIG. 4. Refined 40-mesh wood fiber.

TABLE 5. *Summary of mechanical properties of wood flour and 20% wood fiber composites.*

Material	Tensile properties			Flexural properties		Izod impact energy	
	Strength (MPa)	Modulus (GPa)	Elongation (%)	Strength (MPa)	Modulus	Notched (J/m)	Unnotched (J/m)
Wood flour	25.8	2.23	4.4	43.0	1.84	19.5	119
Wood flour and MAPP	29.4	2.35	[4.1]	46.9	[1.90]	[19.1]	[117]
Wood fiber	26.2	[2.27]	[4.2]	[42.9]	[1.88]	[20.8]	141
Wood fiber and MAPP	37.0	[2.31]	4.9	52.2	[1.91]	22.5	197
Polypropylene	28.5	1.53	5.9	38.3	1.19	20.9	656
COV (%)[b]	1	4	4	2	3	6	11

MAPP = 3% maleated polypropylene Brackets indicate values not statistically different from those of uncoupled wood flour composites (α = 0.05)

hanced stress transfer across the matrix to the fiber.

The performance of wood-fiber-reinforced PP composites was compared with that of PP composites filled with commercial wood flour. All filler contents were 20% or 40% by weight. Wood-flour-filled PP was taken as the baseline for our comparisons because it is currently the most common commercial wood/plastic composite. Tables 5 and 6 present the mechanical properties of wood flour and wood fiber composites at 20% and 40% filler levels, respectively, with or without the addition of the coupling agent, MAPP. If the addition of MAPP or the use of wood fiber instead of wood flour resulted in an insignificant (α = 0.05) change in property values, the value appears in brackets (Tables 5 and 6). In general, the addition of wood fiber to PP significantly improved the strength of wood fiber composites compared with that of wood flour composites. At the 40% filler level, wood fiber also improved tensile and flexural modulus. Wood fiber and wood flour composites showed little difference in tensile elongation and notched impact properties. Unnotched impact energy improved with the addition of wood fiber and further improved with the addition of MAPP.

Figures 5 and 6 show the wood fiber/PP interface without and with the addition of MAPP. When no coupling agent was added, the bond between the fiber and PP was poor; the failure at the interface led to fiber pullout (Fig. 5). Addition of MAPP improved the wood fiber/PP interfacial bond dramatically (Fig. 6). Table 7 shows the effect of MAPP on the mechanical properties of wood flour and wood fiber composites. The addition of MAPP did not affect tensile and flexural modulus, but it did improve tensile and flexural strength. We observed greater improvement in tensile and flexural strength of the wood fiber composites. While the addition of MAPP accounts for some improvement in strength, greater increases in strength were obtained for the wood fiber/MAPP composites than the wood flour/MAPP composites. This is direct evidence of

TABLE 6. *Summary of mechanical properties of wood flour and 40% wood fiber composites.*

Material	Tensile properties			Flexural properties		Izod impact energy	
	Strength (MPa)	Modulus (GPa)	Elongation (%)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
Wood flour	25.4	3.87	1.9	44.2	3.03	22.2	73
Wood flour and MAPP	32.3	4.10	[1.9]	53.1	[3.08]	[21.2]	[78]
Wood fiber	28.2	4.20	[2.0]	47.9	3.25	[23.2]	91
Wood fiber and MAPP	52.3	4.23	3.2	12.5	3.22	[21.6]	162
Polypropylene	28.5	1.53	5.9	38.3	1.19	20.9	656
cov (%)[b]	1	4	5	1	3	6	8

MAPP = 3% maleated polypropylene. Brackets indicate values not statistically different from those of uncoupled wood flour composites (α = 0.05).

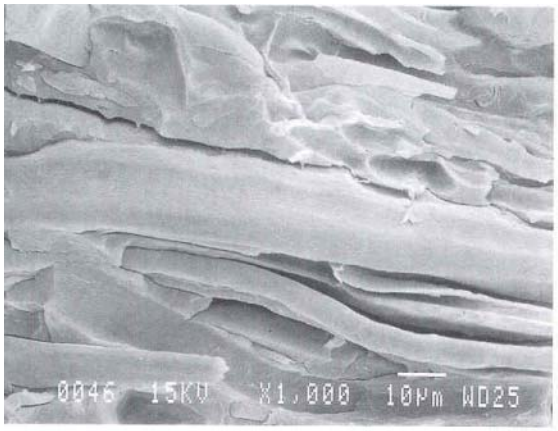

FIG. 5. Hardwood fiber in polypropylene matrix.

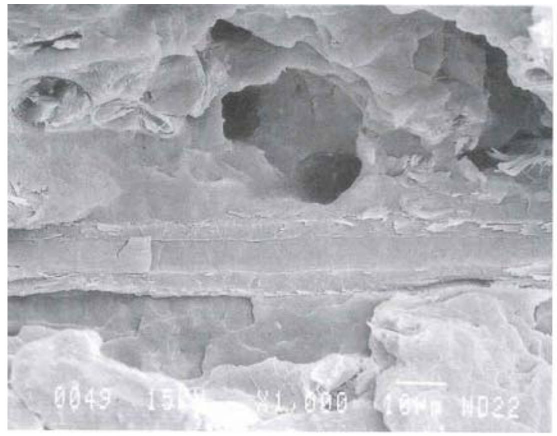

FIG. 6. Hardwood fiber in polypropylene matrix with added maleated polypropylene (MAPP).

more efficient stress transfer from the matrix to the fiber because of the longer fiber length (i.e., higher aspect ratio) of the wood fiber compared with that of the wood flour. The addition of MAPP also improved unnotched impact energy. Unnotched impact energy can be thought of as a measurement of crack initiation. Interfacial bonding lowers the stress concentrations in a composite, thus increasing unnotched impact energy.

CONCLUSIONS

The study of wood flour/polypropylene (PP) composites made with various sizes of wood particles showed that aspect ratio, not particle size, had the greatest effect on strength and stiffness. Wood flour mesh size did affect impact energy. The larger the particle size, the greater the notched impact energy as a result of the increase in fracture surface area. Conversely, the larger the particle size, the lower the unnotched impact energy as a result of higher stress concentrations in the material.

The use of wood fiber instead of wood flour in PP composites resulted in higher strengths at both 20% and 40% filler levels, and higher moduli at the 40% filler level. Pressurized refining increased the aspect ratio of the wood fiber, calculated to be between 16 and 26, relative to 3.35 for 40-mesh wood flour. This higher aspect ratio enhanced stress transfer from the matrix to the fiber. The use of wood fiber had little effect on impact energy.

Addition of the coupling agent MAPP had a greater effect on the properties of wood fiber composites compared to that of wood flour composites. Composites containing 40% wood fiber and 3% MAPP were approximately twice as strong and at least three times as stiff as wood fiber composites without MAPP. The ad-

TABLE 7. *Mechanical properties of wood flour and wood fiber composites with and without MAPP.*

	Tensile properties			Flexural properties		Izod impact energy	
Material	Strength (MPa)	Modulus (GPa)	Elongation (%)	Strength (MPa)	Modulus (GPa)	Notched (J/m)	Unnotched (J/m)
Wood flour–20–MAPP	+14%	+5%	NC	+9%	NC	NC	NC
Wood fiber–20–MAPP	+41%	NC	+17%	+22%	NC	+15%	+40%
Wood flour–40–MAPP	+27%	+6%	NC	+20%	NC	NC	NC
Wood fiber–40–MAPP	+86%	NC	+60%	+51%	NC	NC	+78%

Wood flour is ponderosa pine: wood fiber is hardwood.
NC is no significant change upon the addition of MAPP (α = 0.05).

dition of MAPP did not significantly affect tensile or flexural moduli.

These results clearly support the use of higher aspect ratio wood fibers and coupling agents for increasing the strength of wood/PP composites.

REFERENCES

American Society of Testing and Materials (ASTM). 2000a. Tensile properties of plastics. ASTM D638. Annual Book of ASTM Standards. American Society for Testing and Materials, Philadelphia, PA.

———. 2000b. Flexural properties of unreinforced and reinforced plastics and electrical insulating materials. ASTM D790. Annual Book of ASTM Standards. American Society for Testing and Materials, Philadelphia, PA.

———. 2000c. Impact resistance of plastics and electrical insulating materials. ASTM D256. Annual Book of ASTM Standards. American Society for Testing and Materials. Philadelphia, PA.

Eckert, C. 2000. Opportunities for natural fibers in plastic composites. *In* Proc. Progress in Wood Fiber–Plastic Composites Conference; May 25–25, 2000. Toronto, ON.

Gauthier, R., H. Gauthier, and C. Joy. 1999. Compatibilization between lignocellulosic fibers and a polyolefine matrix. *In* Proc. 5th International Conference on Wood Fiber–Plastic Composites; May 26–27, 1999. Madison, WI.

Osswald, T. A. 1999. Fundamental principles of polymer composites: Processing and design. *In* Proc. 5th International Conference on Wood Fiber–Plastic Composites; May 26–27, 1999. Madison, WI.

———, and G. Menges. 1995. Materials science of polymers for engineers. Hanser/Gardner Publications, Inc., Cincinnati, OH. 475 pp.

Sanadi, A. R., K. Walz, L. Wieloch, R. E. Jacobson, D. F. Caulfield, and R. M. Rowell. 1995. Effect of matrix modification on lignocellulosic composite properties. *In* Proc. 3rd International Conference on Wood Fiber–Plastic Composites; May 1–3, 1995. Madison, WI.

Zaini, M. J., M. Y. A. Faud, Z. Ismail, M. S. Mansor, and J. Mustafah. 1996. The effect of filler content and size on the mechanical properties of polypropylene/oil palm wood flour composites. Polym. Int. 40:51–55.