

## **EFFECTS OF FIBRE CHARACTERISTICS ON THE PHYSICAL AND MECHANICAL PROPERTIES OF WOOD PLASTIC COMPOSITES**

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**SUMMARY:** We investigated the effects of species, wood type and fibre size on the mechanical and physical properties of wood plastic composites (WPC). Wood-based particles from three softwood species (white cedar, jack pine and black spruce) and three wood fibres (sapwood, heartwood and bark) were investigated. Wood particles (24, 42 and 65 mesh) were compounded into pellets at 25%, 35% and 45% by weight with HDPE by twin-screw extrusion. Flexural, tensile and water absorption test specimens were prepared by injection moulding. Toughness and elongation at break of the resultant composite materials decreased with larger particle size. Although strength of the resultant composites improved with larger particle size, toughness decreased. Substantial differences between average mechanical and physical properties of WPC filled with the three different wood species were observed. Black spruce and jack pine produced the best strength and elasticity. Composites made from heartwood and bark showed significantly higher toughness and elongation at break than those made entirely from sapwood. This effect was more pronounced when particle concentration increased. The water uptake of the WPC slightly varied among species and increased with fibre size increasing but to a negligible extent as compared with other wood-based composites and solid wood.

**KEYWORDS:** wood species variability, injection moulding, white cedar, jack pine, black spruce, bark, HDPE, mechanical properties, water uptake.

## INTRODUCTION

Effective use of wood-based particles and fibres as fillers or reinforcements in thermoplastic composites requires a fundamental understanding of the structural and chemical characteristics of wood [1]. English and Falk [2] provide a comprehensive overview of factors that affect properties of wood-plastic composites (WPC). Although several studies have shown that fibre-polymer compatibility can be enhanced by selecting suitable coupling agents [3-4], compatibility between polar wood fibre and non-polar thermoplastics still remains a key to extend the application limits of the resultant composites [5]. Another, frequently cited key factor in natural fibre thermoplastic composites is thermal degradation [6]. Furthermore, the various wood species have different anatomies. These structural differences impact the use of these materials in WPC. For example, fibre dimensions, strength, variability and structure are important considerations. Maldas et al. [7] are among the few researchers who have investigated the effect of wood species on the mechanical properties of wood/thermoplastic composites. They reported that differences in morphology, density and aspect ratios across wood species accounted for varying reinforcement in thermoplastic composites. Recently, Neagu et al. [8] investigated the stiffness contribution of various wood fibres to composite materials. They observed a correlation between lignin content and longitudinal Young's modulus, and an optimal lignin content range for maximum fibre stiffness was recorded for softwood Kraft fibres. Several attempts have been made to correlate wood-based particles and fibre properties to WPC properties [9-11]. A high aspect ratio (length/width) is very important in fibre reinforced composites, as it indicates potential strength properties [12]. Stark and Berger [11] investigated the effects of particle size on the properties of polypropylene filled with wood flour. They concluded that melt flow index, heat deflection temperature, notched impact energy and flexural and tensile modulus and strength increased with increasing particle size. Later, Stark and Rowlands [10] reported that aspect ratio, rather than particle size, had the greatest effect on strength and stiffness. They suggested that particle size does not affect specific gravity. The objective of this study was to determine the effects of wood particle size and species on the physical and mechanical properties of wood particle-reinforced high-density polyethylene (HDPE).

## MATERIALS AND METHODS

Five types of wood sawdust were investigated in this study: Eastern white cedar where; sapwood and heartwood sawdust were treated separately, jack pine wood sawdust and bark shavings and black spruce sawdust. Sawdust and shavings were hammer-milled into particles. Particles were then screened and classified into 3 mesh sizes (Table 1) using an oscillating multideck screen classifier. At this stage, the particles had a moisture content of 10.5 %.

Table 1 Hammer-milled particle classes

Mean class	Class interval	
	<i>mesh</i>	$\mu m$
24 mesh	]20,28]	]850, 600]
42 mesh	]35,48]	]425, 300]
65 mesh	]48,100]	]300, 150]

The HDPE polymer (Goodfellow Corp., USA) used was a semi-crystalline polymer (70–80%) with 0.95 density, 9.0 g/10 min melt index and 135°C melting point. Ethylene-Maleic Anhydride Copolymer (MAPE, A-C® 575A, Honeywell Int., USA) was used as a coupling agent. WPC were produced in a two-stage process. In the first stage, wood particles were compounded into pellets at 25%, 35%, and 45% by weight with the HDPE, using a co-rotating twin-screw extruder. Barrel temperatures of the 4 zones ranged from 180 to 190°C from feeding to die zones. Screw speed was 240 rpm and melt pressure at the die varied between 15 and 25 bars, depending on wood particle content. In the second stage, injection WPC test specimens were made using a reciprocating-screw injection moulding machine in the conditions shown in Table 2. Specimens were made according to ASTM specifications for tensile, impact and bending strength testing. They were stored in controlled conditions (50% relative humidity and 23°C) for 40 hours prior to testing. Tensile and bending tests were performed according to ASTM D638 [13] and ASTM D790 [14], respectively. Toughness is defined as the energy equivalent to the area under the stress-strain curve. Composites were immersed in water at room temperature for two months. Specimens were weighed at regular intervals using an analytical balance and water uptake was calculated.

Table 2 Injection moulding machine settings

Mould temperature – fixed/mobile	38°C/38°C
Injection pressure	900kPa
Injection pressure time	10s
Hold pressure	900kPa
Hold pressure time	4s
Barrel temperature profile: feed, zone1, zone2, nozzle	160, 190, 190, 190°C
Screw speed	135rpm
Cooling time	15s

## RESULTS AND DISCUSSION

Fig.1a shows the tensile modulus of elasticity results and Fig.1b shows the tensile strength of wood particle-reinforced HDPE with particle size. The effect of particle size is unclear with low wood particle content. In contrast, at high wood particle content (45 wt %), tensile modulus of elasticity and strength increase significantly. This is consistent with previous reports on wood-particle thermoplastic composites [10, 11, 15]. As for tensile modulus, flexural modulus of elasticity shows a steady increase with increasing particle size at higher filler content (Fig.1c). It rises from 2.1 GPa at particle sizes ranging from 100–48 mesh (150–300 µm) to 2.7 GPa at average particle size of 24 mesh (≈710 µm) and 45 wt% filler content. These results are in good agreement with previously reported data [11, 15].

Flexural strength development (Fig.1d) also demonstrates that particle size has greater impact at higher fibre load (45 wt %), with approximately 24% higher strength when average particle size increases from 65 to 24 mesh. On the other hand, the incorporation of wood particles in the HDPE matrix steadily increases tensile strength (Fig.1b) independently of filler content. When average particle size increases from 65 mesh (≈230 µm) to 24 mesh (≈710 µm), tensile strength improves by 43%, 10% and 12% at 25 wt%, 35 wt% and 45 wt % filler content, respectively. Zaini et al. [15] reported increasing maximum tensile strength with increasing particle size for

isotactic polypropylene filled with 250 to 63 mesh oil palm wood flour, while Stark and Rowlands [10] recommended the use of higher aspect ratio wood fibres, rather than larger size, to increase WPC strength. However, Stark and Berger [11] found that larger filler particles (greater than 250  $\mu\text{m}$ ) decreased tensile properties.

Figs. 2a and 2b show toughness and elongation at break of the resultant composite materials, respectively. As expected, both properties decrease with larger particle size. The effect is more pronounced as particle concentration increases. This is a common tendency, and has been reported with inorganic filler as well [16]. The strength of composites depends on the debonding process at the fibre end and the fibre's pull-out process during interface failure [17]. The phenomenon is more pronounced as particle size increases. Since cracks travel around the wood particles, the fracture surface area increases with increasing particle size. As a result, less energy is required to fracture a specimen with larger particles [11].

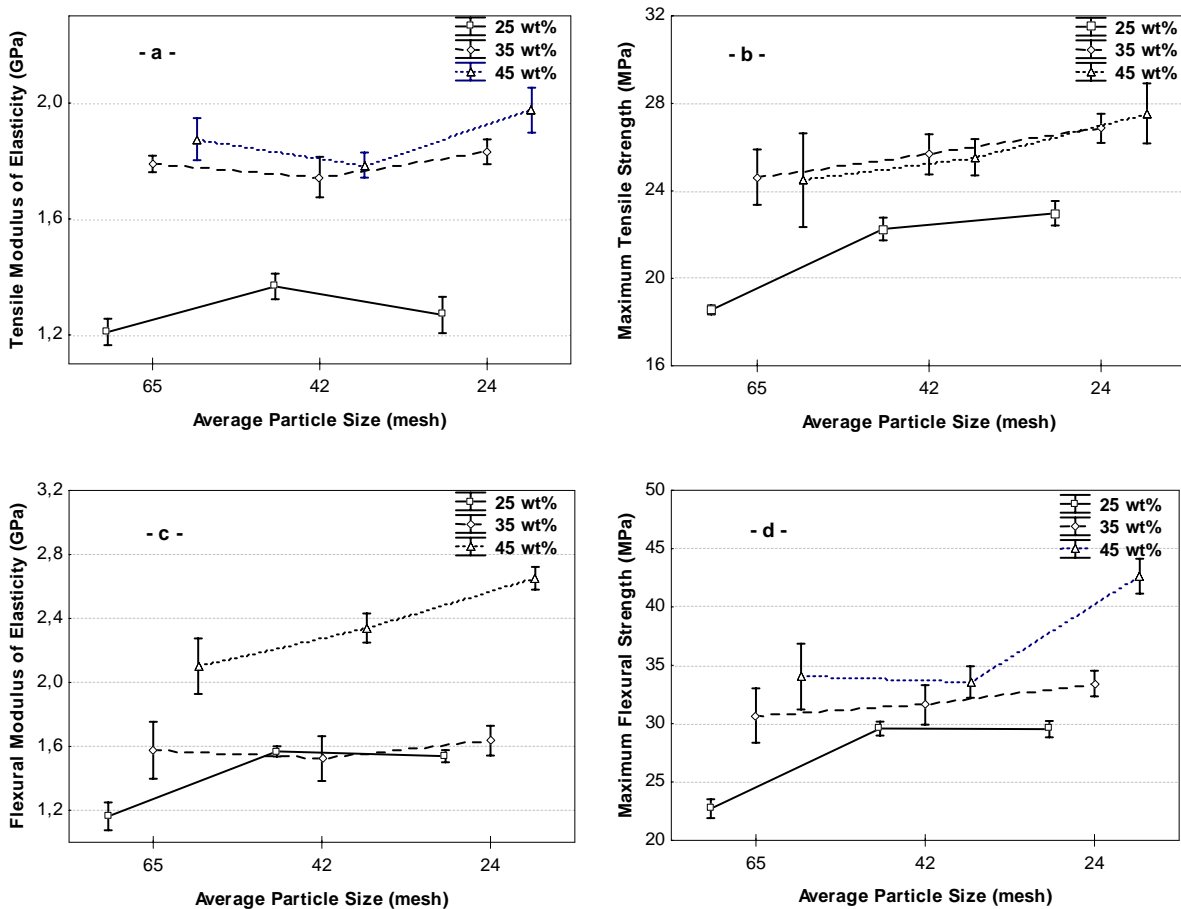


Fig. 1 Effect of wood particle size with various filler contents on: (a) tensile modulus of elasticity; (b) tensile strength; (c) flexural modulus of elasticity; (d) flexural strength of WPC.

In summary, although incorporating particles of larger sizes into the HDPE matrix proved effective in improving strength properties, this turned out to be detrimental to toughness. Thus, depending on end use, the composite should be optimized for either stiffness or toughness by adjusting both filler particle size and concentration. Fig. 3a and 3b illustrate the modulus of

elasticity and maximum strength, respectively, of wood particle-reinforced HDPE composites with various wood types. The composite contained 35 % by weight wood particles at 42 mesh. WPC made with jack pine and black spruce particles exhibits the highest modulus of elasticity and strength, whereas bark and Eastern white cedar WPC show the lowest properties. Average flexural modulus of elasticity ranges between 2.3 and 2.2 GPa for jack pine and black spruce WPC respectively, while maximum flexural strength is approximately equal (40 MPa) for both species. In other words, both the modulus of elasticity and maximum strength of HDPE were enhanced at least 1.5 times when jack pine or black spruce was added. During the hammer-milling process, Eastern white cedar and bark showed a higher vulnerability, producing a large proportion of fines when the aspect ratio was lower than 4 (results not shown). This vulnerability can be explained by the low specific gravity of cedar wood (0.32) and bark (0.40) compared to jack pine (0.43) and black spruce (0.46). The high thermal sensitivity of the bark particles contributes significantly to generate fines. As discussed above, this high fraction of fines reduces WPC stiffness.

Fig. 4a and 4b depict the toughness and elongation at break, respectively, of the obtained composite materials. Composites filled with Eastern white cedar sapwood and bark showed brittle behaviour, with 90% and 60% lower toughness than black spruce. This result is mainly explained by the poor adhesion between the particles of some species and the HDPE matrix. Since white cedar wood and bark contain high amounts of extractives at the surface compared to jack pine and black spruce, a weak surface boundary layer can be formed, making the coupling agent less effective in forming a cross-linking network with the cellulose [18]. This unexpected result was recorded with the bark-particle WPC, which showed the second highest elongation at break after black spruce WPC. This might be explained by the good dispersion of this particle type. Some lipophilic extractives might help to disperse the particles during WPC preparation. These results agree with Maldas et al. [7], who reported that differences in morphology, density and aspect ratios across wood species accounted for varying reinforcement in thermoplastic composites. Thus, it is important to select the appropriate wood species for optimal WPC end use.

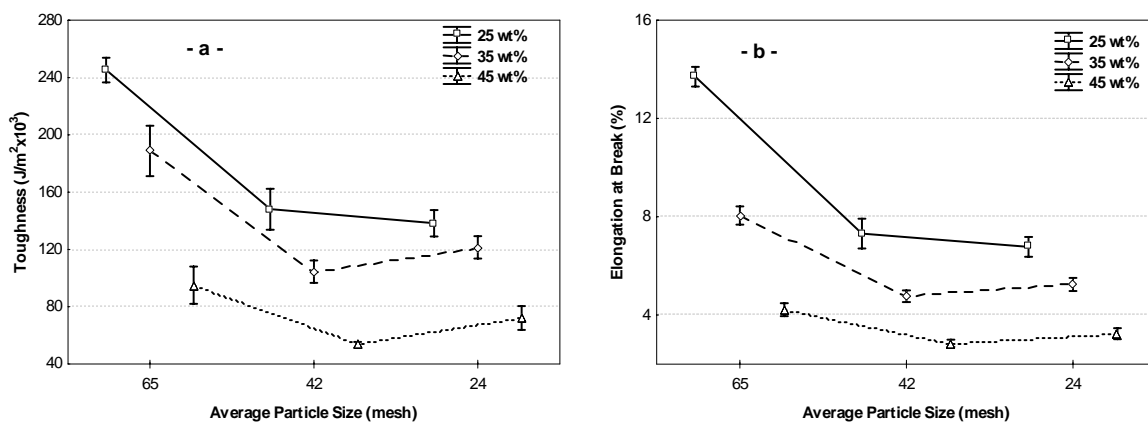


Fig. 2 Effect of wood particle size and filler content on: (a) toughness (tensile energy); (b) tensile elongation at break.

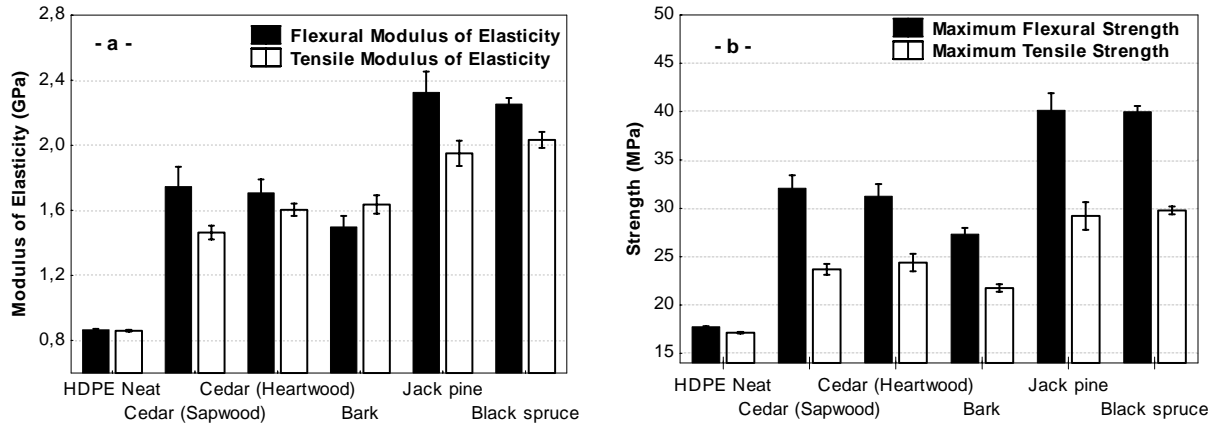


Fig. 3 Wood particle/HDPE injection-moulded composites with different filler species: (a) tensile and flexural modulus of elasticity; (b) maximum flexural and tensile strength.

Fig. 5a depicts the effect of particle size on WPC water uptake. As previously reported [19-20], the higher the particle size, the higher the water absorption. This can be explained in two ways: (i) larger particles lead to greater hydrophilic exposed surfaces; and (ii) poor adhesion between wood particles and the polymer matrix generates void spaces around the wood particles. These voids in the bulk matrix are readily filled with water. As an unexpected result, no difference was observed between composites filled with 24 mesh and 42 mesh (average) particles. This might be explained by the development of particle dimensions during processing. We investigated this development, mainly after compounding (results not shown). It was observed that particles with higher aspect ratios were subject to severe damage due to higher shear stresses that developed in the conical counter-rotating intermeshing twin-screw extruder during mixing and compounding. Apparently, particle length distribution after compounding skewed towards the shortest particle length.

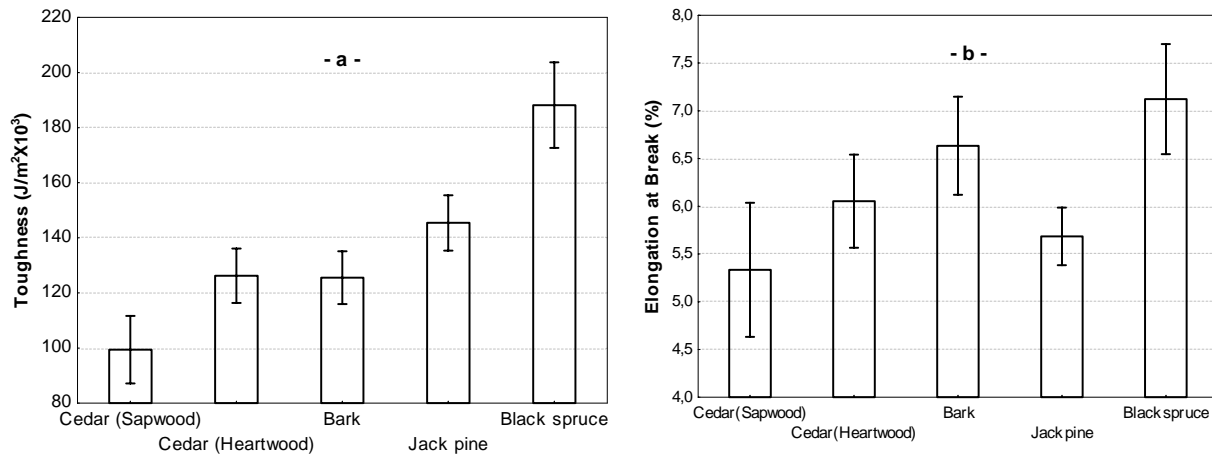


Fig. 4 (a) Variation in toughness; (b) maximum tensile elongation at break for wood particle/HDPE injection-moulded composites with different filler species.

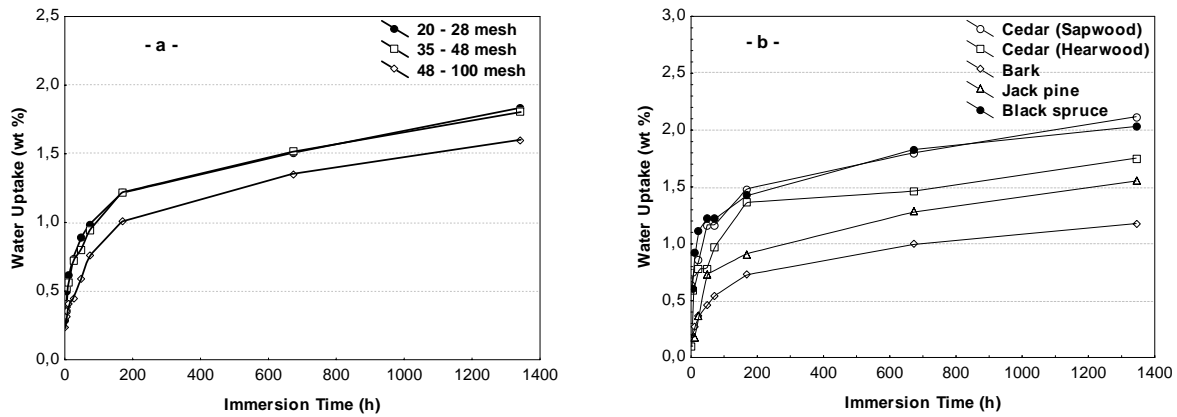


Fig. 5 Average water uptake in composite materials with (a) wood particle size and (b) wood filler species after two months of water immersion.

WPC water absorption tended to increase up to 8 weeks immersion. This negates the argument that WPC reach saturation at short and medium immersion times. For example, assuming that moisture was absorbed by the wood particles alone, then average moisture content of the wood particles was 10.4% in composites filled with 42 mesh particles at 35 wt%. However, when neat wood particles are immersed in water, moisture content readily exceeds 30%, which corresponds to fibre saturation. Therefore, WPC water uptake may actually be slowed down rather than delayed.

The fractional water-uptake curves, as a function of composites filled with different wood particle species, at 35 wt%, and immersion time are shown in Fig. 5b. As expected, since the filler particles are organic, water absorption increases with immersion time. Maximum water uptake is observed at about 2.5% for Eastern white cedar (sapwood) WPC, and about 1.5% for bark WPC. Bark WPC exhibit lower water absorption compared to those made with wood particles, which can be explained by differences in chemical composition between bark and wood. Since bark contains higher amounts of hydrophobic components (lignin and extractives) and lower amounts of hydrophilic components (cellulose and hemicelluloses), it would be expected to show higher water uptake. In addition, WPC water uptake varies slightly across species and increases with particle size, although negligibly compared to other wood-based products such as solid wood and particleboard.

## CONCLUSIONS

The effect of particle size was unclear at lower wood particle content. At higher wood particle content (45 wt %), however, the impact in tensile modulus was substantial. Moreover, flexural modulus of elasticity showed a steady increase with increasing particle size. At 45 wt %, approximately 24% improvement in flexural strength was recorded when average particle size increased from 65 mesh to 24 mesh. On the other hand, toughness and elongation at break for the resultant WPC decreased with larger particle size. The effect was more pronounced as particle content increased.

Composites made from jack pine and black spruce particles exhibited the highest modulus of elasticity and strength. The lower elasticity and strength of Eastern wood cedar and bark based composites is explained by the low specific gravity and thermal sensitivity of the fillers. Composites made from bark particles exhibited lower water absorption compared to those made from wood particles. Differences in chemical composition between bark and wood are among the plausible explanations for the variability of water uptake.

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## REFERENCES

1. D.D. Stokke and D.J. Gardner, "Fundamental Aspects of Wood as a Component of Thermoplastic Composites", *Journal of Vinyl and Additive Technology*, Vol. 9, no. 2, 2003, pp. 96.
2. B.W. English and R.H. Falk, "Factors that Affect the Application of Woodfiber-Plastic Composites", *Proc. of the Woodfiber-Plastic Composites Conference*, 1995, pp. 183.
3. A.K. Bledzki, S. Reihmane, and J. Gassan, "Thermoplastics Reinforced with Wood Fillers: a Literature Review", *Polymer - Plastics Technology and Engineering*, Vol. 37, no. 4, 1998, pp. 451.
4. D. Maldas, B.V. Kokta, R.G. Raj, and C. Daneault, "Improvement of the Mechanical Properties of Sawdust Wood Fibre-Polystyrene Composites by Chemical Treatment", *Polymer*, Vol. 29, no. 7, 1988, pp. 1255.
5. P. Zadorecki and A. J. Michell, "Future Prospects for Wood Cellulose as Reinforcement in Organic Polymer Composites", *Polymer Composites*, Vol. 10, no. 2, 1989, pp. 69.
6. C. Burgstaller, "Processing of Thermal Sensitive Materials - A Case Study for Wood Plastic Composites", *Monatshefte fur Chemie*, Vol. 138, no. 4, 2007, pp. 343.
7. D. Maldas, B.V. Kokta, and C. Daneault, "Thermoplastic Composites of Polystyrene: Effect of Different Wood Species on Mechanical Properties", *Journal of Applied Polymer Science*, Vol. 38, no. 3, 1989, pp. 413.
8. R.C. Neagu, E. K. Gamstedt, and F. Berthold, "Stiffness Contribution of Various Wood Fibers to Composite Materials", *Journal of Composite Materials*, Vol. 40, no. 8, 2006, pp. 663.



9. A.K. Bledzki and O. Faruk, "Microcellular Injection Molded Wood Fiber-PP Composites: Part II- Effect of Wood Fiber Length and Content on Cell Morphology and Physico-Mechanical Properties", *Journal of Cellular Plastics*, Vol. 42, no. 1, 2006, pp. 77.
10. N. M. Stark and R. E. Rowlands, "Effects of Wood Fiber Characteristics on Mechanical Properties of Wood/Polypropylene Composites", *Wood and Fiber Science*, Vol. 35, no. 2, 2003, pp. 167.
11. N.M. Stark and M.J. Berger, "Effect of Particle Size on Properties of Wood-Flour Reinforced Polypropylene Composites", *Fourth International Conference on Woodfiber-Plastic Composites*, Madison, Wisconsin, 1997.
12. R.M. Rowell, J.S. Han, and J.S. Rowell, "Characterization and Factors Effecting Fiber Properties", *Natural Polymers and Agrofibers Composites*, Vol. 2000, pp. 292.
13. ASTM D638-03, "Standard Test Method for Tensile Properties of Plastics", *ASTM International*, West Conshohocken, 2003.
14. ASTM D790-03, "Standard Test Method for Flexural Properties of Unreinforced and Reinforced Plastics and Electrical Insulating Materials Plastics", *ASTM International*, West Conshohocken, 2003.
15. M.J. Zaini, M.Y.A. Fuad, Z. Ismail, M.S. Mansor, and J. Mustafah, "The Effect of Filler Content and Size on the Mechanical Properties of Polypropylene/Oil Palm Wood Flour Composites", *Polymer International*, Vol. 40, no. 1, 1996, pp. 51.
16. I. L. Dubnikova, S. M. Berezina, and A. V. Antonov, "Effect of Rigid Particle Size on the Toughness of Filled Polypropylene", *Journal of Applied Polymer Science*, Vol. 94, no. 5, 2004, pp. 1917.
17. Y.H. Liu, J. Q. Xu, and H.J. Ding, "Mechanical Behavior of a Fiber End in Short Fiber Reinforced Composites", *International Journal of Engineering Science*, Vol. 37, no. 6, 1999, pp. 753.
18. H. Saputra, J. Simonsen, and K. Li, "Effect of Extractives on the Flexural Properties of Wood/Plastic Composites", *Composite Interfaces*, Vol. 11, no. 7, 2004, pp. 515.
19. M. Tajvidi, S.K. Najafi, and N. Moteei, "Long-Term Water Uptake Behavior of Natural Fiber/Polypropylene Composites", *Journal of Applied Polymer Science*, Vol. 99, no. 5, 2006, pp. 2199.
20. V. Steckel, C.M. Clemons, and H. Thoemen, "Effects of Material Parameters on the Diffusion and Sorption Properties of Wood-Flour/Polypropylene Composites", *Journal of Applied Polymer Science*, Vol. 103, no. 2, 2007, pp. 752.