WOOD POLYMER COMPOSITES

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ABSTRACT

This paper is an update of several earlier review articles on wood polymer composites (WPC). Notable polymer types that have been used to make WPC are phenol-formaldehyde (PF), various vinyls, and furfuryl alcohol. Phenol-formaldehyde and similar chemicals typically enter wood cell walls, whereas the vinyls usually fill cell lumens. Combination treatments that modify both lumens and walls are possible. Diffusion and mass flow are two possible methods of introducing the chemical into the wood. Curing can be accomplished using a catalyst and heat or penetrating radiation. Type of treating chemical and final density are major determinants of WPC properties. Physical and mechanical properties of WPC have been extensively studied. Major WPC improvements over untreated wood are in hardness, finishability, and dimensional stability. There is much yet to be learned about chemical formulations, treating and curing, and their relationships to property enhancements, WPC recyclability, and chemicals from renewable sources for making WPC.

Keywords: Wood polymer composites, WPC, wood polymer combinations, acrylic wood, impregnated wood, stabilized wood, modified wood.

INTRODUCTION

Wood polymer composites (WPC) are made by impregnating wood with a polymerizable monomer or prepolymer and then curing the monomer or prepolymer to a solid. Solid wood (lumber) or any wood composite (such as waferboard and medium density fiberboard) can be used to make WPC, although solid wood is usually used. WPC has polymer in cell lumens or in cell lumens and walls. WPCs have many changed and improved physical properties compared with the parent wood. Notable are an increase in surface hardness and dimensional stability, and the possibility of fine finishing without surface coating.

Over the years, researchers have impregnated wood with a variety of chemicals to produce WPC. A few of these have found commercial applications, some for a limited time. Currently, there are several companies producing WPC products, mainly flooring. There are many potential applications for the material.

There are several earlier review articles about wood polymer composite materials processes, properties, and products. A book chapter by A. J. Stamm, one of the pioneers in many types of modified wood, covers processes and the products formed (Stamm 1977a). Bryant (1966) reviews modified woods including WPC. A. E. Witt (1981) wrote a review of radiation-polymerized vinyl WPC. He directed a pioneer company producing gamma-radiation-cured, vinyl WPC, mainly hardwood flooring. Two pioneers and prolific workers in modified woods, I. S. Goldstein and W. E. Loos (1973), published an informative book chapter. Autio and Miettinen (1970) summarized early work

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in Finland, and Schaudy and Proksch (1982) reported on work in central Europe. J. A. Meyer, who is both a polymer and a radiation chemist, pioneered the heat-catalyst method of producing vinyl WPCs. He has written several reviews (e.g., Meyer 1982, 1984, 1987). Rowell and Konkol (1987) of the U.S. Forest Products Laboratory have written a review of treatments that change wood physical properties including WPC.

This article is intended as an update and extension of these earlier reviews.

WOOD TREATABILITY

It is hardwoods (deciduous species) that have mainly been used in making WPC. One reason is that WPC is used for finished products, which are traditionally made from hardwoods. Another is that heartwood of many softwoods (conifers) does not impregnate well. Also, softwoods usually have lower density than hardwoods, requiring more treating chemical for comparable properties. For these reasons, this paper will discuss mainly hardwoods.

Nicholas and Siau (1973) review wood factors that influence its treatability. In hardwoods, pores (vessels) provide major longitudinal flow paths for fluids. Pits between pores and fibers, fiber lumens, and rays also play roles in fluid flow, but less is known about their contributions to the distribution of fluid in the wood.

Sapwood of most species (hardwoods and softwoods) treats well, but heartwood treatability is species-dependent (MacLean 1960). Treatability also varies within species and within trees. Often there are areas of single boards that treat differently than adjacent areas (Schneider 1993).

A great many woods, including tropical species, have been used to make WPC. Some are listed in Table 1.

TYPES OF POLYMERS IN WPC

Representative types of monomers and prepolymers that have been used to make WPC and some of their characteristics are given in Table 2.
 TABLE 1. Some wood species that have shown useful impregnability when producing WPC.

Alder (Alnus spp.) Yellow poplar (Liriodendron tulipifera) Ash (Fraxinus spp.) Maple (Acer spp.) Walnut (Juglans spp.) Birch (Betula spp.) Basswood (Tilia americana) Poplar (Populus spp.) Cherry (Prunus spp.) Red gum (Liquidambar styraciflua) Beech (Fagus spp.) Red and white pine (Pinus resinosa and strobus)

(from Sordo 1975; Bryant 1966; Witt et al. 1981; Young and Meyer 1968; Schneider et al. 1985, 1990; Schneider 1993)

TREATING CHEMICAL COSTS

Basic monomers and prepolymers that can be used to produce WPCs have a wide cost range. Widely used industrial monomers, such as styrene and methyl methacrylate, are the cheapest WPC base chemicals. Unit cost of such chemicals generally decreases with quantity purchased. Conversion efficiency, evaporation during cure, pot life, additives, ease of use, and fire and environmental safety precautions have an effect on final cost of chemicals used to make WPC.

WPC PRODUCTION METHODS

There are two basic processes for moving WPC-producing fluids into wood. Low- and medium-viscosity treating fluids can be forced into wood using vacuum and pressure. Vinyl monomers are examples of low-viscosity fluids used in such WPC preparation. Some epoxies with medium viscosities have been used to treat wood (Langwig et al. 1969; Moore et al. 1983). Higher-viscosity treating fluids, such as phenol-formaldehyde prepolymers, typically move into wood by a combination of flow and diffusion-displacement.

Diffusion method of producing WPC

The Impreg and Compreg processes (Stamm 1964) rely largely upon diffusion to move the

Starting mix	Characteristics Usually clear materials with added crosslinker (Meyer 1968) that can have dye added for color. Usually do not swell cell wall, thus pro- ducing cell-lumen-filled WPC. Styrene has a particularly difficult odor to eliminate from wood products.				
Vinyl and similar monomers [e.g., methyl methacrylate, styrene (Meyer 1984), diallyl phthalate (Raech 1965), polyester-styrene (Autio and Miettinen 1970)]					
Water- and alcohol-soluble prepolymers [e.g., phenol-, urea- and melamine-formaldehyde (Stamm 1964)]	Used with veneers to produce Compreg and Impreg. Phenol resins are light to dark brown. Melamine and urea resins are light-col- ored and can be dyed.				
Low-viscosity epoxy resins (Moore et al. 1983; Langwig et al. 1968)	Formulated for polarity so some wood swelling and wood substance dimensional stabilization occurs.				
Polar monomers [e.g., furfuryl alcohol (Stamm 1977a, b)]	Swell cell wall to give high chemical resistance and dimensional sta- bility in water. Dark-brown material.				
Modified vinyl monomers (e.g., Mathias and Wright 1989; Rowell 1983; Schaudy and Proksch 1982)	Modified with polar groups for cell wall swelling, or fungicide or fire retardant groups.				
Isocyanates (polyurethanes) [e.g., Hartman 1969]	Reactive with wood. Can be modified to include preservatives.				

 TABLE 2. Some types of monomers and prepolymers that have been used for making WPC and their characteristics.

solids of a phenol-formaldehyde resin solution into wood veneer. Veneer is used because of the short distance the fluid must move. After the chemical has diffused into the veneer, it is cured using heat. Impregnated veneers can be cured singly and then glued together to form a board, or they may be stacked together in a press and heated to cure and bond simultaneously. The Compreg process uses pressure beyond that needed for bonding to increase the density of the material. The laminated WPC produced by either process is typically machined into products.

Vacuum-pressure method of producing WPC

Preparing WPCs from solid wood or wood composites is accomplished in two stages. The first stage is identical to full-cell pressure treating with liquid preservatives, except that there is no final vacuum. The second stage is similar to bulk polymerization of liquid monomers or prepolymers.

Mass fluid flow into wood under pressure gradients occurs several orders of magnitude more quickly along the grain than across (Siau 1984); therefore, except for very long samples, most of the treating solution enters through end grain. Low viscosity fluids flow more readily into wood, which is a reason monomers are often used to make WPC. Useful treating fluid viscosities at room temperature using vacuum are in the few cP range (for example, styrene at 0.73 cP and water at 1.8 cP). With overpressure, viscosities from about 30 cP (Autio and Miettinen 1970) to 130 cP (Hartman 1969) are usable.

Before the impregnation stage, water and air must be removed from the wood to provide space for the treating fluid. Water is removed by a drying process. Kiln-dried lumber (approximately 6% MC) or dried particle- or waferboard treats well.

After drying, wood is placed in a pressure vessel and a vacuum drawn to remove air. The evacuated wood has empty cells ready to receive fluid. The wood is left in the pressure vessel under vacuum while the treating fluid flows into the chamber, covering the wood. Vacuum is then released. Sufficient retention may be possible in reasonable time using vacuum followed by atmospheric pressure. For large samples or wood of low permeability, an overpressure may be used. The maximum allowable pressure is the maximum crushing strength of the wood. Practical maximum is normally about 10 atmospheres (1,000 kPa or 150 psig). At the end of the pressure treating stage, the wood contains liquid. The liquid must

be changed into a solid polymer before the material is a WPC.

Polymerization of polymerizable monomers or prepolymers can be initiated using penetrating ionizing radiation (electrons, gamma rays, or X-rays) or a chemical catalyst and heat (Meyer 1965; Siau and Meyer 1966), including radio frequency heating (Beall et al. 1966). For vinyls, the heated catalyst breaks down into free radicals that can initiate polymerization. The heat polymerization process normally occurs in ovens separate from the pressure treating apparatus. Since flammable vapors can be generated in this step, apparatus that maintains a low vapor concentration and/or does not have a vapor ignition source should be used.

Rosen (1974) gives details of how to produce heat-catalyst, vinyl WPC on a small scale.

TYPES OF WPC

There are two orders of porosity in woodcell cavities and micropores in the cell walls. Cell-wall micropores are transient. They are at maximum volume in fully water-swollen wood and disappear nearly linearly with moisture content as wood dries below the fiber saturation point. They can be largely (there is some hysteresis) restored by re-exposing to moisture or another polar fluid. The cyclic moistening and drying wood undergoes in use, increasing and decreasing micropore volume, are the reason for its dimensional instability. Polymers in WPC can largely fill cell lumens or they can enter the micropore volume.

Cell-lumen WPC

If a chemical introduced into dry wood does not cause swelling, the chemical remains in cell lumens. Most of the common vinyl monomers (such as styrene and methyl methacrylate) are in the nonswelling or little-swelling category over normal treating times (Siau 1969) and so produce essentially cell lumen WPC. When a nonswelling chemical is changed into a polymer, the polymer will occupy the cell cavities but not the cell walls. Since the cell cavities are a major path for moisture movement in wood (Siau 1984), plugging them with a polymer makes the wood more resistant to rapid changes in moisture content, especially along the grain. The effect is greater dimensional stability over the short term. There is speculation that physical restraint by polymers in cell lumens reduces swelling at a given moisture condition (Schneider et al. 1991). This would contribute to dimensional stability.

Cell lumen wood polymer composites are internally mechanically reinforced wood with improved finishing characteristics. The reinforcement afforded by cell lumens filled with polymer increases such properties as elastic moduli, rupture moduli, surface hardness (Schneider et al. 1990) and toughness (Schneider et al. 1989). Fine finishes can be obtained by sanding and polishing. Adding dye to the monomer colors the material. Since the dye remains largely in the polymer (nonswelling monomers do not take it into the wood substance), low density woods with high polymer content give most brilliant colors. Denser woods have more unstained wood substance per unit volume, diluting the color.

Cell-wall WPC

Dramatic increases in dimensional stability result from stabilizing the transient cell-wall micropores. This has been accomplished by using low molecular weight chemicals, which swell the cell wall as they enter it and subsequently are cured into a solid, nondissolving polymer. This holds the wall in a swollen state permanently. Solvent exchange techniques have also been used to stabilize cell-wall micropores. Impreg and Compreg processes, for example, use phenol-formaldehyde prepolymer dissolved in water or alcohol (Stamm 1977a). The solvent produces the micropores, and the prepolymer exchanges with the solvent in the cell wall. Curing the prepolymer holds the wood swollen. Polyethylene glycol (PEG) is a polymer that is used to stabilize wood in an exchange method (Stamm 1977a). The PEG is a waxy solid that retains wood mechanical properties near those of green wood, so does not really develop a wood polymer composite.

The PEG remains water-soluble and hygroscopic, limiting its usefulness in wood.

Cell-wall WPCs are particularly useful when extremes of pH, temperature, moisture, or exposure to decay organisms will be encountered in the material's use. Phenol-formaldehyde cell-wall WPC is high temperature, organic solvent, and acid resistant; furfuryl alcohol WPC is highly resistant to alkali as well (Goldstein and Loos 1973). High polish can be obtained with sanding and buffing. Since furfuryl alcohol and some phenol-formaldehyde polymers are dark and penetrate cell walls, dark material is formed regardless of the original wood density. This can be valuable when dark, tropical woods are being simulated using lighter, temperate zone woods. Light-colored cell-wall WPC is made using light-colored, phenol-formaldehyde resins and can also be made using urea- and melamine-formaldehyde formulations (Stamm 1964). The melamine resins have not found industrial acceptance, possibly because of cost. With urea formulations, contact with wood causes pH changes in treating solutions, which can lead to premature resinification. Without very careful control, the resin precipitates, making the solution useless for further treating.

Combination treatments

It is possible to combine cell-lumen and cellwall treatments. Monomers that are essentially nonswelling have been diluted with swelling solvents to obtain the combination effects (Furuno and Goto 1973). The solvents evaporate during cure, which limits solids retention. Cellwall followed by cell-lumen treatments (Rowell et al. 1982), neat (100% active ingredient), swelling monomers or mixtures of swelling and nonswelling monomers (Loos and Robinson 1968; Schaudy and Proksch 1982; Rowell et al. 1982) and silane coupling agents combined with nonswelling monomers (Brebner and Schneider 1985; Schneider and Brebner 1985) have shown improved dimensional stability over cell-lumen treatments. Considering the large number of chemicals available, combination formulations that develop desirable properties of both cell-wall and cell-lumen treatments have promise.

TREATING AND CURING EFFECTIVENESS

Monomer retention

In a wood of a particular relative density, there is a maximum space available (void volume fraction or porosity, Siau 1984) for monomer to fill. In the case of nonswelling chemicals, maximum retention occurs when the total lumen volume is filled. With swelling chemicals, transient volume in the cell wall is also available, but the volume of the sample is increased by the swelling. Maximum uptake (based upon wood weight at 6% MC) expected by fully filling wood voids with nonswelling fluids of 0.9 g/cc and 1.1 g/cc is shown in Fig. 1. The 6% MC weight is used because kilndried lumber is normally used for WPC production.

Polymer retention

Curing processes may evaporate some monomer and may force partially cured chemical from wood. Most monomers shrink upon polymerization. Polymer shrinkage can develop air space in cell lumens and can also cause the wood to shrink. Shrinkage, evaporation, and exudation lower polymer retention.

Polymerization reactions typically do not change all of the starting chemical to polymer. In vinyl chain growth (addition) polymerizations, for example, some monomer remains unreacted. With some step growth (condensation) polymers, volatiles are lost during the reaction. It may be desirable to leave some monomer unreacted in the polymer to act as plasticizer. Monomers may cause objectionable odor, however. Condensation reaction byproducts may be water, carbon dioxide, or formaldehyde. These are typically volatilized and lost from the product. Knowing how much of these are expected, based on chemical stoichiometry, allows calculation of polymerization efficiency. Stamm (1977b) did this for furfuryl alcohol resinification.

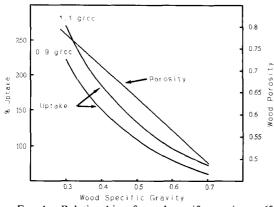


FIG. 1. Relationship of wood specific gravity at 6% MC to its porosity (void volume fraction) and the uptake of two monomer densities (0.9 g/cc and 1.0 g/cc) based upon wood weight at 6% MC.

Final WPC density determines many properties of the material. WPC density, calculated from wood porosity assuming perfect filling and using 3 polymer densities, is shown graphically in Fig. 2. Wood porosity was calculated from: Va = 1 - G(0.667 + (MC/100)) (Siau 1984, Eq. 1.19). Percent uptake is: (Va \times fluid density)/G, where Va is porosity and G is moist wood specific gravity. Oven-dry wood is used in the calculation because heat curing dries the wood. The figure shows retentions of lowerdensity (1.0 g/cc and 1.2 g/cc) chain-growth vinyl polymers with assumed shrinkage of 15% during polymerization and additional depletion of 5% from evaporation during cure. The figure also predicts retention of higher-density (1.5 g/cc), step-growth polymers, many of which lose small molecules during polymerization, causing high shrinkage. For the figure, a shrinkage of 30% and an evaporation loss of 5% was assumed. Step-growth polymers used for WPC (such as phenol-formaldehyde and furfuryl alcohol) swell the wood, which was not accounted for in the calculation. Figure 2 is in reasonable agreement with treating results of both vinyl and step-growth monomers in readily treated wood.

Depth of polymer penetration

Most WPC is produced from wood that is quite uniformly penetrated with monomer, and

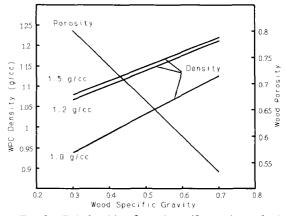


FIG. 2. Relationship of wood specific gravity to final WPC density for polymers of several densities. The wood specific gravity versus porosity line is included.

thus the cured product has a fairly uniform density throughout. With some large cross-section products such as gunstocks, furniture parts, and thick cutting boards, such complete loading produces a very heavy product. In such products, only an outer layer requires enhanced properties offered by WPC. Fully loading the surface volume of such products would give desired property enhancements without undue weight. One method of achieving surface volume loading is to control chemical viscosity using a solution of prepolymer in monomer (Brebner 1989).

MATERIAL PROPERTIES

Wood polymer composites have changed properties compared to the parent wood. Notable properties that are changed upon WPC formation, and methods of measuring them, are shown in Table 3. Table 4 lists some specific density and mechanical property changes when producing vinyl WPC. In another example, the average of 15 Shore Durometer D-scale values for untreated sugar maple was 71 and when treated with furfuryl alcohol or MMA increased to 81 (Schneider 1993). WPC, when polymer filling is good, has a smaller density range than various wood species from which it was made (Table 4). Figure 2 shows expected final densities for a range of wood

Property	Measure				
Hardness	Ball indentation (Beall et al. 1973), Shore Durometer value (Schnei- der 1993)				
Dimensional stability, hygroscopicity	Antishrink efficiency, relative swell- ing, sorption isotherms (Rosen 1976; Stamm 1977a; Rowell and Youngs 1981; Schneider et al. 1991)				
Moisture diffusion	Vapometer tests, sorption iso- therms (Hartley and Schneider 1992)				
Toughness	U.S. Forest Products Laboratory toughness tester value (Schneider et al. 1989), Charpy toughness value (Stamm 1977b)				
Moduli of elasticity and rupture, creep	Bending, tensile, compressive (Langwig et al. 1968; Autio and Miettinen 1970; Brebner et al. 1985; Brebner et al. 1988; Schneider et al. 1990; Schneider and Phillips 1991), long-term tests (Boey 1989)				
Density	Weight and volume or Archimedes principle				
Machinability and finishability	Machining and finishing trials (Rowell and Konkol 1987; Meye 1982)				
Weatherability	Outdoor exposure, artificial weath- ering trials (Autio and Miettinen 1970)				
Resistance to biodeterioration	Fungal resistance trials, insect resis- tance trials, marine borer resis- tance trials (Rowell 1983)				
Chemical resistance	Trials using organic solvents, acids and bases (Hills 1972; Stamm 1964)				
Abrasion resistance	Trials using standard abraders such as Taber (lannazzi et al. 1964; Stamm 1977a)				
Fire resistance	Flame spread and fire tube test val ues (Siau et al. 1972)				

TABLE 3. Some properties of WPCs and techniques by

specific gravities for polystyrene WPC (polymer density 1.0 g/cc, 20% shrinkage and monomer loss), methyl methacrylate, and other acrylic WPC (polymer density 1.2 g/cc, 20%) shrinkage and monomer loss) and furfurl al-

Adhesive trials (Rowell 1987)

Gluability

cohol and phenol-formaldehyde WPC (1.5 g/cc polymer, 35% polymerization loss). Lower specific gravity woods have greater mechanical property enhancements than woods with higher starting specific gravities. This results in a smaller range of mechanical properties values for WPC than for the parent woods.

Vinyl WPC has greatly reduced (60% to 90%) less, Rosen 1976) rate of swelling in moist (97% RH) air compared to untreated wood. Antishrink efficiencies (ASE) are about 70% for phenol-formaldehyde (Stamm 1977a), furfuryl alcohol (Stamm 1977b; Schneider 1993) and acrylonitrile (Stamm 1977a) cell-wall treatments. Cell-lumen treatments give very small (0 to 20%, Rosen 1976; Rowell et al. 1982; Schneider 1993) ASE. Moisture diffusion coefficients are typically higher for cell-wall treatments than for cell-lumen treatments (Rowell et al. 1982; Schneider et al. 1991).

WPCs can be glued before or after treatment, with specific techniques dependent upon the adhesive, treating formulation, and treating process. Finishing can be accomplished by sanding or sanding and buffing (Stamm 1977a; Schneider 1993). Machining properties are similar to those of hard plastics. Woodworking machines can be used, but metalworking machines work better for some operations and the harder WPCs. Mechanical fastening, because of hardness, generally requires predrilling.

QUALITY CONTROL IN PRODUCTION

For a particular wood polymer composite, physical and mechanical properties are usually closely related to density. Therefore, once testing has established property values at different loadings with a particular combination of wood and polymer, measurement of density alone should be a good indicator. Density is relatively easy to measure and may thus be one of the best routine quality control tools for producers and buyers. Experience can indicate actual loadings and densities to be expected with particular woods, chemical formulations, and curing schedules.

Surface hardness of WPC depends upon

			MOE						
	Specific gravity		% increase at OD			% increase at 12%			
Species	OD	Treated	Ten.	Comp.	Bend.	Ten.	Comp.	Bend	
Schneider and Phi	llips (1991):								
Basswood	0.35	1.00	-1	48	18				
Sugar maple	0.73	1.20	9	19	16	13	32	20	
Beall et al. (1973):		% hardness modulus increase							
Aspen	0.45	0.98		800					
Sugar maple	0.61	1.06		420					
Oak	0.64	0.94		270					
Young and Meyer	(1968):			% tangent	ial compress	ion strengtl	h increase		
Basswood	0.34	0.90		435					
Red pine	0.51	1.02		636					
Red gum	0.52	0.97		175					
Red maple	0.58	1.06		171					
Black cherry	0.58	1.04		202					
Sugar maple	0.64	1.06		160					
Beech	0.66	1.01		201					
Yellow birch	0.72	1.14		146					

TABLE 4. Some property changes as a result of treating.

polymer loading at the surface and polymer hardness. Polymer hardness, for a given polymer, can be related to degree of cure and amount of monomer remaining. Hardness, like density, can be related to WPC performance and can be a useful quality control measurement. Using a hand-held device such as a Shore Durometer, measurement can be simple.

RECOMMENDATIONS

In processes requiring flow of heterogeneous fluids into wood, the wood may act as a chromatographic column, separating the components of the fluid. Better understanding of such chemical separations during WPC production could be helpful in designing treating fluids.

Even though species of wood have been categorized into treatability classes, experience (Schneider 1993) has shown that there can be wide variability in the same species and even in different parts of the same tree. Heartwood and sapwood of easily treatable species may treat similarly or differently (Young and Meyer 1968). Understanding the wood anatomical and ultrastructural reasons for such variability and being able to predetermine the treatability of particular wood pieces or batches would be useful.

Once a reactable treating solution is in wood, the wood becomes a reaction vessel. The size of the individual cells containing reacting fluid, the restrictions to convective fluid flow as heat is absorbed or generated, the insulating properties of the wood, wood extractives, and perhaps the surface chemical activity of the wood substance influence reactions taking place in the wood. Some evidence for this is the different reaction isotherms observed from the same chemical polymerized in wood and in bulk (Schneider 1993). Better understanding of polymerization reactions in wood could help in chemical formulation and curing process design.

Recyclability may be important in future WPCs. Comminuted WPCs, for example, may be effective furnish for molded or extruded products.

Materials have environmental advantages when produced from renewable resources. Wood is one such material, but most of the monomers used in WPCs are produced from petroleum. It would be to WPCs advantage to use polymers derivable from renewable resources. Such polymers, such as those derived from furfuryl alcohol, have been developed and used to treat wood in the past, but apparently none are currently used commercially.

Dense, high-extractive-content tropical woods have found use over the years where hard, resistant, and often beautiful woods are desired. Such woods are becoming increasingly scarce and expensive. Being able to simulate properties and appearance of such woods using more plentiful woods should help the market for WPCs.

Properties of wood polymer composites can be varied using properties, interactions, and relative quantities of the wood and polymer from which they are made. WPCs thus have potential for being customized to match specific end uses.

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