MEETING SOCIETY'S CHALLENGE: VALUE-ADDED PRODUCTS FROM RECYCLED MATERIALS

BRENT ENGLISH

US Forest Products Laboratory,
Madison, Wisconsin, U.S.A.

ABSTRACT

Technology has brought many benefits to the world. Along with these benefits come problems, one of which is the disposal of solid waste materials. For example, in the United States in 1988, paper and paperboard and wood and plastics accounted for nearly 60% of the municipal solid waste stream. Developing alternative uses for recovered materials helps solve landfill problems and creates new materials that balance performance properties and cost. Composites can be made from combinations of waste wood and plastic materials. Wood-plastic composites research is being conducted using either melt-blend or fiber mat technology. With melt-blend processing, where the ratio of waste wood to thermoplastic is approximately 50:50, the materials are intimately blended in either a high-intensity mixer or a twin-screw extruder. The resulting blend can be processed by conventional plastic processing equipment into sheets, films, or molded parts. The blend can be augmented by a variety of compatablizers or coupling agents. Fiber mat processing involves the mixing of a variety of materials and air laying the materials into a m at. This m at can then be pressed into a flat panel or molded into shapes. This paper provides a brief overview of the research in progress at the USDA Forest Service, Forest Products Laboratory, to make value-added composites. In addition, research and development needs are outlined that will maximize the benefits of using recovered waste materials for composite products.

INTRODUCTION

The word waste projects a vision of a material with no value or useful purpose. However, technology is evolving that holds promise for using waste or recycled wood and, in some cases, even plastics to make an array of high-performance composite products that are in themselves potentially recyclable.

A comprehensive waste management program must rely on the aggregate impact of several courses of action: waste reduction, recycling, waste-to-energy schemes, and landfill (Kovacs 1988). The greatest impact that is likely to result from further research is in the area of recycling. Increased use of recycled biobased resources will allow the markets for fiber composites to grow without increasing the use of virgin timber. Therefore, forest products industries will benefit from such research, because inexpensive raw materials will be available for producing high-quality composites.

When fibers, resins, and other materials are used as raw materials for products, such as paper, extensive cleaning and refinement are required. When recovered fibers, resins, and other materials are used for the manufacture of composites, extensive preparation is not required and thus greatly reduces the potential cost of manufacturing.

This paper focuses on wood-based resources that either presently enter or could enter the recycling stream, both commercially and residentially. This includes newspaper, packaging, wood pallets, other forms of wood-based fiber products, and all forms of industrial and residential solid-wood and wood-based composite waste. The use of resources that coexist with wood-based resources is also considered, including a variety of plastics and other resources that "contaminate" the wood-based resource.

The purpose of this paper is to describe the potential for producing selected composites from waste wood, paper, and plastics. Discussed first is the availability of waste materials in the municipal solid waste stream and the desirability of developing ways to recycle these materials into useful, high-performance, value-added composites. Methods for making selected composites are then described, and discussion is offered on product properties and attributes. Finally, research and development needs are outlined that will maximize the benefits of using recovered waste materials for composite products.

MUNICIPAL SOLID WASTE AS SOURCE OF MATERIAL FOR COMPOSITES

A considerable amount of data is related to the inventory of the U.S. municipal solid waste (MSW) stream. In 1988, paper and paperboard, wood, and plastics in the MSW stream accounted for approximately 71.8, 6.5, and 14.4×10^6 tons, respectively. These figures account for nearly 60% of MSW. By the year 2000, these figures are expected to increase to 96.1, 8.4, and 21.1×10^6 tons annually (EPA 1990). In addition to the wood fiber in the MSW stream, low-grade wood, wood residues, and industry-generated wood waste in the form of sawdust, planer shavings, and chips are now being burned or otherwise disposed.

Many problems are associated with the use of waste materials, including collection, analysis, separation, clean up, uniformity, form, and costs. Assuming that these problems can be overcome on a cost-effective basis, some of the resultant reclaimed materials should be useful ingredients for a range of valuable composites.

Source separation and recycling not only extend the life of landfills by removing materials from the MSW stream but also make available large volumes of valuable raw m aterials for use by industry in place of virgin *resources.* Industrial use of such materials reduces both costs for raw materials and the energy it takes to make a finished product (New York Legislative Commission 1986). The main requirements are that the recycled ingredients meet the quality and quantity of the consuming production operation.

THERMOFORMABLE WOOD-PLASTIC COMPOSITES

Thermoformable composites are classified into two general types based on the manufacturing process: melt-blend and air-laid nonwoven mat formation. Both processes allow and tolerate differences in composition and in the lignocellulosic component.

Currently, the primary application of thermoformed composites, both melt-blend and air-laid nonwoven, is for interior door panels and trunk liners in automobiles. Additional large-volume, low-to-moderate cost applications are expected in areas such as packaging (trays, cartons), interior building panels, and door skins.

For many applications, it is certain that virgin ingredients can be replaced by some recycled ingredients in melt-blend and nonwoven mat formation. For example, the thermoplastic polymer might be totally or partially replaced by high-density polyethylene (HDPE) from milk bottles, polyethylene terephthalate (PET) from beverage bottles, or even nonsegregated plastic mixtures from MSW. Large quantities of a variety of industrial waste plastics are also available and should be considered. The virgin lignocellulosic component might be replaced by fibers from wastepaper or waste wood. These substitutions offer potential benefits in reducing both MSW and the cost of the composite processes. In some cases, the properties of the composite will probably be improved, for example, by substituting wastepaper fibers for wood flour in the melt-blend process.

The following sections are not intended *to* be a comprehensive review of recent research on wood fiber-thermoplastic composites. Described are the effects of some important composition and processing variables in the- composite processes, including preliminary indications of the effects of recycled ingredients.

Melt-Blend Composites

The use of lignocellulosic fillers in plastics is widely known and practiced. Thermosetting plastics have been filled with wood flour since the turn of the century. The fillers have served mainly as extenders to the m ore expensive plastic resins. Recent developments have increased the interest in mixing wood flour and fibers into thermoplastics as both fillers and reinforcements.

A typical composition for a thermoplastic melt-blend composite is 40 to 60 weight percent wood flour or cellulose pulp fiber with a powdered or pelletized thermoplastic, such as polypropylene or polyethylene. In the melt-blend process, the wood-based fiber or flour is blended with the melted thermoplastic matrix by high-intensity shearing or kneading. Currently, the primary commercial processes employ twin-screw extruders or high shear thermokinetic-type mixers. The mixture is extruded into sheets or pelletized for subsequent operations. Limits on the melt viscosity of the mixture restrict the amount of fiber or flour to about 50 weight percent. Fiber length is limited by fiber breakage as a result of the high shear forces during the melt-blend process.

Publications are beginning to appear on the effect of recycled ingredients on the behavior of melt-blend lignocellulosic-polyolefin composites. Selke and colleagues (1988) showed that composites from aspen fiber and once-recycled blow-molding HDPE from m ilk bottles possessed essentially equivalent strength and modulus properties as did those of composites made from virgin HDPE; however, impact energy was reduced. Woodhams and others (1991) found that composites made from polypropylene and pulp fibers or fiberized old newspaper possessed strength and impact properties similar or superior to those of composites made from wood flour-polypropylene systems. In preliminary work at the Forset Product Laboratory (FPL), we compared the properties of wood flour-polypropylene and wood flour-HDPE systems with properties of a fiberized old newspaper-HDPE composite. The differences between wood flour-polypropylene and wood flour-HDPE systems were qualitatively consistent with expectations based on the lower strength and greater flexibility of HDPE relative to polypropylene. Also, strength was improved by substituting fiberized old newspaper for wood flour.

Properties of Selected Melt-Blend Composites

Scientists at the FPL believe a great opportunity in thermoformable wood-plastic composites lies in the area of the melt-blend process. Research in this area has focused on process parameters, coupling agents, and product properties. Both recycled and virgin materials have been tested.

The primary commercial product made by the melt-blend process is an extruded blend of 50% virgin polypropylene (PP) and wood flour (WF). Early testing at FPL (unpublished) and at the University of Toronto (Woodhams, et al. 1991) showed that certain properties could be increased by substituting old newspaper fiber or cellulose fiber for the wood flour. Tests were made at the FPL to determine this effect more thoroughly, as well as the effect of coupling agents and the use of recycled HDPE from old milk jugs.

Materials- - The PP was Fortilene 1602[1] from Solvay Polymers, Inc. (Deer Park, Texas). The HDPE was recycled from milk bottles. The wood flour was western pine (-40, +80 mesh) from American Woodfiber Co, (Schofield, Wisconsin). Cellulose fiber was Solka-Floc BW-40 from James River Corp. (Hackensack, New Jersey). The old newsprint (ONP) was hammermilled to 15-mm flakes. The coupling agent used was E-43 powder, a maleated PP. obtained from Eastman Chemical Products. , Inc.

Methods--A 1-L thermokinetic high-shear mixer (K-mixer) was used to blend the composites. The fillers were mixed alone for 1 min. The polymers were then added and mixed for an additional minute. The HDPE composites were ejected from the K-mixer at a temperature of 185°C at 2,800 rpm. The PP composites were ejected at 200°C at 3,000 rpm. The material was then injection molded, and the specimens were tested according to ASTM standards (ASTM 1984a,b). Results are shown in Table 1.

Melt-Blend Composite Conclusions -- Melt blending the polymers has a significant negative effect on impact strengths. In most cases, however, the tensile strength values were improved. Compared with the wood flour, both the BW-40 fibers and the ONP fibers improved the tensile strengths of composites with either polymer. However, the fibers increased the impact energy with PP but decreased the impact with HDPE. This difference may be due to a better compatibility of the E-43 with PP than with HDPE. Consistent with the properties of the unfilled polymers, the filled PP composites had greater

----

[1]The use of trade or firm names anywhere in this publication is for reader information and does not imply endorsement by the U.S. Department of Agriculture of any product or service.

Table 1.--Mechanical properties of selected melt-blend composites using E-43 as a coupling agent.[a]

Polymer	Filler	Izod Impact Energy (ASTM D256-84) Notched (J/m)	Izod Impact Energy (ASTM D256-84) Unnotched (J/m)	Tensile Tests (ASTM D638-84) MOE (GPa)	Tensile Tests (ASTM D638-84) Strength (MPa)
PP	WF	18.7	72	3.72	34.1
HDPE	WF	36.4	81	2.60	27.7
PP	BW-40	24.7	114	4.80	48.2
HDPE	BW-40	30.7	68	3.78	36.3
PP	ONP	20.8	109	4.89	47.1
HDPE	ONP	28.6	73	3.79	37.6
Virgin PP		16.0	640	--	33.1
Virgin HDPE		130.0	--	--	29.6

[a]--, not available.

tensile strength and modulus values and lower notched impact energy values than did the filled HDPE composites.

Nonwoven Mat Composites

In contrast to the melt-blend process, nonwoven mat composite technology involves room temperature air mixing of lignocellulosic fibers and fiber bundles with thermoplastic fibers or granules. The resultant mixture passes through a needling step that produces a low-density mat in which the fibers are mechanically entangled. The mat is then shaped and densified by a thermoforming step. With this technology, the amount of lignocellulosic fiber can be greater than 90 weight percent. In addition, the lignocellulosic fiber can be precoated with a thermosetting resin, for example, phenol-formaldehyde. After thermoforming, the product possesses good temperature resistance. Because longer fibers are required, this product can achieve better mechanical properties than that obtained with the melt-blend process. However, high wood fiber contents lead to increased moisture sensitivity.

Numerous articles and technical papers have been written, and several patents have been issued on both the manufacture and use of nonwoven fiber webs containing combinations of textile and cellulosic fibers. This technology is particularly well-known in the consumer products industry. For example, Bither (1980) found that polyolefin pulps can serve as effective binders in nonwoven products.

Brooks (1990) published a historical review of the development, production, and use of air-laid, nonwoven, moldable mat processes and products. The first moldable wood product using the wet slurry process was developed by Deutche Fibrit during 1945 to 1946 in Krefeld, West Germany (Brooks 1990). A moldable cellulose composition containing pine wood resin was patented by Roberts (1955) as well as a process for producing molded products from this composition (Roberts 1956). The composition consisted of a mixture of comminuted cellulose material and at least 10% of a thermoplastic pine wood resin that was derived from the solvent refining of crude rosin. Both of Roberts' patents were assigned to the Weyerhaeuser Company.

From 1966 to 1968, a series of patents (Caron and Grove 1966a,b,c; Grove and Caron 1966) were issued and assigned to the Weyerhaeuser Company. These patents cover the use of a wood fiber-thermoplastic resin system in conjunction with a thermosetting resin system. In the early 1970's, Brooks developed a process that produced a very flexible m at using a thermoplastic Vinyon fiber in combination with a thermosetting resin system (Brooks 1990). The m at was fed through an oven to melt and set the Vinyon fiber without affecting the setting of the thermosetting resin component. This process was patented by Doerer and Karpik (1984) and assigned to the Van Dresser Corporation.

Properties of Selected Nonwoven Mat Composites

The FPL has made and tested a wide variety of nonwoven air-laid composite materials. These materials include a wide variety of recycled and virgin wood fibers, recycled and virgin synthetic fibers, agricultural fibers, phenolic powders, and plastic granules. Almost any dry fiber can be air-laid into a mat that can be subsequently pressed into a panel or molded shape. For the purpose of this paper, only a brief discussion is given on several selected materials.

A comparison was made between two wood fibers derived from virgin Western Hemlock (Tsuga heterophylla) and recycled demolition wood. The wood fibers were blended with either recycled *or* virgin polyester (PET) fibers and a phenol formaldehyde resin.

Materials and Methods--Hemlock wood fibers, obtained from Canfor, LTD (Vancouver, B.C.) were produced from 100% pulp-grade chips, steamed for 2 min. at 0.759 MPa steam pressure, disk refined, and flash dried at 160°C in a tube dryer. This processing sequence produced fibrous strands made of individual fibers, pieces of fiber, and fiber bundles. In this report, these fibrous strands are referred to as fibers. The demolition wood fiber, obtained from Wood Recycling, Incorporated, (Peabody, Massachusetts) was prepared from wood waste taken from the Boston area. The wood waste was shredded, hammermilled, and cleaned before it was pressurized refined in a manner similar to the hemlock.

Two types of PET fibers were obtained from E.I. DuPont deNemours, Inc. Wilmington, Delaware). The virgin fibers were 5.5 denier (6.1×10^{-7} kg/m), 38 mm long, crimped, with a bonding temperature greater than 215°C. The recycled fibers were of a similar configuration. The phenolic resin had a solids content of 52%, a viscosity of 50 to 100 cps (0.05 to 0.1 Pa-s) at 25°C, and a pH of 9.5 to 10.0.

The processing began by spraying the phenolic resin onto the wood fibers at a 10% weight basis. The wood and plastic fibers were then mixed by passing them through a spiked drum, transferred through an air stream to a moving support bed, and subsequently formed into a continuous, low-density mat of intertwined fibers. The mat then went through a needling process where barbed-shank needles pass through the mat thickness, and in so doing, resulted in an increased interlocking of the fibers. The mats were 330 mm wide and 12 to 18 m long. The mats were then cut into 330- by 330-mm squares. The squares were then stacked to get the required panel weight. A manually controlled, steam-heated press was used to press all panels. They were pressed at 190°C to a thickness of 3.2 mm and a specific gravity of 1.0.

Panels were conditioned at 65% relative humidity at 20°C. After conditioning, the panels were cut into test specimens and tested in conformance with ASTM (1987) or TAPPI (1989). Results are shown in Table 2.

Table 2. -- Mechanical and physical properties of air-formed composites from recycled and virgin materials.[a]

Property	Panel composition: 80% Hemlock 10% V-PET 10% resin	80% Hemlock 10% R-PET 10% resin	80% Demo 10% V-PET 10% resin	80% Demo 10% R-PET 10% resin	American Hardboard Association Standard(1982)
Static Bending MOR (MPa)	50.6	47.1	43.2	47.8	31.0
Static Bending MOE (GPa)	3.66	4.36	3.23	3.74	--
Tensile Strength (MPa)	33.0	28.4	28.3	30.0	15.2
Tensile MOE (GPa)	4.84	5.12	4.26	4.56	--
Impact energy (J)	36.1	28.7	34.2	30.7	--
24-h water soak/ absorption (%)	43.3	41.3	48.2	44.1	35.0
24-h water soak thickness swell (%)	25.2	22.3	29.8	26.9	25.0
Linear Expansion Oven dry to					
30% RH (%)	0.25	0.21	0.20	0.20	--
65% RH (%)	0.46	0.44	0.43	0.45	--
90% RH (%)	0.65	0.70	0.64	0.71	--

[a] Demo is recycled demolition wood; RH is relative humidity; --, not available.

Nonwoven Mat Composite Conclusions -- Air-laid nonwoven mat technology permits the use of a wide range of lignocellulosic and synthetic fibers. The lignocellulosic components can range from recycled or virgin wood materials to agricultural fibers. The lignocellulosic component can be chemically modified to improve the performance of the composite in adverse conditions, as appropriate. Products can be made from 100% plastic fibers, 100% lignocellulosic fibers, or m any different combinations of the two materials. Thermosetting resins can be either coated on the lignocellulosic fibers or added in powder form during web formation. Additionally, granulated plastics may be added.

RESEARCH AND DEVELOPMENT NEEDS

The USDA Forest Service, by virtue of its role as steward of the National Forests, its research mission, and its longstanding expertise in wood-based composites and recycling research, is actively engaged in a high-priority research program on alternative uses for recovered materials from the MSW stream. The FPL research program is focusing on developing value-added composites from waste materials, including wood-plastic fiber composites, dry-formed wood fiber-based composites, and composites fabricated with inorganic binders. For each of these program areas, FPL scientists are

1. developing methods for converting recovered fibers into forms suitable for subsequent processing into alternative end-use applications,
2. optimizing laboratory methods for making prototype products,
3. developing a performance database, including determining mechanical and physical properties of wood-based products and conducting analytical tests,
4. determining the potential for recycling composites with minimal loss of properties, and
5. studying product applications and economic viability of alternative end-use applications.

In each of these research areas, economic and laboratory studies are being conducted on an iterative basis as a means of setting research priorities and guiding process development. The research will focus on the components of successful recycling systems through determining the supply and availability of waste wood fiber, analyzing the economic efficiency of processing concepts, and studying the market potential for products made from recovered fiber. Studies will examine the effect of new technologies on the environment, such as the projected impact on the landfill burden and on the quality of the air, forests, soil, and water. They will also examine the broader economic impact of these technologies on timber markets and trade.

CONCLUDING REMARKS

Recycling is a critical element in the long-term management of renewable resources. Using recycled wood and fiber for wood-based composites presents tremendous opportunities for growth, progress, and further industry competitiveness in a world that is rapidly consuming m any nonrenewable resources at an ever-increasing rate.

ACKNOWLEDGMENTS

I thank John Youngquist, Roger Rowell, George Myers, Craig Clemens, and Anrzej Krzysik, all of the USDA Forest Service, Forest Products Laboratory.

REFERENCES CITED

AMERICAN HARDBOARD ASSOCIATION. 1982. Voluntary product standard. Basic Hardboard ANSI/AHA A135.4-1982. Palatine, IL.

ASTM. 1984. Standard test methods for impact resistance of plastics and electrical insulating materials. ASTM D256-84. American Society for Testing and Materials, Philadelphia, PA.

ASTM. 1984. Standard test methods for tensile properties of plastics. ASTM D638-84. American Society for Testing and Materials, Philadelphia, PA.

ASTM. 1987. Standard methods of evaluating the properties of wood-base fiber and particle panel materials. ASTM D1037-87. Vol. 04.09. American Society for Testing and Materials, Philadelphia, PA.

BITHER, P. 1980. Polyolefin pulps as nonwoven binders. Proceedings, Air-Laid and Advanced Forming Conference. November 16-18, 1980. Hilton Head Island, SC.

BROOKS, S. HUNTER. 1990. Air lay nonwoven moldable mat process and products produced by using this mat process. Proceedings, 1990 Tappi Nonwovens Conference. pp. 87-108.

CARON, PHILLIP E. and GROVE, G.A. 1966a. Process for manufacturing moldable fibrous panels. U.S. Patent 3,230,287. U.S. Patent Office. Washington, D.C.

CARON, PHILLIP E. and GROVE, G.A. 1966b. Production of hot-pressed three-dimensional fiber articles. U.S. Patent 3,261,898. U.S. Patent Office. Washington, D.C.

CARON, PHILLI[E. and GROVE, G.A. 1966c. Method of die-baking moldable wood fiber parts. U.S. Patent 3,265,791. U.S. Patent Office. Washington, D.C.

DOERER, RICHARD P. and KARPIK, J.T. 1984. Moldable fibrous mat and product. U.S. Patent 4,474,846. U.S. Patent Office. Washington, D.C.

EPA. 1990. Characterization of municipal solid waste in the United States: 1990 update. Report EPA/530-SW-90-042. U.S. Environmental Protection Agency. Washington, DC.

GROVE, GENE A. and CARON, P.E. 1966. Method of making a moldable wood fiber mat with metal insert. U.S. Patent 3,279,048. U.S. Patent Office. Washington, D.C.

KOVACS, W. L. 1988. The coming era of conservation and industrial utilization of recyclable materials. Ecology Law Quarterly 15(4):537-625.

NEW YORK LEGISLATIVE COMMISSION on Solid Waste Management. 1986. The economics of recycling municipal waste.

ROBERTS, J.R. 1955. Moldable cellulose composition containing pine wood resin and molded product. U.S. Patent 2,714,072. U.S. Patent Office. Washington, DC.

ROBERTS, J.R. 1956. Process of forming molded cellulose products. U.S. Patent 2,759,837. U.S. Patent Office. Washington, D.C.

SELKE, S.E.; YAM, K.L.; GOGOI, B.; and LAI, C.C. 1988. Compounding wood fibers and recycled high density polyethylene using a twin-screw extruder. Abstract. Cellulose, Paper and Textile Div. ACS Meeting. June.

TAPPI. 1989. TAPPI test methods. Puncture test of containerboard. TAPPI T803 om-88, Vol. 2. Technical Association of the Pulp and Paper Industry, Atlanta, GA.

WOODHAMS, R.T.; LAW, S.; BALANTINECZ, J. J. 1991. Proceedings, Wood Adhesives Symposium; 1990 May 16-18; Madison, WI: 177-182.

In: Proceedings of the Pacific Rim bio-based composites symposium; 1992 November 9–13; Rotorua, New Zealand. FRI Bulletin 177. Rotorua, New Zealand: New Zealand Forest Research Institute; 1992: 326–335.

on recycled paper