Chapter 4

Lignocellulosic–Plastic Composites from Recycled Materials

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Reductions are urgently needed in the quantities of municipal solid waste (MSW) materials that are currently being landfilled. Waste wood, waste paper, and waste plastics are major components of MSW and offer great opportunities as recycled ingredients in wood fiber-plastic composites. With cooperation from the U.S. Environmental Protection Agency, a research and development program has been initiated at the Forest Products Laboratory and University of Wisconsin–Madison to investigate the processing, properties, and commercial Potential of composites containing these recycled ingredients. Two processing technologies are being employed—melt blending and nonwoven web. Some past research studies axe briefly reviewed to illustrate the behavior of wood fiber- polyolefin composites.

Use of the word "waste" projects a vision of material with no value or useful purpose. However, technology is evolving that holds promise for using waste or recycled wood and plastics to make an array of high-performance products that are, in themselves, potentially recyclable. Preliminary research at the USDA Forest Service, Forest Products Laboratory (FPL), and elsewhere *(1,2)* indicates that recycled plastics such as polyethylene, polypropylene, or polyethylene terephthalate can be combined with wood fiber waste to make useful reinforced thermoplastic composites. Advantages associated with these composite products include lighter weight and improved acoustic, impact, and heat reformability properties—all at a cost less than that of comparable products made from plastics alone. In addition, previous research has shown that composite products can possibly be reclaimed and recycled for the production of second-generation composites *(3).*

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Since the turn of this century, the United States has been dubbed "the throw-away society" *(4),* generating approximately 50 percent of the world's solid and industrial waste. If present trends continue, this nation's solid-waste stream will increase from 157.7 × 10^6 t in 1986 to 192.7 × 106 t by the year 2000 *(5).* It is increasingly difficult to dispose of the growing volume of municipal solid wastes (MSW) in landfills because most people will not tolerate MSW in their neighborhood. In the next 15 years, 75 percent of our landfills will be closed, and by the the 2000, this nation will be short 56 × 10^6 t per year of disposal capacity. As political and conservation pressures increase, it is anticipated that the recovery of recyclable materials from the solid-waste stream will increase from 16.9 to 23.9 × 10^6 t by the year 2000 *(5).*

In many uses, wood fiber-plastic composites can be opaque, colored, painted, or overlaid. Consequently, recovered fibers or resins used in these composites do not require the extreme cleaning and refinement needed when they are to be used as raw materials for printing paper or pure plastic resins. This fact greatly reduces the cost of wood-fiber plastic composites as raw materials and makes composite panels or molded products an unusually favorable opt ion for the recycling of three of our most visible and troublesome classes of MSW—newspapers, waste wood, and plastic bottles.

Research at the FPL on wood fiber-plastic composites focuses on two very different manufacturing technologies—melt blending (for example, extrusion and injection molding) and air laid or nonwoven web. A host of new natural fiber-synthetic plastic fiber products can be made because of the increased processing flexibility inherent in both of these technologies. These products can be produced in various thicknesses, from a thin material of 3 mm to structural panels up to several centimeters thick. A large variety of applications are possible because of the many alternative configurations of the product. A list of potential products could include

1. storage bins for crops or other commodities,
2. furniture components, including both flat and curved surfaces,
3, automobile and truck components,
4. paneling for interior wall sections, partitions, and door systems,
5. floor, wall, and roof systems for light-frame construction, and
6. packaging applications, including containers, cartons, and pallets.

Taken together, these two processing techniques provide options for balancing performance properties and costs, depending upon the product application under consideration.

The overall goal of this paper is to illustrate the potential that currently exists for manufacturing thermoformable composites from waste materials such as waste wood, paper, and plastics. We first discuss the availability of such waste materials from MSW streams and the desirability of developing the means to recycle them. We then describe how these composites are made and point out why materials from the MSW should be suitable ingredients. Next, we illustrate the properties of such composites by describing some recent research on the effects of composition and processing variables, using both virgin and recycled ingredients. We follow this with an outline of the research and development needed to convert wood fiber and plastics into durable products.

Municipal Solid Waste as a Source of Lignocellulosic Fiber and Plastics

A considerable amount of data are related to the inventory of the U.S. MSW stream (Table I). In 1986, paper and paperboard, wood, and plastics in the MSW stream accounted for approximately 65, 5.8, and 10.3 × 106 t, respectively. By the year 2000, these figures are expected to increase to 86.5,6.1, and 15.7 × 10^6 t annually. In addition to the wood fiber in the MSW stream, vast quantities of 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 of.

Table I. Estimated Distribution of Materials in Municipal Solid Waste in 1986[a]

Source	Amount in municipal solid waste (Percentage)	Weight ($\times 10^6$ t)
Paper and paperboard	35.6	56.1
Yard waste	20.1	31.7
Metals	8.9	14.0
Food waste	8.9	14.0
Glass	8.4	13.3
Plastics	7.3	11.5
Textiles	2.0	3.2
Wood	4.1	6.5
Rubber–leather	2.8	4.4
Miscellaneous inorganics	1.9	3.0
Total	100.0	157.7

[a]SOURCE: Adapted from ref. 6.

The data in Table I include all the residential waste products but not all the industrial waste materials. Data are available for the total volume or weight of certain wood-based products in the MSW stream, such as paper, packaging, and pallets, but only incomplete information is available for timber thinnings, leaves, industrial production wastes, bark, and sawdust. These latter categories of wood waste also represent potentially valuable sources of raw materials.

A number of problems are associated with the use of these waste materials, some of which include 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, from low-cost, high-volume materials to high-cost, low-volume materials for a wide range of end-use applications.

Source separation and recycling not only extend the life of landfills by removing materials from the MSW stream, but they also make available large

volumes of valuable raw materials 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 *(4)*. The main requirement is that the recycled ingredients meet the quality and quantity requirements of the consuming production operation.

Thermoformable Composites as Outlets for Waste Paper, Wood, and Plastics

Two general types of thermoformable composites exist, distinguished by their very different manufacturing processes. Both processes allow and require differences in composition and in the lignocellulosic component. The two processes used to produce thermoformable composites are melt blending and air laying or nonwoven mat formation.

A typical composition for a melt-blended 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-blending process, the wood-based fiber or flour is blended with the melted thermoplastic matrix by shearing or kneading. Currently, the primary commercial process employs twin screw extruders for the melting and mixing; the mixture is extruded as sheets that are subsequently shaped by thermoforming into the final product. Limits on the melt viscosity of the mixture restrict the amount of fiber or flour to about 50 weight percent as well as the length of the fibers that can be used. In any event, fiber length is limited by fiber breakage as a result of the high shear forces during melt mixing.

In contrast, the nonwoven mat technology involves a room temperature air mixing of lignocellulosic fibers (or even fiber bundles) with fibers of the thermoplastic. The resultant mixture passes through a needling step that produces a low-density mat in which the fibers are mechanically entangled. That mat must then be 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-blending process. In contrast, high wood fiber contents lead to increased moisture sensitivity.

It is virtually certain that virgin ingredients can be replaced by some recycled ingredients in melt blending and nonwoven mat formations for many applications. For example, the thermoplastic polymer might be totally or partially replaced by high-density polyethylene (HDPE) from milk bottles, polyethyleneterephthalate (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 waste paper or wood. These substitutions offer potential benefits in reducing both the MSW problem and the cost of the composite processes. In some cases, we can also reasonably expect

the properties of the composite to be improved; for example, by substituting waste paper fibers for wood flour in the melt-blending process.

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

Recent Research on Wood Fiber-Thermoplastic Composites

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

Composites Made by Melt Blending. The 1980s brought a resurgence of research into various aspects of melt-blended composites made from wood-based flour or fiber in virgin thermoplastic matrices. For example, Kokta and his colleagues have published numerous papers in this area, emphasizing improvements in the tiller-matrix bond through coupling agents and grafting of polymers on cellulosic fiber surfaces *(7–9)*. Klason and colleagues carried out extensive investigations on the effects of several polymer and fiber types and the influence of a variety of processing aids and coupling agents *(10,11)*. Woodhams and others examined several types of pulp fiber in polypropylene (PP) and HDPE *(12,13)*. Shiraishi and colleagues showed improvements in mechanical properties as a consequence of using high-molecular-weight maleated PP instead of normal PP *(14,15)*. Finally, Maiti and Hassan measured the effects of wood flour on the melt rheology of PP *(16)*.

At the FPL and the University of Wisconsin (UW), we have investigated in some detail the influence of a low-molecular-weight maleated PP (Eastman's Epolene E-43) (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.) on the mechanical and physical properties of wood flour and PP-extruded composites. For example, Figure 1 shows that small amounts of E-43 produce small but statistically significant improvements in maximum flexural strength and that those effects are greater when smaller wood flour particles are present *(17)*. Moreover, Figure 2 indicates that the major gains in strength are produced at less than 2 percent E-43 and that the optimum extrusion temperature for this property is about 210°C *(18)*. In contrast, Figure 3 shows that notched impact energy is significantly reduced by both the E-43 and the high extrusion temperature *(18)*. Other experiments indicated that the E-43 probably is not acting as a true coupling agent but instead has some effectiveness as a dispersing agent *(19)*.

Publications are beginning to appear on the effect of recycled ingredients on the behavior of melt-blended lignocellulosic-polyolefin composites. Selke and colleagues showed that composites from aspen fiber and once-recycled blow-molding HDPE from milk bottles possessed essentially equivalent strength and

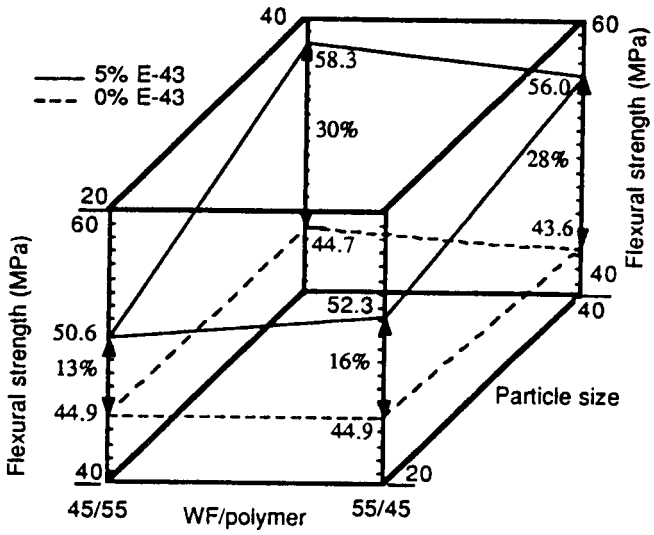

Figure 1. Effect of wood flour particle size, wood flour to polymer ratio, and Epolene E-43 on flexural strength of melt-blended composites. (WF is wood flour.) SOURCE: Adapted from ref. 17.

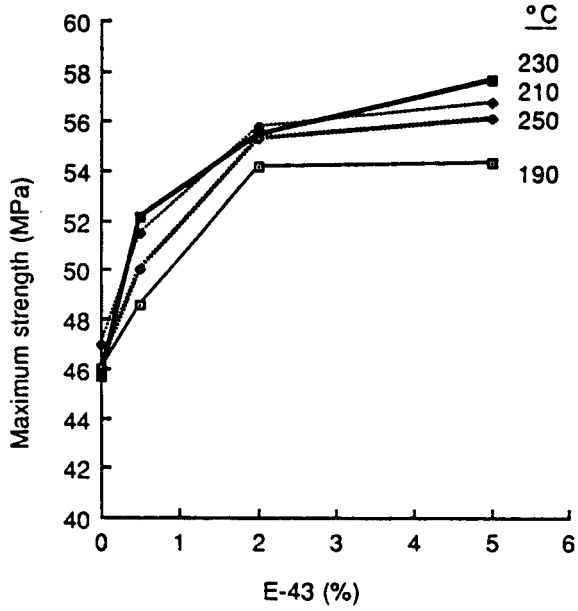

Figure 2. Effect of Epolene E-43 concentration and extrusion temperature on flexural strength of melt-blended wood flour-polypropylene composites. SOURCE: Adapted from ref. 18.

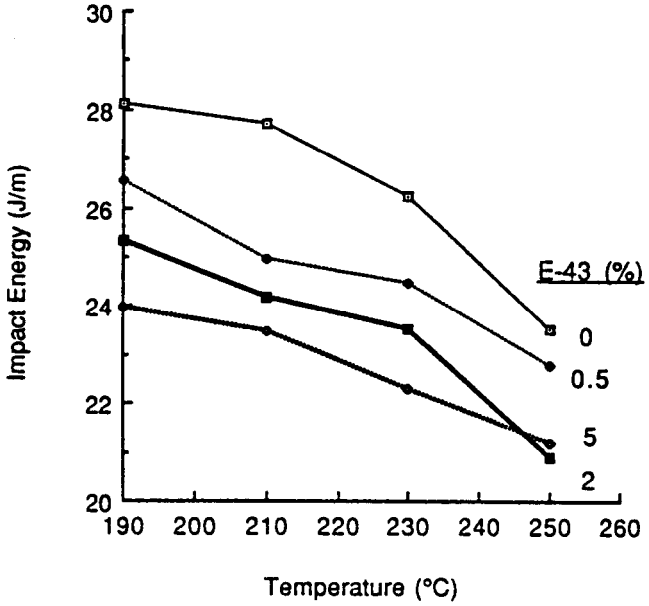

Figure 3. Effect of Epolene E-43 concentration and extrusion temperature on notched impact energy of melt-blended wood flour-polypropylene composites. SOURCE: Adapted from ref. 18.

modulus properties as those of composites made with virgin HDPE; however, impact energy was reduced *(1,2)*. Woodhams and others found that composites made from PP and pulp fibers or fiberized newspaper (ONPF) possessed strength and impact properties that were very similar and apparently much superior to those for wood flour (WF)–PP systems *(13)*. In preliminary work at the FPL, we compared the properties of WF–PP and WF–HDPE systems with an ONPF–HDPE composite (Figures 4 and 5). The differences between WF-PP and WF-HDPE are qualitatively consistent with expectations based on the lower strength and greater flexibility of HDPE relative to PP. Also, strength has been improved by substituting ONPF for WF.

Composites Made by Nonwoven Mat Technology. 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, Sciaraffa and others *(20)* have been issued a patent for producing a nomvoven web that has both fused spot bonds and patterned embossments for use as a liner material for disposable diapers. Bither *(21)* has found that polyolefin pulps can serve as effective binders in nonwoven products. Many additional references could be cited in this area.

S. Hunter Brooks *(22)* has published a review of the history of technology development for the production and use of moldable wood products and 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 *(22)*. A moldable cellulose composition containing pine wood resin was patented by Roberts *(23)* in 1955, and a process for producing molded products from this composition was patented by Roberts *(24)* in 1956. This composition consisted of a mixture of comminuted cellulose material and at least 10 percent of a thermoplastic pine wood resin 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 *(25-29)* was issued to Caron and others 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 1970s, Brooks developed a process that produced a very flexible mat using a thermoplastic Vinyon fiber in combination with a thermosetting resin system *(22)*. The mat 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 in 1984 by Doerer and Karpik and was assigned to the Van Dresser Corporation *(30)*.

Brooks also developed an interesting method of recycling waste cellulosic materials for the production of medium-density fiberboard and paper *(31)*. After being shredded, sorted from other waste materials like plastic and metal, and steamed, the cellulosic fibers and fiber bundles are abraided under heat and pressure to break down any hydrogen bonds and to soften any lignin and other resins. The resultant cellulose fibers are then mixed with resin, formed

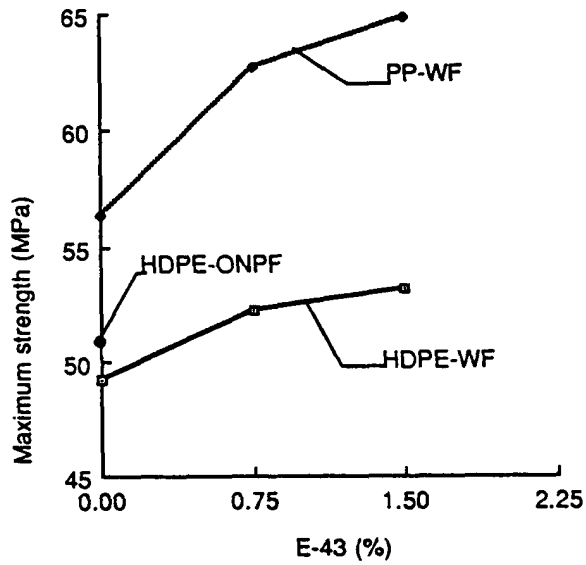

Figure 4. Flexural strength of several melt-blended composites at 50/50 weight ratio filler to polymer. (WF is wood flour, ONPF is newspaper fiber, PP is polypropylene, HDPE is high-density polyethylene.)

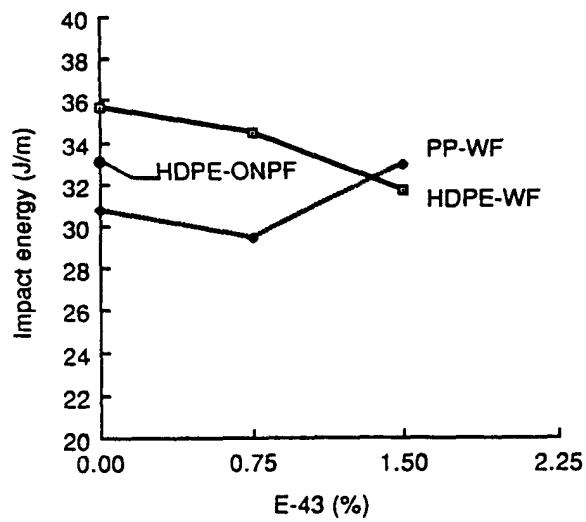

Figure 5. Notched impact energy of several melt-blended composites at 50/50 weight ratio filler to polymer. (WF is wood flour, ONPF is newspaper fiber, PP is polypropylene, HDPE is high-density polyethylene.)

into a mat, and consolidated under pressure to form flat fiberboard and paper products.

A general review of the opportunities for combining wood with nonwood materials was written by Youngquist and Rowell *(32)*. This review included a discussion of the materials and the properties of composites consisting of wood-biomass, wood-metals, wood-plastics, wood-glass, and wood-synthetic fibers.

In a recently published paper, Youngquist and others *(33)* reported on the mechanical and physical properties of wood-plastic fiber composites made with an air-formed dry-process technology. This paper reported the effect of species, the ratios of WF to PP, and the type of plastic fiber or plastic fiber-thermosetting resin blends on the mechanical and dimensional stability properties of pressed panels having a density of 1 g/m^3.

Krzysik and Youngquist *(34)* reported on the bonding of air-formed wood-polypropylene fiber composites using maleated polypropylene as a coupling agent between the hydrophilic wood and the hydrophobic polyolefin materials.

Tables II and III present mechanical and physical property data obtained at FPL on nonwoven webs made into composites having densities of 1.0 and 1.2 g/m^3, respectively.

In many cases, the mechanical properties for each of the three formulations within a density grouping (e.g., 1.0 g/m^3) were statistically different from one another. As the panel densities increased, the level of magnitude of the mechanical properties generally increased correspondingly. Also note that the dimensional stability property levels for each of the formulations at the two density levels did not exhibit large variations when comparing one density level to another. In most cases, as each formulation changed, all properties, with a few exceptions, increased in that order. The use of a thermoplastic polyolefin-like polypropylene greatly improved the dimensional stability properties of the composite, compared to the polyester copolymer-containing composite. These results can probably be explained by the fact that the polypropylene melts, and to some extent, partially encapsulates the wood fibers. In all cases, the ability to absorb impact energy of the polyester copolymer-containing composite was superior to that of the polypropylene-containing composite. This can be attributed to the fact that the polyester maintains a fibrous matrix, whereas the polypropylene fibers melt and flow under heat and pressure. Phenolic resin, in combination with wood and polyester homopolymer fibers, forms a composite that has greatly improved mechanical properties compared to that of the other two composite formulations tested.

A number of preliminary trials have been conducted at the FPL, using recycled office wastepaper, shredded old newspapers, dry fiberized old newspapers, and fiberized demolition waste wood. The raw paper materials, which did not have the ink removed, were reduced to a suitable form using a number of reduction methods, as noted previously. The demolition waste was first sorted mechanically, and then manually, to remove nonwood materials, washed, and then fiberized using a pressurized refiner. These recycled woocl-based fibers were then air mixed with virgin polyester or polypropylene, transferred by an air stream to a moving support bed, needled, and subsequently formed into a continuous, low-density mat of intertwined fibers. When polyester fibers were

Table II. Properties of Nonwoven Web Composite Panels[a] (Density 1.0 g/m^3)

Property measured[c]	Formulation[b]		
	90H/10PE	90H/10PP	80H/10PE/10PR
Static bending strength (MPa)	23.3	25.2	49.3
Static bending modulus (GPa)	2.82	2.99	3.57
Tensile strength (MPa)	13.5	12.5	27.7
Tensile modulus of elasticity (GPa)	3.87	4.20	5.07
Internal bond (MPa)	0.14	0.28	0.81
Impact energy (J)	26.7	21.5	34.3
24-h water-soak thickness swell (percent)	60.8	40.3	21.8
24-h water-soak water absorbency (percent)	85.0	54.7	45.1
2-h water-boil thickness swell (percent)	260.1	77.5	28.2
2-h water-boil water absorbency (percent)	301.6	99.5	55.7
Linear expansion at 65 percent relative humidity (percent)[d]	0.38	0.25	0.76

[a]Averages connected by bars located below the numbers are not statistically different; averages not connected by bars are statistically different at 0.05 significance level.
[b]90H/10PE = 90 percent hemlock and 10 percent polyester copolymer, 90H/10PP = 90 percent hemlock and 10 percent polypropylene, and 80H/10PE/10PR = 80 percent hemlock, 10 percent polyester homopolymer, and 10 percent phenolic resin.
[c]Specimens were conditioned at 65 percent relative humidity and 20°C.
[d]Linear expansion values are based on 0 percent moisture content.

Table III. Properties of Nonwoven Web Composite Panels[a] (Density 1.2 g/m³)

Property measured[c]	Formulation[b]		
	90H/10PE	90H/10PP	80H/10PE/10PR
Static bending strength (MPa)	36.5	36.8	76.4
Static bending modulus[d] (GPa)	4.59	3.81	5.42
Tensile strength (MPa)	20.2	17.8	42.2
Tensile modulus of elasticity (GPa)	6.56	6.36	7.80
Internal bond (MPa)	0.28	0.52	1.44
Impact energy[d] (J)	31.1	27.4	36.6
24-h water-soak thickness swell (percent)	57.1	42.7	23.6
24-h water-soak water absorbency (percent)	57.5	37.5	28.4
2-h water-boil thickness swell	245.0	88.3	32.4
2-h water-boil water absorbency	227.3	79.6	38.6
Linear expansion at 65 percent relative humidity (percent)[e]	0.33	0.20	0.86

[a]Values connected by bars located below the numbers are not statistically different; values not connected by bars are statistically different at 0.05 significance level.
[b]90H/10PE = 90 percent hemlock and 10 percent polyester copolymer, 90H/10PP = 90 percent hemlock and 10 percent polypropylene, and 80H/10PE/10PR = 80 percent hemlock, 10 percent polyester homopolymer, and 10 percent phenolic resin.
[c]Specimens were conditioned at 65 percent relative humidity and 20°C.
[d]The only statistically different values were between the 90H/10PP and 80H/10PE/10PR formulations.
[e]Linear expansion values are based on 0 percent moisture content.

used, the wood fibers were first sprayed with a liquid phenolic resin, prior to web formation.

Panel-sized mats were cut from each roll. Six to eight mats were selected according to their weight and stacked. Demonstration prototype panels and molded shapes were then produced using a steam-heated press. These prototype samples appeared to be reasonably successful; therefore, a series of experiments will be conducted so that physical and mechanical properties can be determined on a number of different composites made from a variety of recycled wood raw mat erials.

Research and Development Needs

At the FPL and the UW, we are conducting a program aimed at developing technology to convert recycled wood fiber and plastics into durable products that are themselves recyclable, are environmentally friendly, and will remove the raw materials from the waste stream. This program is being conducted in cooperation with the U.S. Environmental Protection Agency. In support of this goal, we have defined a number of research and development needs. Some of these are as follows:

1. Processing methods—Processing methods must be improved or developed, first, for converting waste wood and waste plastics into forms suitable for subsequent melt-blending and nonwoven web processing and, second, for carrying out the actual processing into composites. In the melt-blending technology, for example, the necessary short fibers from waste paper (ONPF) possess very low-bulk density, and this creates difficulties in handling and in feeding to an extruder. Therefore, preblending or pelletizing steps may be desirable. In the nonwoven web process, for example, both the wood and plastic must be converted to fibers or at least to a long, slender form (paper strips or shreds). For either technology, the processing conditions required for good composite properties must be established for different ingredients or even for different forms of the same ingredients.
2. Database—A database is needed that systematically describes the effects of ingredient, formulation, and processing variables on the physical and mechanical properties of the composites. For example, variables must include, plastic and wood component specifications, ratio of plastic to wood components, and presence of additives such as coupling agents or dispersing aids. The properties of interest include those of the melt, such as melt viscosity, melt strength, thermoformability, and those of the final composite, such as strength, modulus, impact resistance, and moisture sensitivity.
3. Recyclability—It is necessary to establish the extent to which the composites formed with recycled ingredients can be recycled into similar products without undue loss in composite properties.
4. Industrial implementation—An essential aspect of the program is the identification of potential processing limitations and the practical utility of the products. This will require cooperation with industry processors and producers and extension of the database to confirm that composite systems are available to meet processing and product specifications.

Concluding Remarks

In any approach to recycling, the government and the private sector must be full cooperative partners. Government cannot logically mandate the increased use of recyclable materials without industry involvement because only the industrial sector has the technical knowledge and equipment to separate and process solid waste and to make useful, economically viable products from these materials. Industry is the market for recycled resources, and it must be a full partner in all aspects of the process.

As a society, we must take a broad look at our opportunities and the responsibilities that go with them. We must be concerned with the reliable performance of products and also the health and safety of those making and using the products. We must be concerned with the prudent use of renewable resources and reduce our use of products that will deplete our nation's or the world's resources. These limitations can be considered as constraints or as opportunities.

We believe that using recycled raw materials for wood-based composites presents tremendous opportunities for growth, for progress, and for further industry competitiveness in a world that is rapidly consuming many of our nonrenewable resources at an alarming rate.

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