PROJECT SUMMARY

COMPOSITES FROM RECYCLED WOOD AND PLASTICS

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ABSTRACT

The ultimate goal of our research was to develop technology to convert recycled wood fiber and plastics into durable products that are recyclable and otherwise environmentally friendly. Two processing technologies were used to prepare wood-plastic composites: air-laying and melt-blending. Research was conducted in (1) developing laboratory methods for converting waste wood, wastepaper, and waste plastics into forms suitable for processing into composites; (2) optimizing laboratory methods for making composite panels from the waste materials; (3) establishing a database on the effects of formulation and bonding agent on physical and mechanical properties of composites; (4) establishing the extent to which the composites can be recycled without unacceptable loss in properties; and (5) reaching out to industry to provide education, to develop applications, and to extend the database. Overall, the program demonstrated that both air-laid and melt-blended composites can be made from a variety of waste wood, wastepaper, and waste plastics. The composites exhibit a broad range of properties that should make them useful in a wide variety of commercial applications. For air-laid composites, the waste materials were demolition wood waste and waste plastics from milk bottles (polyethylene) and beverage bottles (polyethylene terephthalate). Results showed that air-laid composites made from these waste ingredients possessed properties very similar to those of composites made from the virgin ingredients. In addition, air-laid

composites containing 20% reground panels possessed some properties that were superior to those of the original composites. For melt-blended composites, waste materials were wastepaper, polyethylene from milk bottles, and polypropylene from automobile battery cases or ketchup bottles. Waste magazines were slightly inferior to waste newspapers as a reinforcing filler; the properties of composites made from waste newspaper were better than those of composites made from wood flour, which is currently used in some commercial composites. Properties of wood-plastic composites were generally parallel to those of the plastics; thus, different balances in composite properties are possible from using waste plastic. Outreach activities included the organization and presentation of two international conferences on wood fiber-plastic composites, presentations at many conferences, publication of several papers, and several spin-off cooperative studies with industry. One major study with industry demonstrated the commercial feasibility of making melt-blended composites from old newspapers and polypropylene.

STUDIES ON AIR-LAID COMPOSITES

Air-laid (AL) web composites provide options for balancing performance properties and costs, depending upon the application under consideration. However, poor attraction and low interracial bonding between the hydrophilic wood and hydrophobic polyolefin limit the reinforcement imparted to the plastic matrix by the wood component. We compared the mechanical and dimensional stability properties of flat panel composites made from virgin and postconsumer waste wood and plastics in two series of tests:

AL Series 1. PET systems

- Virgin hemlock wood fiber (HF), virgin polyester fiber (VPET), phenolic resin
- HF, recycled PET (RPET), phenolic resin
- Demolition waste wood fiber (DF), VPET, phenolic resin
- DF, RPET, phenolic resin

AL Series 2. HDPE systems

- HF and virgin high-density polyethylene (VHDPE)
- HF and recycled HDPE (RHDPE)
- DF and VHDPE
- DF and RHDPE

Proportions of components were based on ovendry fiber weight. For AL Series 1, the proportions were 80% wood fiber/10% PET/10% phenolic resin. For AL Series 2, the proportions were 60% wood fiber/30% HDPE/5% PET/5% tackifier (E-10).

We also studied the recyclability of air-laid composites by testing the mechanical properties of second-generation panels made from AL Series 2 panels. Two formulations were tested. Each formulation consisted of 30% RHDPE, 5% VPET, and 5% E-10 tackifier. In addition, one formulation had 60% DF and no refiberized first-generation panels; the other formulation had 40% DF and 20% refiberized first-generation panels.

Methods and Materials

Experiments consisted of the following sequence of steps: (a) modifying the FPL air-forming equipment to ensure that uniform machine- and cross-machine direction webs could be produced routinely; (b) converting raw materials into forms suitable for use in this equipment; (c) producing air-formed webs; (d) selecting and stacking the webs to produce mats of a given weight; (e) consolidating the mats in a platen press to produce test panels; (f) cutting test specimens from panels; and (g) testing properties.

For both test series, each data set was tested for normality at the 95% confidence level using Shapiro-Wilk statistical analysis. An analysis of variance was performed and the means were compared at the 95% confidence level using Tukey's method of multiple comparisons.

The raw materials studied in the air-forming portion of this research program fell into three general classes: cellulosic fibers, plastics, and additives.

Cellulosic Fibers

Two basic types of wood fiber were used in the AL series of tests. The first was virgin western hemlock wood fiber (HF), which was produced in a pressurized single-disk refiner from 100% pulp-grade chips. The second was demolition wood fiber (DF), which was derived from waste wood from buildings that had been tom down in the Boston, Massachusetts, area.

Plastics

The virgin polyester (VPET) was 5.5 denier (6.1 x 10^{-7} kg/m), 38 mm long, and crimped, and had a softening temperature greater than 215°C. The recycled polyester fiber, which was spun from recycled soft drink containers, was 6.00 denier, 51 mm long, and crimped. For ail the air-laid experiments, VPET served as a matrix to hold the fibers together within the mat.

The virgin high-density polyethylene (VHDPE) was a blow-molding polymer normally used as a feedstock for plastic milk bottles. The flakes were cryogenically ground to a (-)35 mesh size. The melt flow index of the recycled HDPE was 0.7.

Additives

Liquid phenolic resin was used as the binder for the AL Series 1 panels; it had a solids content of 51% to 53% and a pH of 9.5 to 10.0 at 25°C. The resin was sprayed on the wood fiber at 25°C at a level of 10% solids by weight as it rotated in a drum-type blender.

For the AL Series 2 boards, the wood fiber was blended with granulated HDPE. Previous work had showed that a tackifier was needed to retain the granule HDPE in the web during the web formation process. Preliminary testing had also indicated that the tackifier, a wax emulsion of oxidized low molecular weight polyethylene (E-10), did not have an adverse effect on the properties of the resultant test panels. The tackifier was applied to the wood fibers in a rotating drum blender With an air spray gun in a manner similar to the application of phenolic resin in the AL Series 1 studies.

Equipment Modifications and Additions

A 305-mm-wide, laboratory-scale Rando-Webber forming machine was used to make nonwoven mats for the air-laid composites, The equipment was modified to minimize the density gradient across the web, a problem encountered in preliminary experiments.

Panel Fabrication

Nonwoven webs were weighed, sorted, and stacked on the basis of weight and specific gravity. A steam-heated platen press was used to press the panels to a thickness of 3.2 mm and a specific gravity of 1.0. A cooling cycle was used to maintain target thickness.

The recyclability part of the study required that we determine the feasibility of recycling panels made for AL Series 2, which contained DF and RHDPE. We found

that we needed to recycle the boards through the pressurized refiner to be able to produce the desired fiber length and bundles.

Tests on Mechanical and Physical Properties

We evaluated the performance of panels made for AL Series 1 (PET systems), AL Series 2 (HDPE systems), and recyclability.

Results of Tests on PET Systems (AL Series 1)

In general, the mechanical, water resistance, and dimensional stability properties of panels made from recycled materials were equivalent to similar properties obtained from panels containing all virgin or virgin/recycled materials, Therefore, the recycled ingredients tested in AL Series 1 could replace virgin materials with minor consequences.

Mechanical Properties

Panels made with the HF/VPET formulation had the highest bending MOR value (50.6 MPa), although no statistically significant differences were observed for MOR values for either wood fiber or PET variations. The modulus of elasticity (MOE) values followed a different pattern. In both the HF and DF groups, the MOE values of boards containing RPET were significantly higher than those of boards containing VPET; total average increase for this property was 16% for both groups.

For the HF/VPET formulation, tensile strength was 33.0 MPa; tensile strength decreased by 14% for the HF/RPET formulation. However, when RPET or VPET fibers were used with DF fibers, no significant differences were noted. In contrast to tenisile strength, the incorporation of RPET fibers increased tensile modulus (MOE) by 6% and 7% for HF and DF formulations, respectively, although these differences were not statistically significant.

Impact energy of specimens from the HF and OF formulations showed a consistent trend. Impact strength was respectively 20% and 10% higher for HF and DF formulations containing VPET fibers compared to formulations containing RPET fibers.

Table 1. Results of water soak and linear expansion tests for AL series[a]

Composite[b]	24-h water soak		Linear expansion (%)		
	Thickness swell (%)	Water absorption (%)	30% RH	65% RH	90% RH
AL Series 1					
HF-80% VPET-10% PR-10%	25.2 (9)	43.4 (20)	0.19 (10)	0.42 (4)	0.61 (6)
HF-80% RPET-10% PR-10%	22.3 (9)	41.3 (25)	0.21 (13)	0.44 (8)	0.70 (7)
DF-80% VPET-10% PR-10%	29.8 (14)	48.2 (25)	0.20 (11)	0.43 (6)	0.64 (7)
DF-80% RPET-10% PR-10%	26.9 (8)	44.1 (16)	0.20 (12)	0.45 (6)	0.71 (6)
AL Series 2					
HF-60% VHDPE-30% VPET-5% E10 wax-5%	43.8 (14)	54.9 (17)	0.15 (9)	0.39 (5)	0.68 (7)
HF-60% RHDPE-30% VPET-5% E10 wax-5%	42.7 (21)	61.8 (19)	0.17 (12)	0.42 (12)	0.69 (12)
DF-60% VHDPE-30% VPET-5% E10 wax-5%	45.2 (13)	58.7 (13)	0.17 (5)	0.40 (7)	0.64 (9)
DF-60% RHDPE-30% VPET-5% E10 wax-5%	52.8 (15)	65.8 (16)	0.16 (7)	0.44 (6)	0.74 (6)

[a]Values in parentheses are coefficients of variation (%).
[b]DF is demolition wood fiber; E10, epolene-maleated polyethylene; HF, hemlock fiber; VPET, virgin polyester fiber; RPET, recycled polyester; RHDPE, recycled high-density polyethylene; and VHDPE, virgin high-density polyethylene.

Physical and Dimensional Stability Properties

Thickness swell values were significantly different for HF/RPET and DF/VPET specimens (Table 1). The HF/RPET specimens had the lowest thickness swell value (22.3%), and the DF/VPET specimens the highest value (29.8%). Water absorption values were not significantly different.

Linear expansion values at 30% RH were statistically equivalent (Table 1). At 65% and 90% RH, the HF/RPET and DF/RPET formulations had slightly higher values and were statistically different from the HF/VPET and DF/VPET formulations.

Results of Tests on HDPE Systems (AL Series 2)

Panels containing virgin and recycled wood fiber/polyethylene had equivalent mechanical and physical properties. Therefore, as in AL Series 1, the recycled materials used in AL Series 2 could replace virgin materials with minor consequences.

Mechanical Properties

The DF/VHDPE panels had the highest bending MOR value at 19.1 MPa, followed by 18.7 MPa for the HF/RHDPE panels. Generally no statistically significant differences were observed for MOR values for either wood fiber or HOPE variations. In contrast, the HF/RHDPE panels had the highest bending MOE value (2.13 GPa) and the DF/VHDPE panels the lowest MOE value (1.75 GPa); however, these results were not statistically significant.

For tensile strength, the highest value (12.4 MPa) was observed for the DF/VHDPE formulation; tensile strength of the DF/RHDPE panels was 7% lower (11.5 MPa). For both wood fiber variations, the use of either virgin or recycled HOPE did not significantly influence tensile strength values. The HF/VHDPE panels had the highest tensile MOE (2.81 GPa); incorporation of RHDPE lowered tensile MOE by 21% (2.23 GPa), a significant change. Tensile MOE values of DF formulations were about equal, averaging 2.11 GPa.

Type of wood fiber and formulation did not significantly affect impact energy.

Physical and Dimensional Stability Properties

Thickness swelling of DF specimens was an average of 22% higher than that of HF specimens; the highest value (53%) was observed for the DF/RHDPE formulation (Table 1). Particularly notable is the fact that RHDPE had a significant effect on only the DF formulation and not the HF formulation. Thickness swelling was lowest in the HF/RHDPE panels (43%).

The formulation had a consistent influence on water absorption (Table 1). Incorporating RHDPE with either type of wood fiber produced a statistically significant

increase in this property (average 13% increase). The HF/VHDPE and DF/VHDPE formulations showed the lowest water absorption values.

Linear expansion values for all formulations at 30% RH ranged from 0.15 to 0.17% (Table 1). At 65% and 90% RH, the HF/RHDPE and DF/RHDPE formulations had slightly higher values.

Results of Tests on Recyclability

In general, the mechanical, water resistance, and dimensional stability properties of second-generation panels made from recycled materials were essentially equivalent to or better than properties obtained from first-generation panels. Therefore, the second-generation composites, or possibly higher generation composites, can be produced using recycled materials without the consequence of reduced property values.

Mechanical Properties

The MOR of second-generation panels was higher than that of first-generation panels (19.6 MPa compared to 17.4 MPa). On the other hand, the bending MOE values of first-generation panels were higher than that of second-generation panels (2.01 vs. 1.77 GPa). These differences were not statistically significant. The tensile strength of second-generation panels was 19% higher than that of first-generation panels. Similar results were obtained for tensile modulus. The higher properties of the second-generation panels indicate that wood fiber/RHDPE composites can benefit by the addition of refiberized material from first-generation panels.

Impact energy values of specimens made from first-generation panels and second-generation panels were nearly equal and not statistically different.

Physical and Dimensional Stability Properties

In the 24-h water-soak tests, first-generation panels showed 53% thickness swell (Table 2). Incorporating first-generation panel fibers into second-generation panels improved this property by 21%. Similar trends were observed for water absorption values (Table 2). Water absorption of second-generation panels was 18% lower than that of first-generation specimens. These differences were statistically significant. The results suggest that the additional HDPE from refiberized first-generation panels further encapsulated the wood fibers, thus limiting water uptake by the wood fibers.

Linear expansion values were similar for both formulations at 30% RH, although the differences were statistically significant (Table 2). The incorporation of recycled panels into second-generation panels significantly decreased linear expansion at both 65% and 90% RH.

The positive influence of incorporating 20% first-generation panels into the second-generation panels may be the result of several factors. The incorporation of 20% refiberized first-generation panels reduced the percentage of wood fiber (DF) from 60% to 40%. The actual amount of total wood fiber was reduced from 60% to 52%, and the total amount of RHDPE was increased from 30% to 36%. Likewise, the percentage of PET and E-10 each was increased by 1%. The increase of these components, particularly the HDPE, and the decrease of the wood fiber may be a direct cause for some improvements in property values. More wood fiber was able to be encapsulated by plastic, thereby reducing exposure of the wood to moisture.

Table 2. Results of water soak and linear expansion tests on recyclability specimens[a]

Composite	24-h water soak		Linear expansion (%)		
	Thickness swell (%)	Water absorption (%)	30% RH	65% RH	90% RH
First-generation panels[b]	52.8 (15)	65.8 (16)	0.16 (7)	0.44 (6)	0.74 (6)
Second-generation panels[c]	42.0 (12)	54.3 (12)	0.15 (11)	0.37 (9)	0.52 (11)

[a]Values in parentheses are coefficients of variation (%).
[b]DF-60%, RHDPE-30%, VPET-5%, E10 wax-5%. DF is demolition wood fiber; RHDPE, recycled high-density polyethylene; VPET, virgin polyester fiber; and E10, epolene-maleated polyethylene.
[c]DF-40%, recycled panel-20%, RHDPE-30%, VPET-5%, and E10 wax-5%

Conclusions

- The Rando-Webber air-forming equipment can be adapted to handle both long and short synthetic and natural fibers as well as powder. Nonwoven air-laid webs can be produced that have excellent uniformity in both the machine- and cross-machine directions.

- Recycled and granulated HDPE can be used in the FPL air-forming equipment to produce an air-laid web that can be subsequently made into flat panels or shaped sections.

- Pressure refining techniques can convert postconsumer demolition wood or construction waste into fiber bundles that can be processed very successfully in the FPL air-forming equipment and subsequently pressed into flat panels or shaped sections.

- Panels made with recycled materials compare favorably to those made of virgin materials. Mechanical and physical properties of panels made with recycled polyester fiber or high-density polyethylene and demolition waste wood are similar to those of panels made with virgin materials.

- Second-generation composites, or possibly higher generation composites, can be produced using recycled materials without the consequence of reduced property values. Mechanical and physical properties of second-generation panels made from recycled materials were essentially equivalent to or better than properties obtained from first-generation panels.

STUDIES ON MELT-BLENDED COMPOSITES

Melt-blending is an inherently low cost, high production rate process in which wood and/or paper are mixed with molten plastic. These blends can then be formed into products using conventional plastics processing techniques such as extrusion and injection molding. The plastic acts as a means to convey the wood/paper during processing and the wood/paper fiber bears the load in the final composite, offering an effective balance between processability and strength of end product. With melt-blending techniques, wood fiber provides several advantages as reinforcement in thermoplastic composites. These include economy on a cost per unit volume basis, desirable aspect ratios, flexibility (hence less fiber breakage), and low abrasiveness to equipment. Composites can be produced containing up to 50 weight percent wood fiber and are low cost, thermoformable, and relatively insensitive to moisture.

Methods and Materials

In laboratory investigations of melt-blended composites, experimental operations generally proceed through the following sequence of steps: (a) conversion of raw materials into forms suitable for preparing dry mixtures quantitatively and feeding those mixtures into the melt-blending apparatus, (b) quantitative dry mixing, (c) melt-blending by either an extruder or a K-mixer, (d) injection molding of test specimens, and (e) measurement of properties.

Five major studies were undertaken to investigate the effects of a number of variables on mechanical and physical behavior of wood-fiber-reinforced thermoplastics. The studies were statistically designed and analyzed and all comparisons were made at a 95% confidence level. However, for the sake of brevity, the results will be presented as a whole, centering on the larger effects of the variables because of their greater impact on composite performance. Cellulosic fibers, plastics, and coupling agents are descfibed in Table 3.

Table 3. Materials used in melt-blending studies

Material	Abbreviation	Description[a]	Source
Plastic			
Virgin polypropylene	VPP	Fortilene 9101, 1602, 1902; nominal MFI 3, 12, 30.	Solvay Polymers, Inc., Deer Park, TX
Recycled polypropylene from auto battery cases	BPP	Cleaned chips; nominal MFI 10	Gopher Smelting and Refining Co., Eagon, MN
Recycled polypropylene from ketchup bottles	KPP	Cleaned chips; nominal MFI 3	Wheaton Plastic Recycling Co., Milleville, NJ
Recycled polypropylene from milk bottles	HDPE–MB	Cleaned chips; nominal MFI 0.7	Recycle Worlds, Madison, WI
Cellulosic filler			
Wood flour	WF	Western pine; -40+80 mesh	American Woodfiber Co., Schofield, WI
Cellulose	BW40	Pure cellulose fiber; mean length 60 µm	James River Corp., Hackensack, NJ
Waste newspaper	ONP	Over-production issue	Milwaukee Journal/ Sentinel Inc., Milwaukee, WI
Waste magazine	OMG	Representative sample of Madison waste stream	Madison Recycling Center, Madison, WI
Coupling agent			
Epolene E43 powder	E43S	Powdered maleated polypropylene; M = 4,200	Eastman Chemical Co., Kingsport, TN
Epolene E43 emulsion	E43E	Emulsified potassium salt	Eastman Chemical Co.
Epolene G3002 powder	G3002	Powdered maleated polypropylene; M = 11,000	Eastman Chemical Co.

[a]MFI is melt flow index. M is molecular weight.

Plastics

Plastics act as the matrix in the composite. In this program, a baseline matrix plastic, virgin polypropylene (VPP) homopolymer, was compared to three waste plastics: polypropylene from auto battery cases (BPP), recycled ketchup bottle polypropylene (KPP), and recycled high-density polyethylene from blow-molded milk **bottles** (HDPE-MB). These plastics were chosen on the basis of low melting temperature (necessary for use with wood/paper fiber), cost, performance, and availability.

Cellulosic Fibers

Celluloses act as the reinforcing filler in the composite. We used wood flour (WF) as the primary baseline filler because it is currently used commercially with polypropylene in extruded sheets for automobile interior panels. We included relatively expensive (several times the price of WF) pure cellulose fibers (BW40) as another baseline filler for comparing against recycled fibers—waste newspaper (ONP) and old magazines (OMG).

Both plastics and additives can be readily used in traditional plastics processing machinery. However, wood or paper fibers present difficulties during the melt-blending step. Cellulosic filler must be in a form that can be completely dispersed into the molten plastic by the shear forces exerted during melt-blending. Wood flour was readily disaggregated and fully dispersed into individual particles in the plastic using a simple laboratory single-screw extruder. Although difficult to disperse with an extruder, a usable blend of BW40 cellulose fibers was also obtained in this way. Wastepaper fibers were much more difficult to handle because of their low bulk density. For melt-blending in a high intensity K-mixer, the paper was milled or ground into approximately 4- to 8-mm flakes using a small granulator. For melt-blending in an extruder, the paper was first reduced into fibers by a small modified hammermill.

Coupling Agents

Additives aid the dispersion of filler into the matrix plastic and/or enhance the bonding (act as a “coupling agent”) between filler and matrix. In this program, we restricted ourselves to Eastman Epolenes (E43, G3002)—maleated polypropylene “waxes” in which the small degree of maleation provides polar groups capable of bonding to the cellulose while the polypropylene segments, in theory, offer compatibility with the polypropylene matrix.

Mechanical and Physical Properties

Materials were compounded in either a 38-mm single-screw extruder or a 1-L K-mixer. Extrusion parameters were temperature profile 170°C to 190°C, screw speed 15 rpm, and residence time 1 to 2 min. The K-mixer parameters were 4500 to 5500 rpm and discharge temperature 171°C to 200°C. In all studies, test specimens were prepared by injection molding. At least five specimens of each blend were tested for each mechanical property. After molding, the specimens were stored over dessicant for at least 3 days before testing.

Mechanical properties were measured on the dry specimens at approximately 23°C. Specimens and test methods followed ASTM D256, D638, D747, and D790. Apparent melt viscosities and shear rates were calculated from measured volumetric throughput rates and pressure drops across the die during steady-state extrusion in a single-screw extruder at 190°C.

Selected properties from the various studies are summarized in Table 4. This table shows the larger effects of some more-sensitive variables. Because the conditions of each study varied somewhat, care must be taken when comparing data. General trends rather than actual values will be emphasized in the following discussion.

Reinforcement Effects

Addition of 30%-40% of any of the wood/paper reinforcing fillers to the plastics resulted in a composite with higher modulus and strength but lower impact energy and percentage of elongation and energy to maximum load. These effects are not surprising and are typical of reinforced thermoplastics in general. Table 5 summarizes property changes with addition of 40% ONP to several virgin and recycled polypropylenes.

Most major changes in composite performance occurred at filler contents below 30%. For example, aside from an approximate 10% increase in modulus, no major changes in mechanical performance were found over the rather narrow range of 32% to 42% ONP in VPP.

Table 4. Selected mechanical property data from various studies

Plastic	Filler	Coupling agent	Tensile modulus (GPa)	Tensile strength (MPa)	Izod impact energy Notched (J/m)	Izod impact energy Unnotched (J/m)
PPV	None	None	—	31.5	23.8	650
	40% ONP	E43	4.89	47.1	20.8	109
	40% BW40	E43	4.80	48.2	24.7	114
	40% WF	E43	3.72	34.1	18.7	72
HDPE–MB	40% ONP	E43	3.80	37.6	28.6	73
	40% BW40	E43	3.79	36.6	30.8	68
	40% WF	E43	2.61	27.8	36.4	81
PPV	40% ONP	None	4.97	34.0	20.8	113
	40% ONP	E43	4.90	47.4	19.8	144
	40% ONP	G3002	4.56	52.3	20.4	190
KPP	None	None	1.62	36.5	62.0	>800
	40% ONP	G3002	4.03	52.3	30.6	167
	40% OMG	G3002	3.55	38.9	34.2	138
BPP	None	None	1.32	24.5	165.0	>800
	40% ONP	G3002	3.98	42.5	34.3	150
	40% OMG	G3002	3.44	31.8	41.8	125

[a]Coupling agent was added at a level of 3%–5% of fiber weight depending on the study.

Typically, the more fibrous fillers (ONP and BW40) resulted in composites with superior mechanical properties but were more difficult to process when compared to WF. However, the presence of clays and other impurities in the OMG made the material more difficult to disperse and interfered with bonding, resulting in decreased mechanical performance when compared with other fibrous fillers.

Figure 1 demonstrates some effects on viscosity at low shear rates. Viscosity measurements showed dramatic increases when WF was replaced with ONP and as fiber content was increased. This increase in viscosity demonstrated the effects of both aspect ratio and filler contents on composite melt behavior. Even at a fiber loading of 32% ONP, PPV composite melts were significantly more viscous than a 42% WF-filled PPV,

Table 5. Change in property after addition of 40% ONP to plastic

	Filled/Unfilled		
Property	KPP	BPP	VPP
Tensile			
Modulus	2.49	3.00	—
Strength	1.49	1.66	1.66
Energy	0.65	0.73	—
Elongation	0.47	0.46	—
Flexural			
Modulus	2.29	3.01	2.94
Strength	1.52	1.91	2.00
Impact			
Notched	0.49	0.21	0.86
Unnotched	<0.21	<0.19	0.29

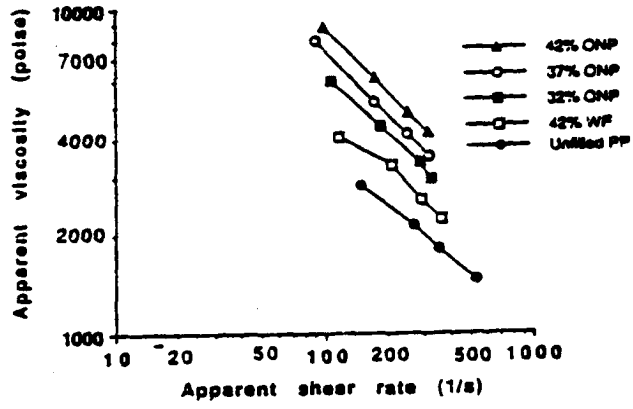

Figure 1. Apparent melt viscosity of composite blends.

Matrix Effects

Changing the melt flow index (MFI) of VPP had a significant effect on viscosity but little effect on mechanical performance over the range studied (3-30 g/10 min). This result suggests that MFI could be used to compensate for increased viscosity of the higher performance fibrous composites.

The polypropylene composites were stronger and stiffer than the HDPE-MB composites but had lower notched impact energy values. The unfilled plastic composites showed the same trend; i.e., composite properties qualitatively followed those of the matrix polymers. Similar trends were also seen for composites made with BPP and KPP. For example, the use of BPP (a block copolymer with higher notched impact strength) instead of VPP resulted in composites with higher notched impact strength, although the difference in this property was not nearly as great as that in the unfilled polymers. In other words, although choice of plastic can affect composite properties, addition of reinforcement can affect the mechanisms by which the plastic achieves its performance.

Analysis of the results of both the filler and plastic effects can lead to some interesting conclusions. For example, the mechanical properties of a recycled ONP/HDPE-MB system were at least as good as those of the current commercial WF/polypropylene system. This may offer some practical utility if problems associated with the high viscosity of the recycled system can be overcome.

Coupling Agent Effects

Coupling agents may be incorporated into composites in different ways to concentrate the material at the interface where it is active. In our studies, adding solid coupling agent (E43) to the melt or applying emulsified E43 directly to the fiber had little effect on mechanical performance of composites. Concentrating the emulsified additive at the fiber/matrix interface by precoating the fibers was apparently counteracted by the much lower chemical reactivity of the potassium salt with the cellulose compared with the reactivity of the anhydride with cellulose. Perhaps the E43 functions more to enhance dispersion of the cellulosic fillers than to bond the cellulose to the polymer.

The higher molecular weight maleated polypropylene, G3002, had a very beneficial effect on composite strength, energy absorption, and unnotched impact behavior. Initial moduli were approximately the same but ultimate strength and area under the curves (a measure of toughness) were much greater for systems with coupling agent (especially G3002). The fact that unnotched rather than notched impact energy was increased indicates that G3002 probably enhances dispersion of the fibers and reduces aggregates that would act as failure loci. For example, unnotched impact energy was more affected by removal of failure loci than was notched impact energy because the notch itself constitutes a failure locus. The apparently greater interaction of G3002 with polypropylene relative to E43 may be attributable to the higher molecular weight of G3002.

Processing Effects

Little preference was shown for compounding in a K-mixer or a single-screw extruder with WF or cellulose fiber. However, those fillers are relatively easily dispersed, and the ONP could not be melt-blended with a single screw. Moreover, the small interaction terms between filler and compounder in the statistical analysis also indicated some differences in dispersibility between WF and cellulose fiber.

We investigated the effects of re-extrusion on the mechanical and theological performance of three different composite blends. For almost all mechanical and theological properties, very little change occurred as the material was recycled over five cycles. The length and therefore the aspect ratio of the ONP fiber decreased, but these decreases were apparently not great enough to result in any large reductions in composite performance. The WF was reduced in both thickness and length, as smaller bundles of wood fibers were sheared off the larger particles. These changes in

dimension resulted in no overall change in aspect ratio and, ultimately, in composite performance.

Conclusions

- Melt-blended composites cannot be prepared with wastepapers as reinforcing filler using a conventional laboratory single-screw extruder. However, these composites can be prepared with a laboratory high-intensity K-mixer, an industrial-scale K-mixer, or a twin-screw extruder that employs a properly designed feeder for the fiber.

- Old newspaper (ONP) as reinforcing filler provides better properties than wood flour, which is currently used as filler in commercial composites. Old magazines (OMG) can also be used as a filler, but they are less easily dispersed into the matrix plastic and result in somewhat lower properties than those of composites containing ONP.

- With the same filler, substituting recycled milk bottle polyethylene for VPP leads to lower strength, stiffness, and unnotched impact energy, but higher notched impact energy.

- Use of recycled high density polyethylene from milk bottles and recycled polypropylene from battery cases as a matrix in composites with ONP results in improved impact performance when compared with that of composites made from VPP and ONP.

- Addition of the coupling agent Epolene G3002 at 3 weight percent of filler results in very useful increases in composite properties, probably as a result of improved fiber dispersion.

- Select composite systems showed little or no loss in mechanical properties when repeatedly reprocessed (re-extruded and injection molded).

COMMERCIAL IMPLEMENTATION

The research program led to many cooperative studies with industry and academia, all with the ultimate goal of commercializing composites made with waste materials.

Commercial Feasibility of Waste Newspaper-Thermoplastic Composites

The program was partially funded by the Forest Products Laboratory (FPL) and the Wisconsin Department of Natural Resources (DNR) and by in-kind contributions from the eight cooperating companies. Laboratory experiments demonstrated that old newspapers could be dispersed as fibers into thermoplastics by melt-blending,

resulting in substantial improvements in some properties compared with the unfilled plastic or plastic filled with wood flour. Major conclusions are as follows:

- Old newspaper/polypropylene (ONP/PP) composites can be compounded on a commercial scale using either the K-mixer with ONP flakes as feed or using a twin-screw extruder with ONP fibers fed separately from the plastic.
- An ONP/PP sheet containing 42 weight percent ONP can be prepared by extrusion on a commercial scale. This sheet meets existing specifications for automobile panels and can be thermoformed into a variety of shapes.
- Given proper design of melt processing equipment, a wide variety of other commercial products could be manufactured from ONP/PP composites with similar ONP content.
- Firm estimates of production costs for ONP/PP composite products must await
  - additional examination of compounding methods to define optimum balance of dispersion ability, throughput rate, and cost
  - improvement in methods to deliver wastepaper in a form and at a cost acceptable to a compounder or a manufacturer of plastic products

Waste LDPE Program

A consortium of companies is investigating the use of waste LDPE "contaminated" by residual fiber from a hydropulping operation that scavenges wood fiber from coated paper stock. The program involves raw material processors, compounders, plastics processors, and research institutions and is being coordinated by the FPL. Major hurdles in this program are the residual moisture in the raw material from the hydropulping process and product applications.

Waste Jute-Polyester Panels As Reinforcing Filler

In response to interest expressed by a U.S. company in the possibility of recycling panels produced by impregnating jute fibers with thermosetting polyester, we granulated the panels and investigated the ability to use the resultant mixture as reinforcing filler in melt-blended composites with a polypropylene matrix. Overall, this waste material produced composite mechanical properties approximately equivalent to those of similar composites containing wood flour as the reinforcement.

Waste Kenaf Core As Reinforcing Filler

This program resulted from a request by the Agrecol Corporation (Madison, Wisconsin) to determine whether kenaf core material could be useful as a reinforcing filler in plastic composites. We granulated the core material and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were approximately equivalent to those of similar composites containing wood flour. Therefore, where kenaf core is readily available at low cost, it could very likely substitute for wood flour as a reinforcing filler.

Wastewood Composite As Reinforcing Filler

The University of Tennessee extension requested an evaluation of waste wood composite as a reinforcing filler in thermoplastic composites. Although such solid waste is available in large quantity; it contains wood with cured thermoset adhesives that might cause problems in melt-processed composites because the adhesives do not melt at processing temperatures. We granulated the plywood and successfully melt-blended the -40 mesh fraction with polypropylene. The composite properties were approximately equivalent to those of similar composites containing wood flour. This waste material could therefore substitute for wood flour as a reinforcing filler in melt-processed composites.

RECOMMENDATIONS

Research

Additional research is needed on both air-laid and melt-blended composites made from recycled wood fiber and plastics to improve properties and processing and to thereby increase potential applications.

1. Evaluate the potential for making composite materials with other major components of the waste stream, including low-density polyethylene, polystyrene, and mixed waste plastics.

2. Verify the recyclability of composites made with reground first-generation ingredients.

3. Improve melt-blending processes to achieve better fiber dispersion with minimal fiber breakage.

4. Improve bonding between wood fiber and plastic matrix to enhance physical and mechanical properties.

5. Improve impact energy and creep resistance (decreased deflection under long-term load), currently the limiting properties of these composite systems.

6. Determine the resistance of these composite systems to relatively extreme environments and develop means to enhance resistance to moisture, biodegradation, and fire.

Commercialization

1. Continue extensive outreach to industry to acquaint companies with these types of composite systems, to develop applications, and to cooperate in product development.

2. To obtain commercial acceptance of melt-blended composites containing wastepaper fiber,

   a. improve methods for converting wastepaper at costs acceptable to industrial users; costs must at least approach current cost of wood flour (about $0.22/kg)

   b. improve methods for melt-blending fiber and plastics on a commercial scale **at** costs acceptable to melt fabricators (extruders, injection molders, etc.)