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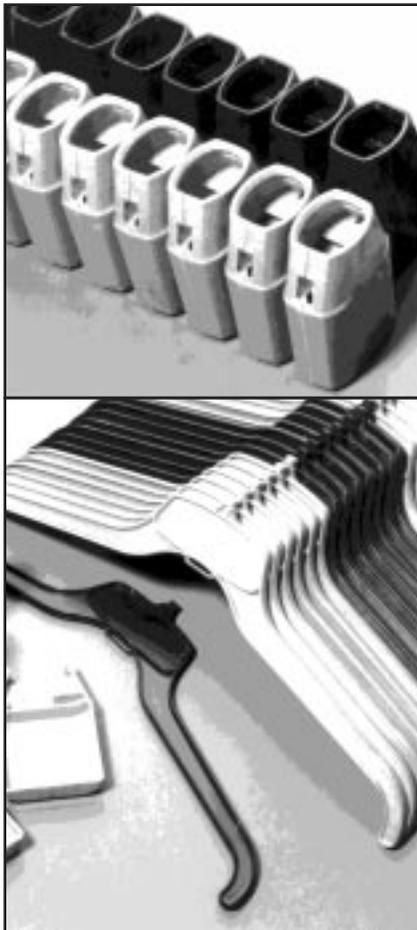
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Waste-Wood-Derived Fillers for Plastics

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Abstract

Filled thermoplastic composites are stiffer, stronger, and more dimensionally stable than their unfilled counterparts. Such thermoplastics are usually provided to the end-user as a precompounded, pelletized feedstock. Typical reinforcing fillers are inorganic materials like talc or fiberglass, but materials derived from waste wood, such as wood flour and recycled paper fiber, are also effective as fillers. The goal of this project was to generate commercial interest in using waste-wood–paper-derived fillers (WPFs) to reinforce thermoplastics. The research strategy was twofold: developmental research and outreach. Specific objectives were (1) to improve wastepaper fiber preparation, feeding, and compounding methods, and optimize composite performance, and (2) to communicate to end-product manufacturers the advantages of WPF thermoplastics.

The research was led and supported by the Forest Products Laboratory (FPL), with input from a consortium of 15 fiber suppliers and plastics manufacturers. Additional funding was provided by the Wisconsin Department of Natural Resources. Equipment was leased and installed at FPL. Eight general purpose formulations were developed—they included extrusion and injection molding grades of both polyethylene and polypropylene, reinforced with WPFs.

An information packet containing performance data, appropriate processing conditions, sample pellets, sample parts, and a questionnaire was sent to nearly 500 commercial

plastics manufacturers in Wisconsin, Illinois, and Michigan. In response to requests for in-house trials, FPL researchers conducted nearly 18 site visits. The researchers ensured proper handling of the material, provided consultation, and gathered information about processing and performance. The trials went very well, and parts were successfully manufactured at all facilities. Products included automobile trim components and housings, vacuum cleaner parts, paint brush handles, bicycle parts, cosmetic cases, and other household items. Great interest has been shown in the use of WPF thermoplastics; one consortium member is establishing a 4 million kg/yr (9 million lb/yr) facility. Total market demand is conservatively expected to exceed 45 million kg/yr (100 million lb/yr).

Keywords: wood fiber, plastic processing, properties of composites, recycling

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Waste-Wood-Derived Fillers for Plastics

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Introduction

This report represents the culmination of Project 94–55 for the Solid Waste Reduction and Recycling Demonstration Grant Program of the Wisconsin Department of Natural Resources. The project was conducted from July 1, 1994 through February 29, 1996.

Background

Previous research at the Forest Products Laboratory (FPL) (Myers and Clemons 1993) and elsewhere had demonstrated the benefits of using waste-wood–paper-derived fillers (WPFs) in thermoplastics. For various reasons, however, the industrial thermoplastic composite industry had been reluctant to accept this technology. The manufacture of thermoplastic composites is often a two-step process: compounding or blending of the raw materials, and formation of the composite blends into a product. Compounders were reluctant to produce thermoplastic blends with WPFs because they were not sure about how to handle the material and were not aware of a market to justify production of the blends. Product manufacturers, on the other hand, did not have access to a supply of compounded pellets for producing the end product and often were not aware of performance advantages and processing limitations of the material. Moreover, few successful demonstrations of this technology had been performed on conventional commercial-scale equipment.

In response to this situation, scientists at the FPL developed a unique program that would overcome some hurdles preventing commercial acceptance of technology for using WPFs in thermoplastics. This program was completed with the cooperation of many industrial partners selected for their particular skills, interests, and abilities.

Project Goals

The overall goal was to generate sufficient commercial interest in WPF thermoplastics to allow large-scale commercial activity. As outlined in Figure 1, specific objectives were as follows:

1. Conduct research, development, and engineering efforts to
 - a. improve methods of preparing wastepaper fiber of needed quality, fiber length, and cost
 - b. select and develop compounding methods to optimize feeding, fiber length retention, and dispersion in the plastic
2. Communicate to end-product manufacturers the cost savings and product properties derived from using WPFs in plastic products

Selection of Materials

Given the large number of processing technologies associated with thermoplastic composites, it was necessary to narrow the focus to the materials and processes with the best chance of success in light of the program objectives. For end-product manufacturing, both injection molding and extrusion technologies were targeted. These are two of the four largest technologies for the production of plastic and thermoplastic composites. The others—blow molding and rotational molding—are not appropriate for the materials used in this program. The study materials are described in general terms in this section; detailed information is provided in the section on composite performance.

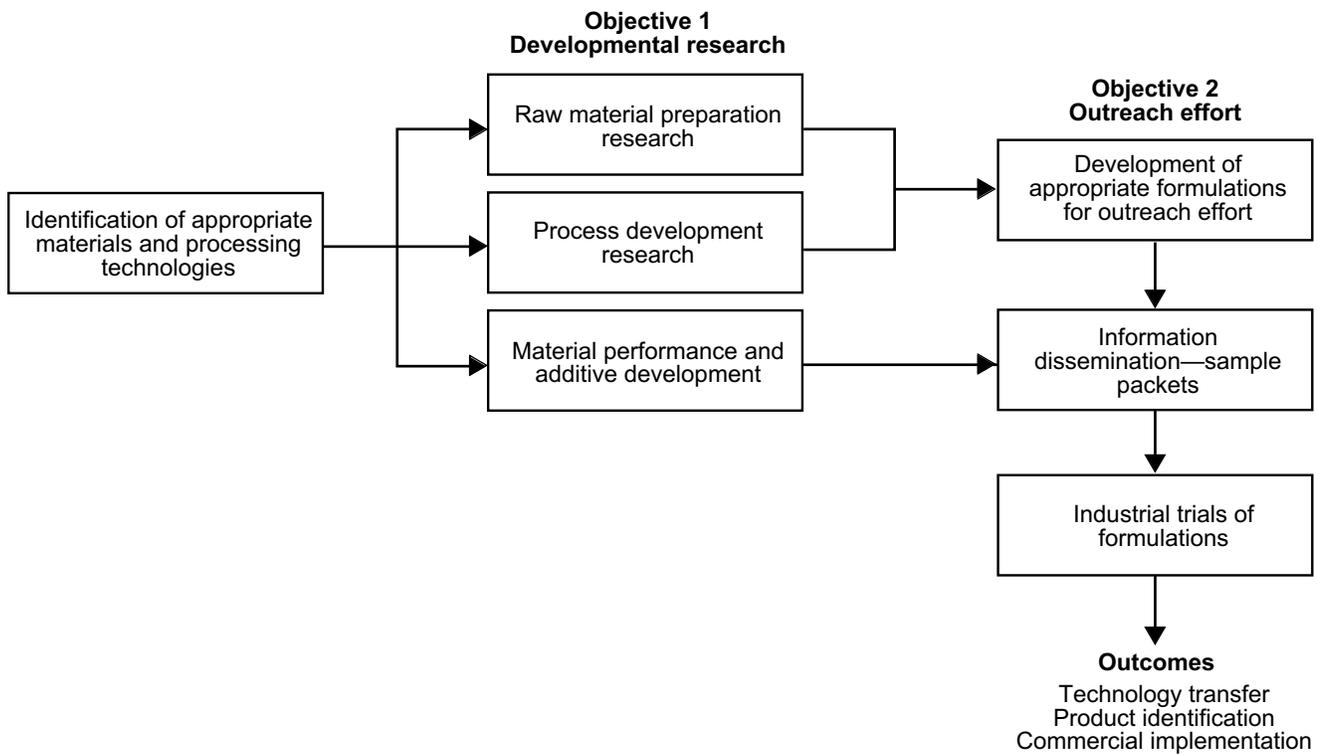


Figure 1—Research strategy.

Wood-Based Fibers

The waste wood and paper fibers had previously been identified for their potential as reinforcing fillers in thermoplastics. Wood flour is an economical, commercially available filler that has been used in thermoplastic composites to a limited extent. Fiber from old newspapers (ONP), another relatively inexpensive filler, has demonstrated improved performance as a reinforcing filler compared to wood flour because of its higher aspect ratio. We initially chose ONP fiber because it has a high percentage of high-yield mechanical pulps and hence short, stiff fibers. If these fibers acted as good reinforcing fibers, then other fibers with higher percentages of chemical pulps and much longer fiber would be expected to perform even better. In addition, ONP fiber was chosen because its fiber quality is more uniform than that of mixed wastepapers.

Plastics

Polypropylene and high-density polyethylene (HDPE) were chosen as the matrix polymers. They are both widely used, are available at low cost, and have good performance for the intended applications. Their low melting points also allow processing below the degradation temperature of wood and paper.

A wide variety of polypropylene and HDPE polymers are available, and careful selection is important because extrusion

and injection molding require different material characteristics. For example, a critical need in injection molding is good flow of the material into the mold, whereas a critical need in extrusion is melt strength to enable handling of the hot material as it comes from the die. Choice of polymers for these technologies is therefore quite different. Injection molding requires a polymer with a low molecular weight to maintain low viscosity. By contrast, extrusion requires a polymer with a higher molecular weight for better melt strength.

Additives

To lower raw material costs and thus the cost of the end products, additives such as impact modifiers and compatibilizers were not added to the formulations used for the outreach portion of the project. However, ways to tailor performance using additives are described in the section on composite performance. The cost increase incurred using additives may be justified when performance specifications for end products are identified.

Processing

Research on processes included methods for feeding WPFs, configuration of compounding equipment, management of moisture, and determination of overall processing conditions. Care was taken to maintain quality of fillers while maximizing filler dispersion, distribution, and extruder throughput.

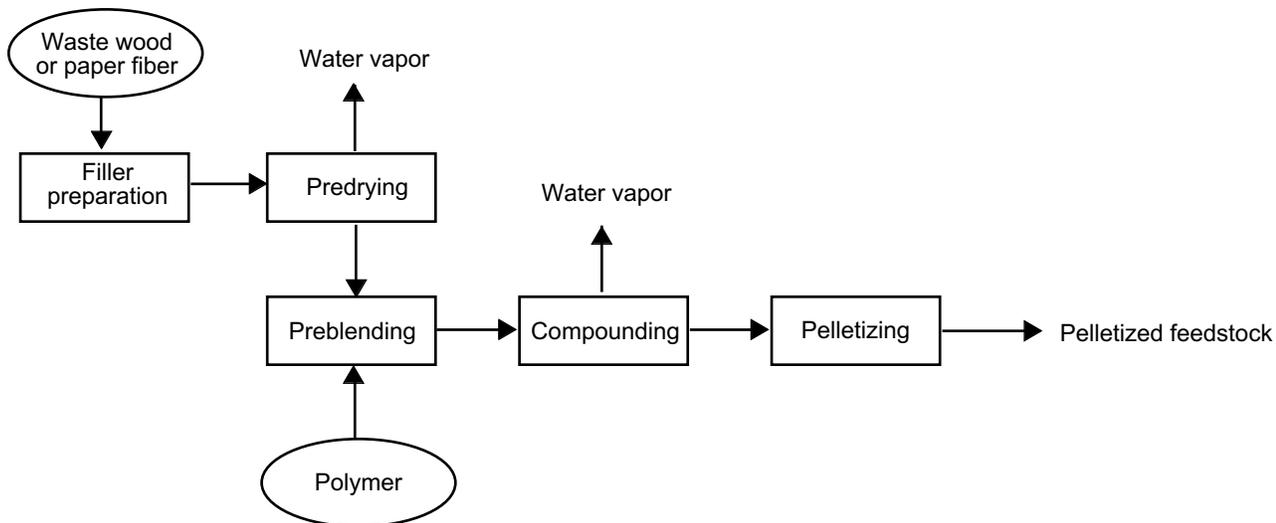


Figure 2—FPL compounding line.

The FPL compounding line is shown schematically in Figure 2.

Preparation and Feeding of Fillers

Conventional plastics equipment is designed to handle materials with a bulk density of approximately 500 kg/m^3 (31 lb/ft^3). Although somewhat lower bulk densities can be handled, material with very low density is difficult to feed, requires specialized equipment, and can reduce processing rates. Because WPFs have lower bulk densities than do thermoplastics, their preparation can be a critical step in the compounding process.

No research on raw material preparation was necessary for wood flour. This material is commercially available and has a bulk density of around $112\text{--}240 \text{ kg/m}^3$ ($7\text{--}15 \text{ lb/ft}^3$). Although lower in bulk density than thermoplastics, wood flour is sufficiently dense to be readily fed and dispersed. The feeding and dispersion of ONP fiber, however, required considerable investigation. Three different forms of ONP fiber were investigated: hammermilled newspaper fiber, crumble pulp, and Szego-milled fiber.

Hammermilled fiber is currently the least expensive dry form of newspaper fiber, and considerable fiber length is maintained during the milling process. This fiber has an extremely low bulk density ($16\text{--}32 \text{ kg/m}^3$ ($1\text{--}2 \text{ lb/ft}^3$)) and is available through a number of insulation manufacturers. Crumble pulp is made by densifying and briquetting damp hammermilled paper and then crumbling the resulting dried briquettes into coarse paper pellets. It has a bulk density of around 192 kg/m^3 (12 lb/ft^3) and is available commercially in the form of bagged absorbent products. Szego-milled paper is paper that has been cut into platelets in a Szego mill. The platelets that we used were about 40 mesh in size, had a

consistency and texture not unlike graphite, and had a bulk density of around 224 kg/m^3 (14 lb/ft^3). This material is currently available only on special order.

Although hammermilled paper is inexpensive and composites made from it perform well, the low bulk density makes it difficult to feed without specialized crammer-type feeders. Budgetary constraints limited us to noncrammer-type feeders. We used both an Acrison (Moonachie, NJ) Model 75-E volumetric feeder and an AccuRate (Whitewater, WI) Model 8000 loss-in-weight feeder. Both of these feeders were very effective for processing wood flour, crumble pulp, and Szego-milled paper.

Unfortunately, the high density of the individual paper pellets that formed the crumble did not readily break apart and disperse in the plastic. Research is being conducted on a crumble pulp compacted to a lower initial density and on a crumble pulp containing waxes as a dispersion aid; significant progress is being made in this area. Because Szego-milled fiber could be fed and dispersed easily and because composites made with it performed at least as well as those made with wood flour, the Szego-milled fiber was chosen for the bulk of the research.

Compounding Equipment

The development of compounding technology, evaluation of feeding of raw materials, and manipulation of formulations required the construction of a small-scale industrial compounding line on site. A search was initiated for a compounding line that used conventional technology and was cost-effective and flexible. Based on the results of this search and previous experience with different compounders, cooperator input, budgetary considerations, and plant trials, we decided that a twin-screw extruder would be most beneficial to the program.



Figure 3—Compounding and pelletizing equipment at FPL.

A leasing agreement was arranged with the Davis Standard Corporation (Pawcutuck, CT), and a twin-screw extruder manufactured by them was installed at FPL (Fig. 3). The extruder has 32-mm (1.26-in.) co-rotating, intermeshing segmented screws with a length-to-diameter ratio of 32:1. There are eight electrically heated, water-cooled barrel sections, two of which have vents for removing volatile materials. Power is supplied by a 15-hp DC drive, and a four-hole strand die is fitted to the discharge end. The machine's capacity is 45 kg (100 lb) of unfilled polypropylene per hour.

To cool the compounded strands discharged by the extruder, FPL staff constructed a waterslide cooling trough similar to that manufactured by Conair Jetro (Franklin, PA). After cooling, the strands were fed into an older model Cumberland (Providence, RI) pelletizer for cutting into pellets.

Moisture Management

Processes for manufacturing plastics tolerate little or no water. Removal of moisture is critical because any moisture remaining in the WPF–plastic blend turns to steam and manifests itself in the form of foam. This can disrupt processes and lead to unacceptable finished parts.

During compounding, moisture was managed by a combination of predrying the fibers from their ambient moisture content of 6–8 percent to 2–3 percent; vacuum was then applied to the vent zones in the extruder barrel during compounding to remove the remaining moisture. Properly done, pelletized feedstock with a moisture content of < 0.1 percent could be manufactured. At that level, the pellets were ready for injection molding or extrusion in unvented conventional systems. Using hot, predried wood and paper fiber also tended to increase throughput.

Processing Conditions

The compounding of WPF with thermoplastics is also limited by the thermal degradation temperature of the wood or paper fiber. Typically, melt temperatures (temperature of molten material) were kept below 204°C (400°F). Above this limit, signs of degradation (smoke, odor, discoloration) were readily apparent with ONP thermoplastics. Strand quality from the extruder rapidly decreased with attempts to raise this limit by as little as 1° or 2°.

Wood-flour-filled strands were somewhat more forgiving. At 204°C (400°F), some discoloration was apparent, indicating some degradation, but strand quality was still sufficient for pelletization. At around 210°C (410°F), smoke and excessive odor were apparent, and strand quality began to rapidly deteriorate.

In general, we found that polyethylene-based formulations could be successfully compounded at 182°C (360°F) or less, whereas polypropylene-based formulations seemed to work well at around 193°C (380°F). These temperatures were typically used regardless of the type of WPF selected.

Several variables could be adjusted to keep melt temperatures at these levels. First, as mentioned previously, one reason for selecting polyethylene and polypropylene was their low melt temperatures and their ability to be effective in these temperature ranges. Two other variables were similarly connected: the intensity and speed (r/min) of the screws.

In general terms, the more mixing elements in the screw configuration, the more intense the mixing action of the fibers into the plastic matrix. This extra mixing increases the mechanical work imparted to the material, thus increasing melt temperature. Screw speed has a similar effect: the higher the rotations/minute, the more mechanical energy imparted, and thus the higher melt temperatures.

We found that a fairly low number of mixing elements was sufficient for satisfactory compounding with wood flour and Szego-milled ONP fiber. Screw speed could also be kept reasonably moderate, at around 240 r/min. Under these conditions, compounded material was routinely produced at 75–100 percent of rated machine capacity.

Composite Performance

Performance of Standard Blends

In support of the objectives of the outreach portion of the project, eight “standard” blends were compounded (Table 1). Melt flow indices are summarized in Table 2. These blends were formulated with ease of processing by the end user as the primary criterion. Since it was likely that many different

Table 1—Mechanical properties of standard blends^a

Blend	Tensile strength ^b (MPa)	Tensile modulus (GPa)	Tensile elongation (%)	Flexural strength ^b (MPa)	Flexural modulus (GPa)	Notched Izod (J/m)	Unnotched Izod (J/m)
PP-WF-X	29.7 [0.3]	4.10 [0.23]	2.4 [0.1]	58.6 [0.4]	4.06 [0.13]	20.8 [0.91]	105 [11]
PE-WF-X	19.7 [0.3]	2.69 [0.09]	2.9 [0.2]	35.8 [0.7]	2.43 [0.13]	26.7 [2.9]	66 [6]
PP-WF-1	27.0 [0.5]	2.92 [0.14]	3.2 [0.2]	51.9 [0.3]	3.07 [0.06]	16.2 [0.63]	109 [9]
PE-WF-I	18.7 [0.21]	2.43 [0.09]	2.5 [0.2]	32.4 [0.5]	2.13 [0.14]	17.8 [0.45]	52 [4]
PP-SZ-X	29.2 [0.31]	4.34 [0.21]	2.2 [0.1]	57.2 [0.5]	4.24 [0.10]	19.1 [0.91]	103 [7]
PE-SZ-X	23.4 [0.3]	3.27 [0.11]	2.2 [0.1]	40.2 [0.4]	2.88 [0.15]	22.2 [0.74]	65 [3]
PP-SZ-1	26.3 [0.06]	3.28 [0.09]	2.6 [0.1]	51.1 [0.3]	3.41 [0.11]	14.2 [0.67]	89 [7]
PE-SZ-1	18.7 [0.2]	2.21 [0.1]	2.6 [0.1]	32.4 [0.5]	2.13 [0.13]	14.7 [0.78]	53 [3]

^aBracketed numbers are standard deviations. PP is polypropylene; PE, high-density polyethylene; WF, wood flour; SZ, Szego-milled newspaper; X, extrusion grade; and I, injection molding grade.

^bMaximum values.

Table 2—Melt flow indices (MFIs)^a

Blend	Filler (%)	Filler type	Polymer MFI at 230°C	Polymer MFI at 190°C	Composite MFI at 190°C
PP-WF-X	40	WF	4	2	0.4
PE-WF-X	40	WF	—	4	0.7
PP-WF-I	30	WF	35	18	4.2
PE-WF-I	30	WF	—	44	8.8
PP-SZ-X	40	Sz	4	2	0.3
PE-SZ-X	40	Sz	—	4	0.3
PP-SZ-I	30	Sz	35	18	2.0
PE-SZ-I	30	Sz	—	44	5.4

^aSee footnote to Table 1 for definitions of terms. Unit of measurement for MFIs is grams/10 min. $t_F = 1.8 t_C + 32$.

products with different performance requirements would be molded or extruded, we recognized at the outset that the standard blends would not represent an optimal formulation. Optimization of formulations for a given product is an iterative process involving the specific product, product manufacturer, and compounder, and, as such, lay outside the scope of this project. The work on standard formulations promised to fulfill the requirements of the outreach portion of the project in that such formulations would allow processors some initial hands-on experience with the processing of this class of materials and a rough idea of performance.

Polymers

The polypropylenes for the extrusion and injection molding grades were Fortilene 9200 and Fortilene 3907 homopolymers, melt flow 4 and 36.5 g/10 min, respectively (Solvay Polymers, Inc., Deer Park, TX). The high-density polyethylenes for extrusion and injection molding grades were LS 6402-00 and LS 3420-00 polyethylene copolymers, melt flow 4.2 and 44 g/10 min, respectively (Quantum Chemical Corporation, Cincinnati, OH).

Fillers

The wood flour was a standard 80-mesh pine (#8020) from American Wood Fibers (Schofield, WI). When appropriate, a standard 40-mesh pine (#4020) was also used. The ONP fiber selected for the outreach portion of the project was Szego-milled paper (American Wood Fibers) because of our early success with handling and processing this material. The newspaper was milled at General Communion, Inc. (Toronto, Ontario) to a -40 mesh.

The materials were premixed and compounded in a twin-screw extruder as described in the section on processing. ASTM standard test specimens for mechanical testing were molded at 190°C (374°F) in a 33-t reciprocating screw injection molder (Cincinnati Milacron, Batavia, OH). Izod impact, flexural, and tensile properties were then measured according to ASTM D 256, D 790, and D 638, respectively (ASTM 1990a-c). As a rough measure of viscosity, the melt flow indices (MFIs) of the blends were measured at 190°C (374°F) and 2.16 kg (4.76 lb) plunger weight.

Melt Flow Indices

The addition of fillers/reinforcements to thermoplastics can greatly reduce the flow properties of a polymer. This reduction in flow properties becomes especially important in highly filled blends. To provide processors with blends with appropriate flow properties for the processes, targets for MFIs for the injection molding and extrusion grades were identified.

Targets for MFIs of the composite formulations were 4-10 g/10 min for injection molding grades and fractional

(< 1 g/10 min) for extrusion grades. As a general rule, WPF-thermoplastic blends are kept below 204°C (400°F) during processing to prevent degradation. Because of the low processing temperatures, MFI was measured at 190°C (37°F). This is particularly important to consider with the polypropylene blends since the MFI of polypropylene is usually measured at 230°C (446°F). This difference in temperature has a significant effect on the MFI.

These temperature and filler effects must be taken into account when choosing an appropriate polymer for a particular application. Consequently, high MFI (low viscosity) base polymers were chosen as a starting point for the injection molding grades. The MFIs of the base polymers and standard blends are shown in Table 2. All of the targeted MFIs were obtained except for the injection molding grade of polypropylene that contained ONP, which was a little low. The HDPE blends had higher MFIs than the polypropylene blends, which was not surprising considering that the base resins had lower MFIs when measured at 190 °C (374 °F). Wood-flour-filled blends had higher MFIs than did blends containing ONP fibers.

Mechanical Properties

The mechanical property data are summarized in Table 3. For most properties, the polypropylene blends performed better than the HDPE blends, undoubtedly because of the relative performance of the base polymers. Extrusion-grade blends performed better than injection molding grades. This was not surprising considering that the extrusion grades contained higher molecular weight polymers with better mechanical performance. Few differences were seen between the wood flour and Szego-milled blends. These results contradict those of previous studies in which ONP fibers performed better than wood flour as a reinforcement in polypropylene (Myers and others 1992, Gonzales and others 1992). This reduction in ONP fiber performance as a reinforcement can be attributed to the reduction of fiber length in the Szego mill. Work on ONP preparation methods is in progress.

Comparison of Standard and Commercial Blends

Although a large body of literature is available on the properties of thermoplastics filled with minerals, no direct comparisons could be found to WPF thermoplastics. The project study plan did not include the comparison of standard and commercial blends. The comparison of various reported data is inconclusive because the polymers selected for study significantly affect performance. Comparison of the extrusion-grade and injection molding polypropylenes will show how properties can vary in apparently similar homopolymers from the same manufacturer. (See section on future work.)

Table 3—Mechanical properties of additives^a

Blend	Additive type	Additive amount (%)	Tensile strength ^b (MPa)	Tensile modulus (GPa)	Tensile elongation (%)	Flexural strength ^b (MPa)	Flexural modulus (GPa)	Notched Izod (J/m)	Unnotched Izod (J/m)
PP-WF-X	—	—	26.6 [0.1]	3.93 [0.26]	2.4 [0.1]	50.7 [1.0]	3.86 [0.11]	19.3 [4.7]	84 [12]
	Nucleating agent	0.15	27.4 [0.4]	4.02 [0.07]	2.2 [0.2]	51.0 [0.6]	3.79 [0.11]	18.8 [1.4]	94 [7]
	EPDM1	5.5	28.2 [0.4]	3.53 [0.11]	2.8 [0.1]	52.4 [0.6]	3.20 [0.08]	22.5 [0.4]	123 [14]
	EPDM2	5.5	25.0 [0.5]	3.62 [0.24]	2.5 [0.1]	47.1 [0.8]	3.54 [0.09]	21.6 [0.3]	96 [8]
	MAPP	1.8	31.7 [0.2]	4.17 [0.34]	2.3 [0.2]	59.3 [1.0]	3.86 [0.10]	16.9 [0.9]	95 [6]
PP-SZ-X	—	—	31.1 [0.1]	5.41 [0.58]	1.6 [0.1]	55.4 [1.3]	4.36 [0.11]	17.6 [0.8]	89 [7]
	Nucleating agent	0.15	32.9 [0.2]	4.96 [0.35]	2.4 [0.1]	58.4 [0.5]	3.65 [0.02]	16.7 [0.4]	109 [16]
	EPDM1	5.5	32.8 [0.2]	4.27 [0.47]	3.4 [0.1]	57.8 [1.2]	2.94 [0.05]	27.1 [2.0]	163 [24]
	EPDM2	5.5	33.7 [0.5]	4.48 [0.12]	2.8 [0.1]	59.6 [1.6]	3.26 [0.15]	22.3 [0.6]	142 [16]
	MAPP	1.8	42.0 [0.2]	5.59 [0.7]	2.6 [0.1]	72.6 [2.2]	4.12 [0.20]	16.8 [0.6]	142 [2]

^aAdditives were added as part of polymer content. Bracketed numbers are standard deviations.

^bStrength at yield.

Study of Additives

A brief study on additives was undertaken to demonstrate how some common additives can be used to tailor mechanical properties of composite blends. The purpose of this investigation was not to recommend an optimized formulation but to demonstrate how manipulation of the formulation can lead to better balances of properties. Specific parts with specific mechanical requirements would have to be identified to justify the use of these additives; otherwise, even small additional costs would be prohibitive. The additives and their level of addition are somewhat arbitrary without a mechanical property target; they were chosen at supplier-recommended levels or were based on previous experience, with a concern for cost. The extrusion grade of polypropylene was used for all blends (Fortilene 9200, Solvay Polymers, Deer Park, TX). The additives used in the investigation were a coupling agent, impact modifiers, and a nucleating agent.

Coupling Agent

Hydrophilic WPFs are not chemically compatible with the hydrophobic polypropylene polymers. To improve bonding between the two components, a maleated (MA) poly-

propylene was added as a coupling agent (MP 880, Aristech Chemical Corporation, Pittsburgh, PA). In previous investigations (Sanadi and others 1994), the addition of a similar MA polypropylene G-3002 (Eastman Chemical Products, Inc., Kingsport, TN), which has a relatively high molecular weight and acid number, to these types of composites markedly improved performance.

Impact Modifiers

Elastomers or rubbers are often added to filled and unfilled thermoplastics to improve impact performance. At low levels, these modifiers often form a separate phase in the polymer matrix. Applied stresses can be transferred to the softer elastomeric phase rather than accumulate in unfavorable locations, which may lead to failure. Ethylene-propylene-diene copolymers (EPDMs) are commonly added to polypropylenes as impact modifiers. Several EPDMs have also been chemically modified to improve compatibility of the WPF with polypropylene. Two EPDMs were used in this study—Fusabond 227D and Fusabond 280D, both from Dupont Canada, Inc. (Mississauga, Ontario); they are referred to as EPDM1 and EPDM2, respectively.

Nucleating Agent

Nucleating agents can be added to a formulation to affect the crystal growth of the polymer during cooling. By affecting the crystal structure, these nucleating agents can improve such properties as transparency and stiffness. The effect of Millad 3988 (Milliken Chemical Company, Inc., Spartanburg, SC) on mechanical properties was investigated.

Summary of Additives Study

The materials were blended in a 1-L (1.06-quart) thermokinetic mixer (K mixer, Synergistics Industries Inc., St. Remi de Napierville, Quebec). The thermokinetic mixer, a high-intensity batch mixer, was used because of its ability to handle formulation changes quickly and to accurately control the small additive concentrations. The blends were then injection molded and tested in the same manner as were the standard blends.

Table 3 summarizes the results of the study on additives; Table 4 shows the increases in property values of blends with additives compared to baseline blends. The nucleating agent had very little effect, if any, on the mechanical properties of the composite blends, despite its reported effectiveness in unfilled polypropylenes (Gächter and Müller 1990). One possible explanation is that the WPFs themselves act as nucleating sites for the polypropylene and, therefore, addition of a nucleating agent does not have a great effect on crystalline development. The effects of cellulose fibers on crystallization of polypropylene were reported by Quillen and others (1993).

The EPDM effects on mechanical properties of the WPF-filled polypropylenes were typical of elastomer-modified composite blends. The tradeoff between increases in impact performance and decreases in moduli with addition of elastomer has been well documented. Impact performance of these ternary composites is affected by many factors, including weight fraction of the components, EPDM type (e.g., EPDMs with different ethylene/propylene ratios, chemically modified EPDMs), processing parameters (e.g., intensity), and polymer matrix properties (e.g., viscosity, compatibility with fillers or EPDM). Manipulation of these variables will depend on considerations related to finished parts, processes, and cost.

The effects of addition of MA polypropylene on composite performance were similar to those found in previous studies (Sanadi and others 1994). Improved bonding of the WPF component resulted in improved tensile/flexural and unnotched Izod impact strengths. Better transfer of applied stresses because of better bonding allows higher stresses to be reached (higher strength properties) and makes it more difficult for cracks to be initiated at stress concentrations such as fiber ends (higher unnotched impact strength). Since the initial modulus of the composites is a result of the moduli of

Table 4—Effects of various additives on mechanical properties of 4-MFI polypropylene with 40% filler

Additive	Change (%) compared to base blend ^a			
	Tensile strength	Flexural modulus	Notched Izod	Unnotched Izod
Nucleating agent, 0.15%	LTP	LTP	LTP	18
EPDM1, 5.5%	LTP	-25	35	65
EPDM2, 5.5%	LTP	-17	20	37
MAPP, 1.8%	27	LTP	LTP	37

^aAverage value of both fillers. LTP refers to <10% property change.

the components, not the bonding between them, flexural and tensile moduli are not affected.

Outreach

An information packet (Fig. 4) containing general information on the material, performance data, appropriate processing conditions, sample pellets, sample extruded and injection molded parts, and a questionnaire was sent to nearly 500 commercial plastic product manufacturers in Wisconsin, Illinois, and Michigan. Response rate to the questionnaire exceeded 16 percent; half the respondents requested materials for in-plant evaluation.

The outreach effort was part of the informational activities prescribed by the Wisconsin Department of Natural Resources for this project. Other informational activities have included the wide distribution of this report to the general public as well as presentations at conferences and technical workshops, the first for the “Progress in Wood Fibre–Plastics Conference” in Toronto, Canada, on April 29, 1996.

Manufacturers who had submitted favorable responses to the questionnaire were contacted for trials at their manufacturing facility. A total of 18 site visits were conducted; scientists from FPL ensured proper handling of material and gathered information from the manufacturers on processability, performance, and potential end-uses.

The FPL supplied between 20 and 200 kg (44 and 440 lb) of pelletized material for each trial (Fig. 5). Material was always redried before the trial to remove any moisture absorbed by the pellets. All the trials were conducted at injection molding facilities; a straight 193°C (380°F) temperature profile was used for polypropylene formulations and a straight 182°C (360°F) temperature profile for HDPE formulations.



Figure 4—WOOD-COM marketing and information package.



Figure 5—Examples of pelletized thermoplastic feedstock (left to right): virgin unfilled feedstock, feedstock reinforced with waste wood fiber, feedstock reinforced with waste newspaper (ONP) fiber.

Mold temperature varied between 15°C and 55°C (60°F and 130°F). Injection pressures and times were usually reduced for the wood-flour-filled formulations and occasionally reduced for the ONP-filled formulations. All other conditions were also well within normal operating range.

All manufacturers were impressed with the moldability of the formulations, and parts were successfully made at all trials. Depending upon the type of processing, the part being made, and the material currently being used, the use of WPF thermoplastics resulted in shorter cycle times, superior performance or appearance, or environmental benefits.

A producer of cosmetics cases liked the natural appearance of cases molded with wood-filled polypropylene. A manufacturer of home repair tools thought the recycled content afforded by WPF would give his products a market edge in the “green” products section of some major home repair supply stores. Several manufacturers stated that the reduced cycle times will significantly increase profits and make them more competitive. Automotive suppliers envisioned the WPF-thermoplastics as being able to replace higher cost resins

Table 5—Estimated capital costs of 4 million kg/yr (9 million lb/yr) wastepaper fiber-reinforced thermo-plastic compounding facility

Item	Cost
2 acre lot, 6,000 ft ² building, office furnishings ^a	\$ 240,000
Handling and drying of WPF	150,000
Compounding system (extruder, feeders, pelletizer)	600,000
Handling and packaging of finished materials	80,000
Total	\$1,070,000

^a1 acre = 4.046 × 10³ m². 1 ft² = 0.0929 m².

without the associated weight gain of mineral-filled thermoplastics. A manufacturer of paint brush and roller handles thought that wood-flour-filled plastic handles resembled solid wood enough to replace it in their line of products.

Photographs of many manufactured items are included in the Appendix. The positive attributes of the products and concerns raised during the industrial trials are described in the following section.

Costs, Benefits, and Concerns

Successful adoption of WPF thermoplastics by the conventional plastics industry will depend upon costs and real and/or perceived benefits. Several resolvable concerns also need to be addressed.

Capital Costs

A 4 million kg/yr (9 million lb/yr) facility has been suggested as the minimum size for a successful commercial compounding venture. Such a facility would employ 13 persons: 8 production workers, 1 production supervisor, 1 engineer, 1 salesperson, 1 clerical worker, and 1 general manager. Total sales would be approximately \$4–\$5 million, based on the assumptions described in this section. At 45 million kg/yr (100 million lb/yr), total employment would probably approach 100, with total sales of \$50–\$60 million. The estimated capital costs of a 4 million kg/yr (9 million lb/yr) facility are shown in Table 5.

Additional employment and economic activity would be created in the existing wastepaper recovery and processing infrastructure, and through product changes.

Material Costs

To fully discuss the cost benefits of WPF thermoplastics, several assumptions will need to be made. For this discussion, we will assume a cost of \$0.50/lb for thermoplastic polymer and \$0.10/lb for prepared (hammermilled) wastepaper fiber.¹ We will also assume that compounding can be profitably conducted at \$0.20/lb at maximum throughput. This figure is derived from the capital cost, operating cost, assumed 80-percent operating time, and requisite return on investment. Based on these assumptions, the cost per pound of precompounded pelletized feedstock can be determined using the following formula:

$$\$/\text{lb} = \frac{p(0.50) + f(0.10) + 0.20}{e}$$

where

- p = polymer weight (percent)
- f = filler weight (percent)
- e = throughput efficiency at assumed 80-percent operating time

As an example, a 50/50 formulation compounded at 100 percent efficiency would have a price of

$$\$/\text{lb} = \frac{(0.50)(0.50) + (0.50)(0.10) + 0.20}{1.00} = 0.50$$

This scenario was selected as an example because the cost of the compounded feedstock equals that of the virgin resin. As the formula indicates, increasing the filler content decreases cost; either increasing the polymer content or decreasing the efficiency increases cost.

In general, most manufacturers contacted during the industrial trials thought that the costs were reasonable. Most manufacturers lowered their operating temperature when molding the materials, resulting in significant energy savings. Some manufacturers did not routinely process filled materials, while others did. Because filled and unfilled thermoplastics have different purposes and uses, comparisons to WPF thermoplastics are discussed separately in the following sections.

Comparison to Unfilled Polymers

One obvious justification for using WPFs is improved performance. From a product standpoint, the improvements may allow the user to manufacture products in a higher performance category, to reduce overall material use through better engineering, and to increase the life of the product. Some users contacted in the technology transfer activity also

anticipated replacing higher priced, higher performing resins like ABS, which currently costs about \$1.00/lb.

A less obvious advantage, but one with great significance, is reduced cycle time. During the technology transfer stage of the research, most users of unfilled thermoplastic were able to reduce product cycle times, some in excess of 25 percent. The resultant savings far outweighed a 5- to 10-percent cost premium. Cycles can be reduced because WPF can be used to displace a considerable volume of polymer, thereby reducing the amount of polymer to be chilled to solid form. In addition, and perhaps more important, the WPF helps to dimensionally stabilize the part at elevated temperatures, allowing it to be removed from the mold at a higher temperature without fear of distortion.

Comparison to Filled Polymers

One of the greatest benefits of using WPFs as opposed to inorganic materials is weight savings. Fiberglass has a specific gravity of 2.5; talc or calcium carbonate, around 2.8. When used as a reinforcing filler, WPF has a maximum specific gravity of about 1.4. Equal volume loadings will result in a molded part that weighs less, which means the user will be able to buy less material to mold the same part (Fig. 6). This is particularly important in large volume applications where even minute weight reductions in parts provide huge cumulative savings.

The lower specific gravity of WPFs also means that on a weight basis, they will displace roughly twice the volume of polymer as will an inorganic filler (Fig. 7). Therefore, higher loadings are possible without weight gain. These savings in weight also allow many wastepaper fiber formulations to be made with a higher specific stiffness than are formulations using minerals; that is, they are stiffer on a weight comparison basis.

For some users contacted in the technology transfer activities, cycle time was reduced compared to the time required for filled resins, particularly polypropylene filled with talc and calcium carbonate. This was probably due to the higher polymer displacement afforded by the WPF formulations. Anecdotal information also indicated that WPF is less abrasive to processing equipment than are inorganic fillers, particularly fiberglass.

Concerns

The greatest concern raised by manufacturers involved the rate of moisture uptake by the dried feedstock. As mentioned previously, for most applications >0.1 percent moisture content resulted in foaming. Articles made of WPF plastic have an equilibrium moisture content of 1 to 2 weight percent. Pelletized feedstock is no exception, which means that

¹ 1 lb = 0.454 kg.

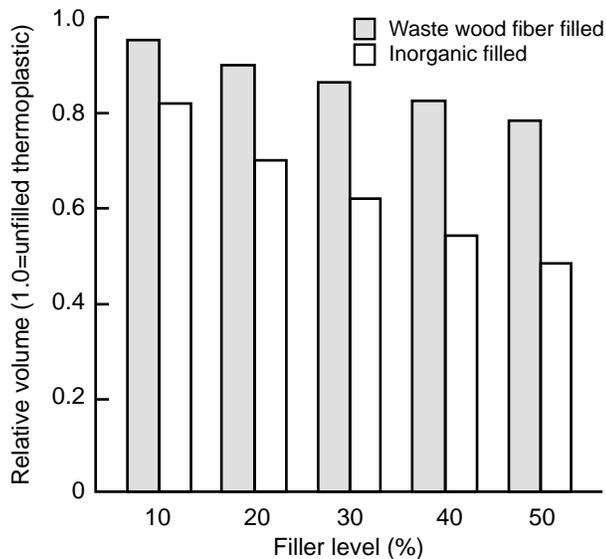


Figure 6—Relative volumes of thermoplastic composites reinforced with inorganic fillers compared to WPF-reinforced composites.

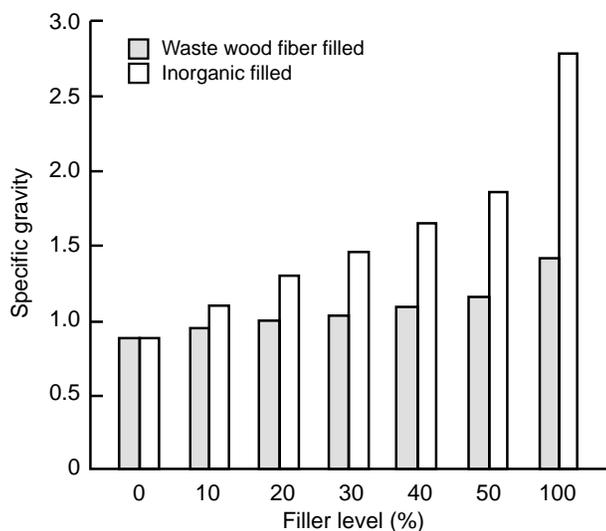


Figure 7—Specific gravity of thermoplastic composites reinforced with inorganic fillers compared to WPR-reinforced composites.

dried feedstock will have a working life before it needs to be redried.

The length of the working life depends on several factors, including relative humidity, filler content, and storage techniques. To better determine the working life of the feedstock, several formulations were oven dried and placed in various temperature- and humidity-controlled rooms at FPL. Rates of moisture uptake are shown in Figure 8.

Another concern was shrinkage rates. Plastic shrinks as it solidifies; in response, molds are made oversize by a

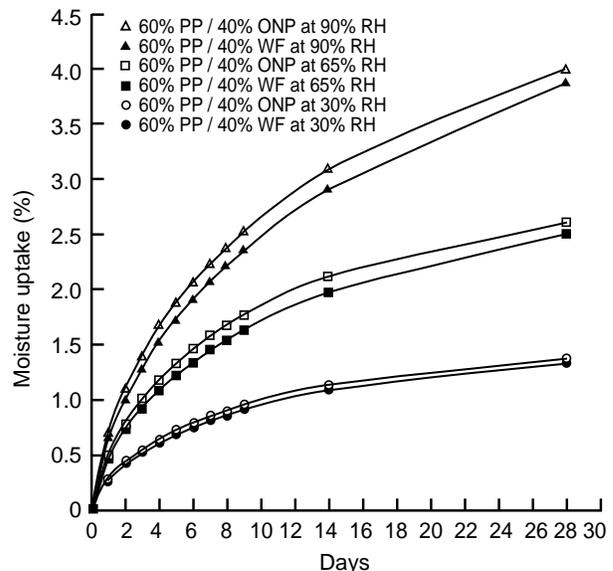


Figure 8—Moisture uptake of ovedried polypropylene (PP) pellets reinforced with wood flour (WF) or newspaper (ONP) fiber at various humidities.

prescribed amount. The thermal coefficient of expansion of most plastics is 250 to 400 times greater than that of wood. Therefore, plastics filled with WPF do not shrink as much as their unfilled counterparts, and they are thus somewhat larger than those made in the same molds from unfilled material. For parts that are not components of an assembly, this is not much of a problem. For parts that are components of an assembly, either the mold or the mating parts may need to be modified. The problem may not occur with molding equipment designed for filled materials because shrinkage rates are already reduced. Filler content can also be manipulated to give equivalent shrinkage rates. Of course, physical and mechanical properties will change correspondingly.

Product Applications

A list of all applications for this class of materials is beyond the scope of this report. The following description of two product areas may provide some indications of usage.

Automotive Applications

Recycling is a high priority research area for the automotive industry. The motivation for this is twofold: one from customer demand, the other from existing and anticipated government mandates. Thermoplastics reinforced with postconsumer wastepaper fibers are very attractive for these reasons. However, the adoption of WPF thermoplastics for automotive applications may rest on cost and performance. The automotive industry uses much ABS; for nonimpact sensitive applications, filled polypropylenes can offer performance similar to that of ABS for reduced cost. Parts made

using WPF-filled polypropylene also weigh less than those made with mineral fillers, and weight savings economize fuel expenditure.

Pallets and Other Shipping Applications

Another potential high volume use of WPF thermoplastics is for pallets and other related shipping containers. Plastic pallets and other returnable plastic containers have already made significant in-roads to this huge market, which has traditionally been dominated by wood pallets and containers. As an example, the Postal Service purchases 2.5 million plastic pallets each year. For many applications, however, plastic pallets do not perform as well as wood pallets. Typically, plastic pallets suffer from a lack of stiffness. Performance is enhanced by using thick plastic components or fiber-glass/metal reinforcement. The added weight contributes to both the purchase price and the shipping costs, which are incurred every time the product is used. Using WPF as a reinforcing filler can significantly improve performance without significantly adding weight.

Environmental Impact

Reasons for using WPF thermoplastics extend beyond performance and cost advantages to far-reaching environmental impact.

The technology described here provides a high volume outlet for both postindustrial and postconsumer waste wood and paper fibers. From the paper perspective, the fibers do not require the cleaning methods needed for paper-to-paper recycling. No sludge is produced, no waste water needs to be treated, and there is no need for deinking. Although most of our research has focused on old newspapers, many other grades of paper, like mixed office waste and bulk mailings, could also be used.

Because of the thermoplastic component, the composite material itself is recyclable, and previous research has shown that recycling can be accomplished with little loss in performance (Youngquist and others 1993). Recycled plastics can also be used in these systems, diverting this valuable material from landfills.

The products made from these materials will often have long life cycles. Life expectancy for most automobiles exceeds 10 years, and automobile recycling technology is among the most advanced. Plastic pallet manufacturers also recycle their products. In fact, the buy-back of damaged plastic pallets for raw materials is one of their major selling points.

Recycling is not a fad, nor is the use of WPF as a reinforcing filler in thermoplastics. The WPF thermoplastics will help make automobiles lighter and more fuel-efficient, increase the shipping efficiency of a wide class of goods, and reduce the

demand on landfills. These materials use a renewable resource to extend the life of a nonrenewable one, and in so doing, retain their recyclability.

Conclusions

The overall goal of the project has been met. At the time of this writing, one cooperator is undertaking tasks to establish a commercial facility of the scale described in this report. Customers identified during the outreach phase of the project will form the basis for this commercial endeavor.

Both study objectives have been met. Quality feedstocks can be produced using the guidelines described in this report. The plastics industry is receptive to pelletized feedstocks reinforced with fillers derived from waste wood and paper fiber. Users of unfilled plastics have found the feedstocks attractive for their combination of high performance and low cost, as well as reduced molding cycle times. Users of filled resins have found similar advantages, as well as weight savings. No advantages on a strict cost-only basis have been identified.

The formulations developed for the outreach effort were general purpose ones. While all manufacturers who tried the formulations were able to mold parts successfully, each application would benefit from a more tailor-made formulation. Research presented in this document shows how performance can be tailored, and continued work by the FPL will address that issue and others related to material science.

Future Work

Future work concerns both old and new problems. An old problem remains: feeding low-bulk density, fiberized waste-paper into an extruder, while making good use of the fibers present. Ongoing work at FPL is directed toward examining medium-density crumble pulp with a preapplied wax dispersant. Researchers will also examine several grades of Szegomilled paper to determine which size yields the best performance.

New problems include development of formulations for specific end-uses. This work will include various recycled fiber types, virgin and recycled plastics, homopolymers and copolymers, and additives. A formal study has also been initiated to make a direct comparison of WPF and mineral fillers. This new project will use the injection molding grade polypropylene used in the work reported here. The materials will be compared on the basis of equal weights and volumes of filler.

Acknowledgments

The success of a large project like this does not happen without the efforts of many people. We thank the following industrial cooperators (in alphabetical order): Chris Anderson and Mike Ford, Badger USA, for development of the mailing list, help with the marketing packet, and assistance during plant trials; Mark Berger and Tom Forcey, American Wood Fibers, for prompt deliveries of wood flour and Szego-milled paper as well as technical assistance; Mark Billian and Mike Dahl, Eaglebrook Products, for allowing us to conduct trials and for loan of the Cumberland pelletizer; Jim Giatras, Bemis Manufacturing, for allowing us to conduct plant trials and for valuable insight about the possibilities of wood fiber-plastic composites; Kevin Gohr, Sheboygan Substrates, for consultation, valuable contacts, and loan of the Acrison feeder; Bernie Kiernan and Mark Lindenfelder, Davis Standard Corporation, for lease of the extruder, excellent startup service, and continuing technical advice; Mike Killough, Solvay Polymers, for generous donations of polypropylene, guidance, and advice; Paul Koch, Penn State-Erie, for sample production and insight; Dennis Kopcha, Penn Pro Manufacturing, for backup supply of crumble pulp; John Kraus, Lamico, for being an early supporter and helping with applications; Terry Laver, Strandex, for allowing us to conduct trials and for technical assistance; Milt Risgaard and his entire staff at Teel Plastics, for continuing support, assistance, and willingness to try new things; George Tedder, Conair Reclaim Technologies, for arranging trials; and Jim Van Hulle, Quantum Chemical Co., for timely donations of HDPE, advice, and guidance.

A complete listing of all the FPL staff who contributed to the project would be quite lengthy, but we would like to single out several people. In no particular order, we would like to thank Jerry Saeman and George Myers (both retired) for their pioneering promotion and research that led to this project; John Youngquist for excellent facilitation; John Bachhuber and Pat Brumm for negotiating contracts and shortening red tape; and finally, Gary Lichtenberg and his entire maintenance staff for prompt and excellent service.

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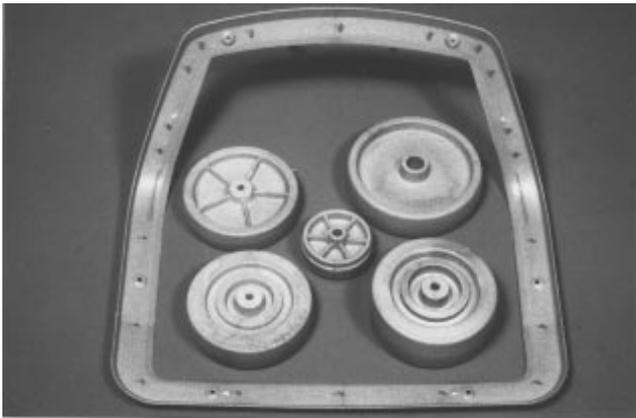
Appendix—Examples of WPF Thermoplastic Composite Products



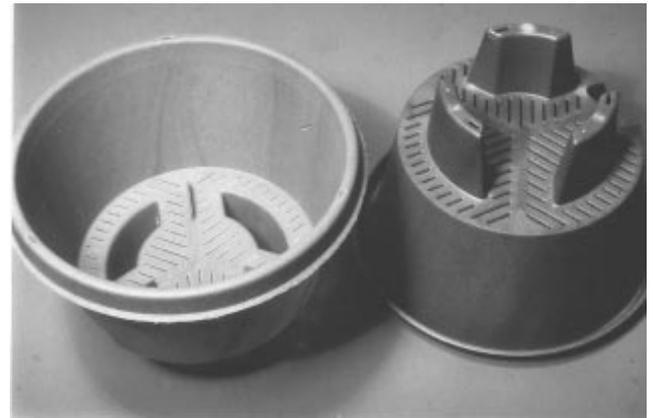
Vacuum cleaner beater bars and battery terminal covers (wood flour/polypropylene).



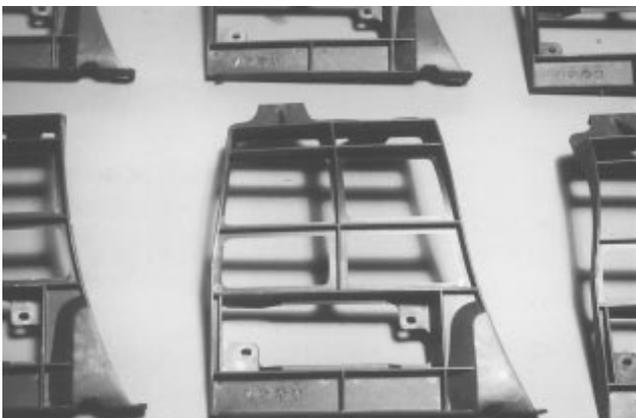
Brackets for curtain valance (colored wood flour/polypropylene).



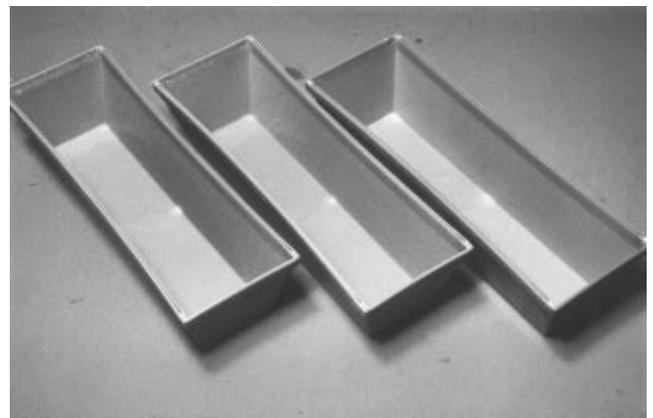
Lawn tractor seat trim piece and castors (wood flour/polypropylene).



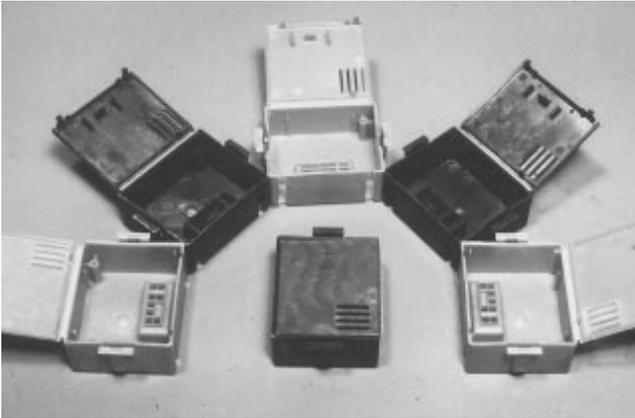
Flower pots (wood flour/polypropylene).



Grill piece for Mazda (newspaper/polypropylene).



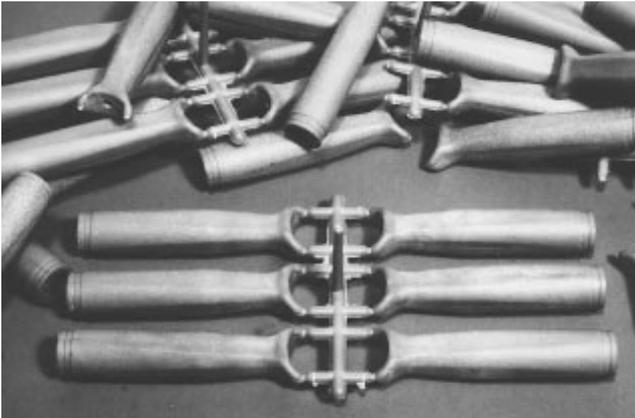
Mudtrays for plaster (wood flour/polypropylene).



Chime boxes for Chrysler (wood flour or newspaper/polypropylene).



Bicycle bottle holders (wood flour or newspaper/polypropylene).



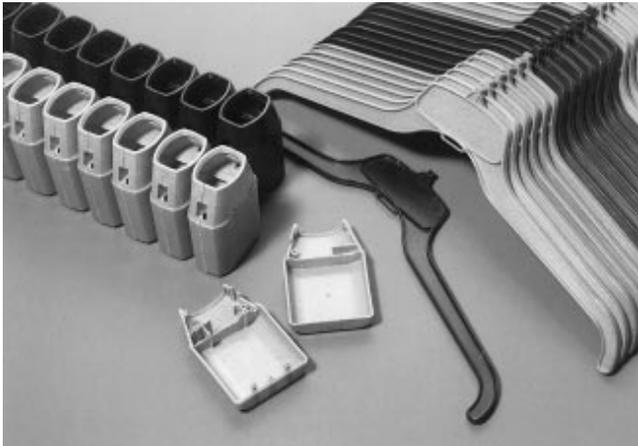
Paint roller handles (wood flour/polypropylene).



Scissor handles (wood flour/polypropylene).



Cosmetic case covers (wood flour/polypropylene).



Flashlight cases and coat hangers (wood flour or newspaper/polypropylene).