EFFECTS OF SEVERAL INGREDIENT VARIABLES ON MECHANICAL PROPERTIES OF WOOD FIBER-POLYOLEFIN COMPOSITES BLENDED IN A THERMOKINETIC MIXER

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ABSTRACT

We examined the influence of several variables on the mechanical properties of wood fiber–polyolefin composites blended in a thermokinetic mixer. A pure cellulose fiber and fibers from old newspaper provided similar performance in matrices of virgin polypropylene or recycled milk bottles (high density polyethylene). Relative to wood flour, these fibrous fillers led to greater strength and modulus with both plastics, to lower impact energy with polyethylene, and to similar impact energy with polypropylene. Compared with the existing commercial wood flour–polypropylene system, the totally recycled polyethylene–newspaper composite provided equivalent strength and modulus, along with greater notched impact energy. Little difference was seen in composites containing a maleated polypropylene additive in the form of the solid anhydride or the emulsified potassium salt, indicating that the additive acted as a dispersing agent and not as a strong coupling agent.

INTRODUCTION

We previously reported the results of studies on wood flour virgin polypropylene composites in which the blending of flour and plastic was performed with a single-screw extruder [1,2]. However, when we attempted the same extrusion blending to systems in which the flour was substituted by fibers from old newspapers, we encountered severe difficulties in maintaining a homogeneous dry blend of plastic pellets with the very low bulk density fibers and in feeding the dry blend into the extruder. Moreover, the extruded product was highly inhomogeneous and contained numerous fiber aggregates.

Investigators at the University of Toronto (UT) demonstrated that excellent dispersion of paper fibers in polypropylene could be obtained by melt blending in a "thermokinetic" blender (the so-called K-mixer) [3,4]. Here, the ingredients were brought above the polypropylene melting point by the transfer of kinetic energy from high speed rotating blades [5] and fluxed briefly before the plastic mass was ejected from the apparatus. Total mixing time was usually <1 min.

We carried out screening experiments with the UT K-mixer and duplicated the UT findings. Subsequently, the UT K-mixer was used to blend a series of wood fiber–thermoplastic mixtures. In these tests, we compared the following sets of variables.

Wood Flour, Commercial Cellulose Fiber, and Newspaper Fiber

The primary commercial, melt-processed wood fiber–thermoplastic composites currently employ wood flour. The data from UT [4] and from our screening tests indicated that properties could be improved significantly by substituting fiber for the flour. We felt

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it important to establish a more direct quantitative comparison between the properties of the commercial wood flour system and those of fiber-containing systems. It was useful, moreover, to obtain a quantitative comparison of the effects on composite properties of a refined cellulose fiber (Solka–Floc)[2] and the less refined lignocellulosic fiber from wastepaper.

Virgin Polypropylene and Recycled High Density Polyethylene

The current commercial wood flour–polypropylene system employs virgin polypropylene (PP-V). A quantitative comparison between that system and one that uses a widely available waste plastic, high density polyethylene from milk bottles (HDPE-MB), was useful.

Maleated Polypropylene Wax Additive

Data from our laboratory [1,2] and elsewhere [6] demonstrated that the powdered, anhydride form of Epolene E-43 (Eastman Chemical Products, Inc., Kingsport, TN) increased the strength and modulus of the wood flour–PP-V system after blending by extrusion. Therefore, the anhydride form might be acting as a coupling agent by chemically bonding to the wood. On the other hand, the emulsified form of E–43 (which could not be forming chemical bonds with the wood) produced even larger increases in the same properties of an air-laid, nonwoven mat composite made from wood fiber and polypropylene fibers [7]. Therefore, we were interested in determining whether one or both of these forms was more beneficial in the melt-processed composites containing fibers instead of flour.

EXPERIMENTAL

The following experimental matrix was tested in a replicated, full-factorial study. Although the data were statistically analyzed, that analysis is not presented here. The variables for polymer, filler, and polypropylene additive (Epolene E-43) were as follows:

1. polymer: PP-V vs. HDPE-MB
2. filler (40/60 weight ratio filler/polymer): wood flour vs. cellulose fiber vs. newspaper fiber (ONP)
3. Epolene E-43 (5 weight percent of filler): solid anhydride vs. emulsified potassium salt

The PP-V was Fortilene 1602 (melt flow index 12) from Solvay Polymers, Inc. (Deer Park, TX); HDPE-MB was recycled from milk bottles (nominal 0.7 melt flow index); wood flour was western pine (-40, +80 mesh (−420 μm, +177 μm)) from American Woodfiber Co. (Schofield, WI); cellulose fiber was Solka–Floc BW-40 (60 μm average length) from James River Corp. (Hackensack, NJ); ONP was an overproduction issue from the Milwaukee Journal/Sentinel Inc. (Milwaukee, WI), hammermilled to approximately 15-mm flakes before use; Epolene E–43 was obtained from Eastman Chemical Products, Inc. The solid E–43 powder was added to the filler prior to mixing in the K-mixer; the emulsified E-43 was applied by room temperature evaporation from a slurry of filler in aqueous emulsion.

The plastics and fillers were blended in a 1-L K-mixer. The fillers were mixed for approximately 1 min, polymer was added, and mixing was continued for about 1 min on average. For HDPE-MB, a 185°C dump temperature and a speed of 2,800 rpm were used; for PP-V, the conditions were 200°C and 3,000 rpm. Several 250-g batches were blended for each composition of each replicate matrix, and the batches were granulated and dry-blended.

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Specimens for testing were made by injection molding, using a Frohring Minijector at 4.5 to 9.0 MPa, 30 to 60 s dwell time, and mold temperature of 30°C. Prior to molding, the granulated mixes were dried at 105°C for 4 to 5 h. After molding, the specimens were kept under desiccant for at least 3 days before testing at 23°C, with minimal exposure to ambient humidity.

Maximum tensile strength and modulus were measured in conformance with ASTM D638M-84, with specimen type IV at a nominal strain rate of 0.15 mm/mm-min [8]. Specimens for flexural (three-point bending) tests were 127 by 12.7 by 3.2 mm; maximum strength and modulus were measured in conformance with ASTM D790-84a, using a support span of 102 mm and a crosshead rate of 5 mm/min [8]. Impact energy was measured with an Izod impact tester; specimens were 64 by 12.7 by 6.4 mm, and tests conformed to ASTM D256-84 [8]. Data points reported here are the average of 12 specimens, 6 each from the replicate matrices. Overall coefficients of variation were approximately as shown in Table I.

RESULTS AND DISCUSSION

Figures 1 to 6 summarize the results for tensile and flexure (three-point bending) strength and modulus and for notched Izod impact energy. Figures 1 to 5 are three-dimensional plots of responses as a function of filler type (horizontal axis) and form of E-43 (angled axis), with separate response surfaces for PP-V and HDPE. Figure 6 is a cross plot of flexure strength against notched impact energy.

Within the errors (see Table I), little or no effect of E-43 form was found for either PP-V or HDPE-MB systems (Figs. 1 to 5). This result indicates that reported increases in strength and modulus and losses in impact (Figs. 1, 2, and 6) upon incorporating E-43 were not due to the form of the carboxyl (anhydride or potassium salt) and the nature of its interaction with the filler or to any ability of the E-43 to co-crystallize with the matrix polymer. Therefore, the E-43 apparently functions primarily as a wetting agent, enhancing the dispersion of the polar fillers into the nonopolar polymers.

In general, the fibrous fillers provided greater reinforcement (higher strength and modulus)(Figs. 1 to 4, Table II) to either matrix than did the wood flour. Moreover, the inexpensive ONP was at least as beneficial in this regard as the cellulose fiber (BW-40). Therefore, despite the breakage of fibers during melt processing, the fibrous fillers appear to have retained a greater aspect ratio than did the wood flour.

The PP-V composites consistently had greater strength and modulus than did the HDPE-MB composites (Figs. 1, 5, Table III). This difference reflects the differences in the matrix polymers themselves and not the fact that the HDPE-MB is recycled; one reprocessing step has been shown to have little effect on the strength and modulus of unfilled or wood-filled HDPE [9].

The impact behavior was less straightforward with respect to the influence of both filler tyype and matrix polymer. However, the data scatter for the unnotched impact energy was so large that we restrict the discussion here to notched impact energy. With a given filler, the notched impact energy for HDPE-MB systems was clrarly superior to that for PP-V systems (Figs. 5 and 6, Table III); this too is consistent with the behavior of the matrices themselves. Substitution of fibrous fillers for wood flour brought about an average 20-percent decreased in notched inpact energy for the HDPE-MB systems (Table II), which was accompanied by larger increases in strength and modulus. This negative correlation between impact energy and strength is part of the more general one illustrated in Figure 6 and is consistent with general observations that increased reinforcement inhibits energy-adsorbing processes during fracture propagation.

In contrast to the behavior for HDPE-MB systems, substitution fibers for wood flour in polypropylene systems may increase notched impact energy (Table II). Why the PP-V

Table I. Approximate coefficients of variation

Property	Coefficient of variation (%)
Tensile strength	4
Tensile modulus	6
Flexure strength	3
Flexure modulus	7
Notched impact energy	8
Unnotched impact energy	13

Table II. Property changes from substituting fiber for flour

Property	Property change (%)[a]	
	PP-V matrix	HDPE-MB matrix
Flexure strength	28	35
Flexure modulus	20	33
Tensile strength	30	33
Tensile modulus	13	36
Notched impact energy	12	–20
Unnotched impact energy	50[b]	–3

[a] 100 × [(Average of property with BW-40 and ONP ÷ Property with WF) - 1].

[b] Values questionable because of data scatter (see Table I).

Table III. Effect of polymer on properties of composites[a]

Variable	Tensile strength (MPa)	Tensile modulus (GPa)	Flexure strength (MPa)	Flexure modulus (GPa)	Impact energy (J/m)	
					Notched	Unnotched
PP-V	40.9	4.30	68.1	3.64	20.7	94.0
HDPE-MB	33.0	3.29	48.1	2.75	33.1	79.0
(PP-V to HDPE-MB ratio)	(1.24)	(1.31)	(1.42)	(1.32)	(0.63)	(1.19)

'Overall average.

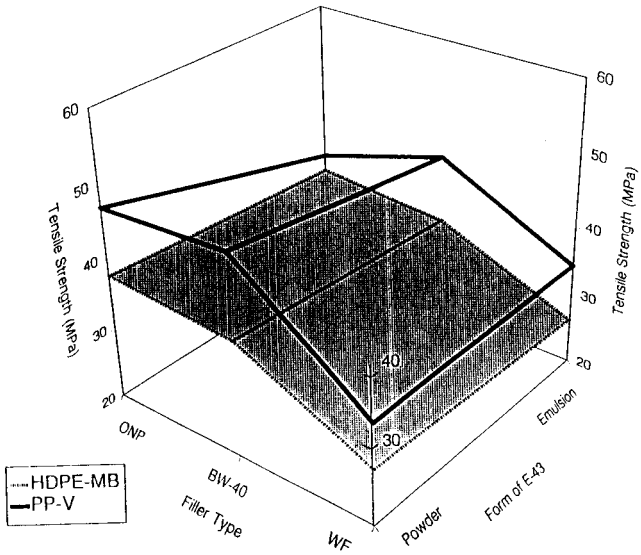

Figure 1–Tensile strength of composites. BW-40 is cellulose fiber, HDPE-MB recycled high density polyethylene from milk bottles, ONP newspaper fiber, PP-V virgin polypropylene, and WF wood flour.

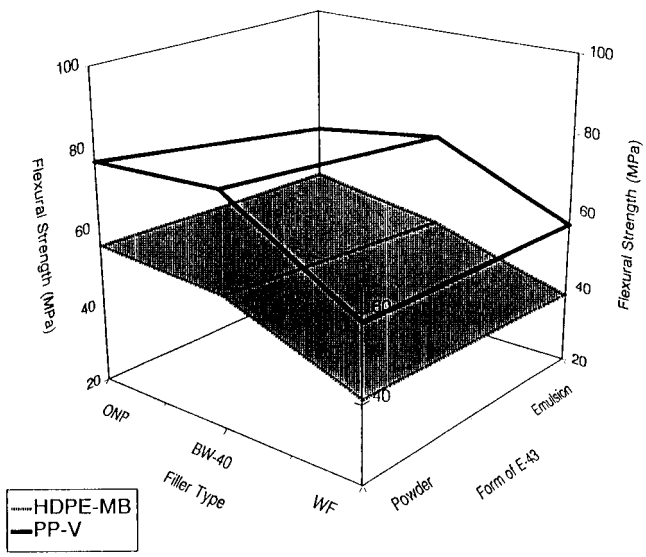

Figure 2–Flexural strength of composites (abbreviations defined in Fig. 1).

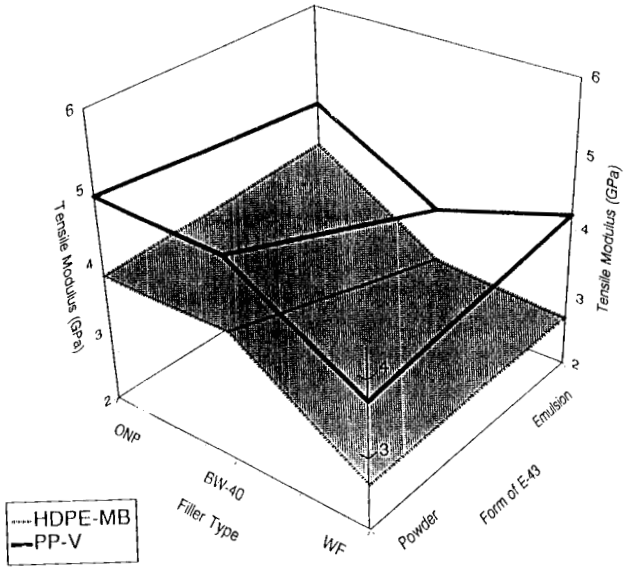

Figure 3–Tensile modulus of composites.

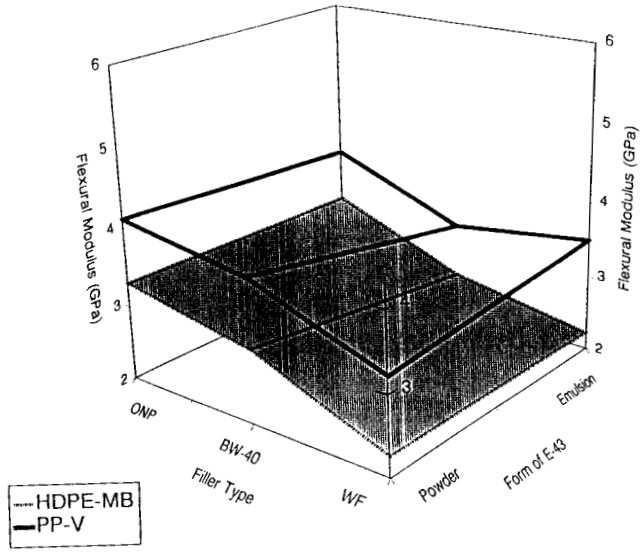

Figure 4–Flexural modulus of composites.

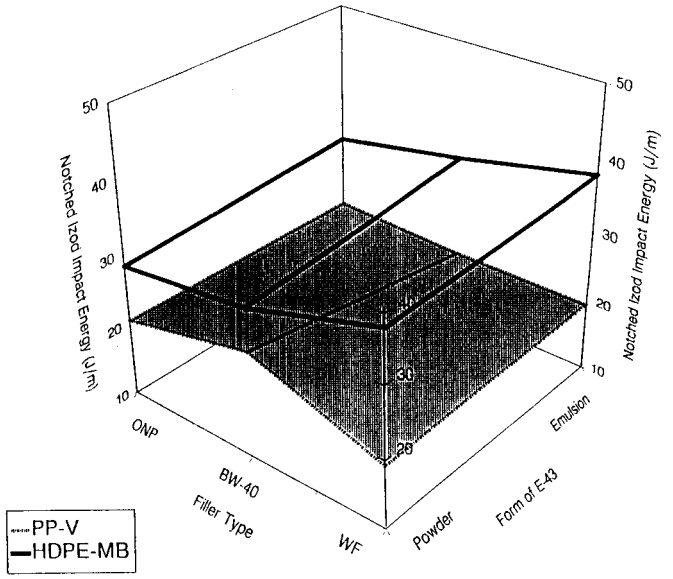

Figure 5-Notched impact energy of composites.

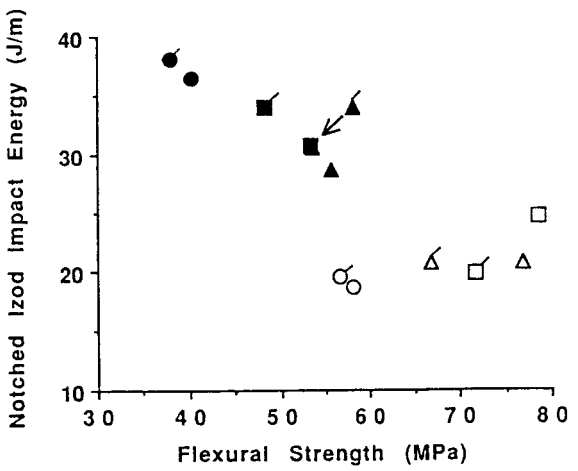

Figure 6-Correlation between notched impact energy and flexural strength,. PP-V, open symbos; HDPE-MB, closed symbols, emulsion E-43 systems, flagged symbolsd; poedered E-43 systems, unflagged symbols; wood flour composites, circles; BW-40 composites, squares; ONP composites, triangles.

Table IV. Comparison of HDPE-MB–ONP and PP-V–wood flour composites

Property	Ratio[a]
Tensile strength	1.07
Tensile modulus	0.98
Flexure strength	0.95
Flexure modulus	0.97
Notched impact energy	1.55
Unnotched impact energy	0.95

[a] Average property value for HDPE-MB–ONP systems to average value for PP-V–wood flour systems.

systems should be able to maintain impact energy in the presence of greater reinforcement is not clear. One possible explanation is that the greater compatibility of the PP-V matrix with the polypropylene segments in E–43 may enhance fiber pull-out resistance and thereby counterbalance the loss of energy absorption resulting from reinforcement.

Table IV compares the properties of the system using recycled ingredients (HDPE-MB–ONP) with those of the current commercial system (PP-V–wood flour). Notched impact energy for the "recycled" system was greatly improved whereas all other properties were identical within experimental error. Based on these mechanical properties, the recycled HDPE-MB–ONP system could therefore advantageously substitute for the PP-V–wood flour system. However, this mechanical property advantage will very likely be offset in some commercial melt processes by the greater melt viscosity of the HDPE-MB system.

CONCLUDING REMARKS

Wood-filled polyolefin composites were blended in a thermokinetic mixer, and mechanical properties were measured on injection-molded specimens. The major findings were as follows:

1. Form of Epolene E-43 (solid powder or emulsified potassium salt) had little effect, leading to the conclusion that the E–43 acts primarily as a dispersing agent and not as a strong coupling agent.
2. The fibrous fillers (cellulose or old newspaper (ONP)) provided greater reinforcement than did the wood flour; for example, on average the strength of the fiber-containing systems was >30 percent greater than that of the wood flour systems. The ONP fibers were at least as beneficial as the pure cellulose fiber in this regard.
3. The fibrous fillers led to lower notched impact energy in the milk bottle polyethylene systems (HDPE–MB) relative to the wood flour (average ratio of fiber wood flour impact energy was 0.8) but caused little change in the virgin polypropylene (PP-V) systems.
4. The PP-V composites had greater strength and modulus than the HDPE-MB composites (average ratio of polypropylene to polyethylene strength or modulus was 1.24 to 1.42) but lower notched impact energy (average ratio was 0.63).
5. The composite containing both recycled filler and matrix (ONP and HDPE-MB) possessed mechanical properties nearly identical to those of the composite made with PP-V and wood flour, with the notable exception of approximately >50-percent notched impact energy for the recycled system.

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