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PROPERTIES OF MELT-BLENDED COMPOSITES FROM POST-CONSUMER POLYPROPYLENES AND WASTEPAPERS

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INTRODUCTION

Paper and plastics are two of the largest components in municipal solid waste (MSW), about 40 and 8 weight percent, respectively [1]. The combination of paper and plastics into fiber-reinforced plastic composites offers an extremely attractive alternative to current disposal by landfilling. This is especially true in view of (1) the relatively high strength and modulus to weight ratio of cellulose fibers and (2) the simplifications introduced because paper contaminants (e.g., ink, coatings) probably have little adverse effect on composite behavior.

In recent years, numerous studies have examined the properties of composites made from virgin or waste plastics and virgin or wastepapers. Particular emphasis has been on high-density polyethylene from milk bottles, virgin polypropylene, and old newspapers [2-6]. The old newspapers fibers clearly provide significant reinforcement to these plastics and improve mechanical properties when compared to wood flour as a filler [2,3-6].

In this study, we extend the database on waste plastic and wastepaper composites to define the potential of some waste polypropylenes when combined with old newspapers or old magazines. The following materiels were investigated:

1. Virgin polypropylene (VPP). This was used as a baseline plastic.
2. Waste polypropylene (PP) from ketchup bottles (KPP). Although PP is present in MSW at less than 5% of the total plastics. KPP could be readily separated from the waste stream if applications would warrant separation.
3. Waste PP from auto battery cases (BPP). Battery case PP appears to offer considerable potential because the cases are nearly 100% recycled and the resultant plastic is available in large quantities in a "pure" state. In addition, BPP is an ethylene–propylene copolymer with better impact resistance than that of PP homopolymer.
4. Old newspapers (ONP). Its potential was previously noted. It is readily separable from the waste stream and constitutes about 6% of the total MSW.
5. Old magazines (OMG). Although OMG constitutes only about 1% of the MSW, it is readily separable. It was also of interest to determine whether OMG coatings and clay fillers would alter the reinforcing capabilities of the fibers.

EXPERIMENTAL PROCEDURES

A full-factorial experimental design was replicated to investigate the mechanical properties of composites from KPP or BPP with 40 weight percent ONP or OMG. Data for the comparable ONP and VPP systems were obtained from previous studies [4,5].

The BPP (melt flow index 10) was obtained from Gopher Smelting and Refining Company[1] (Eagon, Minnesota). The KPP (melt flow index 3) was from Wheaton Plastic Recycling (Millville, New Jersey). Epolene G-3002 (a maleated polypropylene) was from Eastman Chemical Products, Inc. (Kingsport, Tennessee) and was added at 3 weight percent of the fiber. Epolene G-3002 is a maleated PP with acid number 63 and approximate M_n 11,000. Epolene G-3002 was found to improve strength, stiffness, and impact behavior of wood flour/PP composites [7]. The ONP was an overproduction issue from the Milwaukee Journal/Sentinel Inc.. and OMG was a representative sample from the Madison. Wisconsin, wastestream that was provided by the Madison Recycling Center. Both ONP and OMG were ground to approximately 5-mm flakes before use.

The blends were compounded in a thermokinetic mixer (K Mixer; Synergistics, Inc., St. Remi. de Napierville, Quebec) for approximately 1.5 min at 4,500 rpm and a dump temperature of 170°C to 190°C. The resulting blends were granulated and stored over desiccant until molded. Fiberizing OMG in the thermokinetic mixer was more difficult than was fiberizing ONP, resulting in poorer dispersion of the OMG in the matrix plastic.

Test specimens were prepared using a Frohring Minijector model SP50 (Newbury Industries, Inc., Newbury, Ohio) plunger-type injection. molding

Table 1. Measured mechanical properties[a]

Variable		Tensile				Flexural		Izod impact energy (J/m)	
Plastic	Fiber	Strength (MPa)	Modulus (GPa)	Elongation (%)	Energy (J)	Strength (MPa)	Modulus (GPa)	Notched	Unnotched
KPP	None	36.5	1.62	7.8	1.14	48.4	1.49	62.0	>800
	ONP	52.3	4.03	3.7	0.74	73.8	3.30	30.6	167
	OMG	38.9	3.55	3.1	0.48	61.8	3.01	34.2	138
BPP	None	24.5	1.32	7.4	0.80	31.4	1.05	165.0	>800
	ONP	42.5	3.98	3.4	0.58	59.9	3.16	34.3	150
	OMG	31.8	3.44	3.0	0.40	49.2	2.69	41.8	125
VPP	None	31.5	—	—	—	41.1	1.43	23.8	650
	ONP	52.3	4.12	—	—	82.0	4.20	20.4	190

[a]Strength is maximum value; elongation is at maximum strength; energy is area under stress/strain curve to maximum strength. Dashes indicate data not available.

machine at 204°C with residence time approximately 1 min, ram pressure 3.4 to 8.9 MPa, and mold temperature 30°C. The specimens were stored over desiccant for at least 3 days before testing. Specimens and testing procedures followed ASTM specifications [8]. Mechanical properties were measured dry at 23°C and included the following:

Tensile strength and modulus
Tensile elongation at maximum load
Tensile energy to maximum load
Flexural (three-point bending) strength and modulus
Izod impact energy, notched and unnotched

At least five specimens were tested for each composite blend and property

RESULTS AND DISCUSSION

Table 1 summarizes the measured properties and Table 2 the mean coefficients of variation (COVs) for each test. All tests had COVs less than 10% except for unnotched impact energy. The large COV for unnotched impact energy limits the sensitivity of that measurement.

Table 2. Approximate coefficients of variation (COV)

Property	COV (%)
Tensile	
strength	5
modulus	5
energy to maximum load	8
elongation at maximum load	6
Flexural	
strength	2
modulus	5
Impact	
notched	7
unnotched	14

Table 3. Main effects of variables

Property	Overall mean[a]	Main effect[b] KPP to BPP	Main effect[b] OMG to ONP
Tensile			
modulus (GPa)	3.8	[c]	+0.5 (+15)
strength (MPa)	61.4	−8.4 (−18)	+12.1 (+34)
energy (J)	0.55	−0.13 (−20)	+0.22 (+50)
elongation (%)	3.3	−0.2 (−6)	+0.4 (+17)
Flexural			
modulus (GPa)	3.0	−0.2 (−7)	+0.4 (+12)
strength (MPa)	61.2	−13.3 (−19)	+11.3 (+17)
Impact			
Notched (J/m)	35.3	+ 5.6 (+18)	−5.6 (−14)
Unnotched Impact	145	[c]	[c]

[a]Average of all variables and trials.
[b]Main effect is change in property value resulting from the particular variable, averaged over the other variable. Number in parentheses is percentage of change. Interactions were not found at the 95% confidence level.
[c]Not significant at 95% confidence level.

Table 3 is a statistical summary of the factorial design with two variables: matrix type (KPP and BPP) and filler type (OMG and ONP). Significant interaction terms were not found at the 95% confidence level. Unnotched impact energy showed no change. probably because of the test insensitivity as a result of the large COV.

Table 4. Change in property after addition of 40% filler to plastic

Property	Change (%)[a] KPP	BPP	VPP
Tensile			
modulus	+138	+184	—
strength	+28	+52	+66
energy	−47	−40	—
elongation	−56	−58	—
Flexural			
modulus	+113	+164	+194
strength	+40	+74	+100
Impact			
notched	−48	−77	−14
unnotched	−80	−83	−70

[a]Average of ONP- and OMG-filled values used in calculations for KPP and BPP. VPP calculations based on ONP-filled composites; Dashes indicate data not available.

Effects of Filler

Table 4 shows the property changes brought about when 40% filler was added to each polymer. Both tensile and flexural moduli improved more than 100% for KPP and more than 150% for BPP. Modest improvements in strength properties were observed (~30% to 75%), but BPP was again reinforced to a greater extent. Energy to maximum load and elongation at maximum load decreased, and impact energy dropped sharply (e.g., approximately 59% reduction in unnotched impact energy for KPP and 80% for BPP). Thus, incorporating wastepaper fibers led to stiffer and stronger but more brittle materials compared to the unfilled plastics.

All tensile and flexural properties improved when OMG was replaced with ONP (Tables 1, 3); the largest increases were 50% in tensile energy absorbed and 34% in tensile strength. At the same time, a small decrease occurred in notched Izod The increase noted for unnotched Izod in Table 1 was not statistically significant. The greater reinforcement produced by ONP is consistent with its better fiberization and dispersion. Perhaps the clay and other materials in the OMG altered the structure–property relations by hindering both fiber dispersion and effectiveness of the Epolene G3002 for bonding between fiber and plastic. Given the complexity of these systems, speculation about the marginally better notched impact of the OMG system seems unwarranted.

Effects of Matrix Type

All properties except the notched impact energy were decreased by small amounts when BPP was used instead of KPP (Table 3). These changes qualitatively followed the properties of the matrices. The softer, weaker, and tougher BPP was more sensitive than was KPP to filler addition (Table 4), but overall BPP did not achieve as good a balance of properties as did the KPP (Table 1). In particular, the superior impact energy of the unfilled BPP (an ethylene–propylene copolymer) was poorly transferred to the composite system. However, both the KPP snd BPP composites had notched impacts significantly greater than that of the VPP composite (Fig. 1). Thus, BPP and KPP in certain composite applications could provide attractive cost and performance benefits compared to the virgin PP.

CONCLUDING REMARKS

From this comparison of composites contaning waste ONP or OMG fibers in BPP or KPP waste plastics, we present the following:

1. In all cases, adding 40% fiber to the recycled plastics resulted in stiffer, stronger, but more brittle materials.
2. The OMG was more difficult than ONP to disperse into the plastic matrix, and OMG produced a balance of mechanical properties that probably was less desirable than the balance produced by ONP.

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

3. Mechanical properties of KPP and BPP composites qualitatively followed those of the two matrix polymers. The KPP composites possessed somewhat better tensile and flexural properties than did BPP composites but were slightly less resistant to notched impact energy.

4. The BPP composites lost much of the BPP polymer's superior impact behavior. However, both the KPP and BPP composites possessed better impact than that of the analogous virgin PP composite.

5. From the viewpoint of availability, processibility, and composite properties, ONP appears to provide more promise for commericial applications than does OMG. Balancing the factors of cost, availability, and composite properties, BPP should be of more interest than KPP and a useful replacement for virgin PP in some applications

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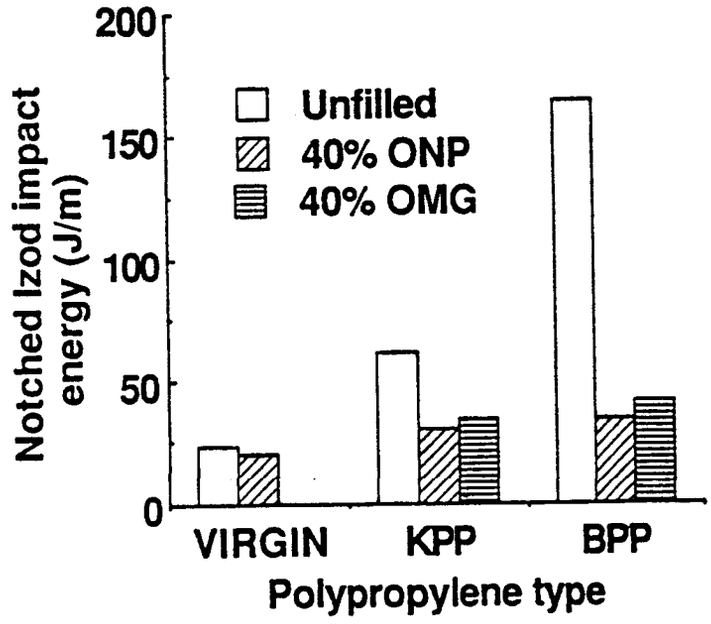

Figure 1. Notched impact energy of various recycled composite systems.