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# Effects of Composition and Polypropylene Melt Flow on Polypropylene—Waste Newspaper Composites

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## INTRODUCTION

In 1986, the United States disposed of 160 million metric tons of municipal solid wastes. Landfill sites that neighbors will tolerate are becoming increasingly hard to find [1]. In the next 15 years, 75 percent of all landfills will be closed, and by the year 2000, the United States will be short 56 million metric tons per year of disposal capacity [1].

Despite advances in paper recycling technology, paper remains the most prominent recyclable constituent of city trash Domestic paper and paperboard manufacture in 1989 amounted to 71 2 million metric tons, much of which was promptly discarded; only 26 percent was recycled [2]

Because of processing cost, waste disposal, and product value considerations, the value placed on the abundant grades of salvaged paper (for example, newspaper) is low, amounting to very few pennies per kilogram. The low cost of wastepaper and the favorable strength to weight ratio of cellulose fibers give such fibers considerable promise as reinforcing fillers in plastic composites. Contaminants (inks, coatings, etc.), which have very adverse effects on paper recycled from wastepaper, have little adverse effect on composites containing wastepaper

Investigations by Woodhams and others [3, 4] and preliminary work at the Forest Products Laboratory (FPL) showed that improvements in mechanical properties should result from the use of old newspaper (ONP) fibers instead of wood flour as a filler in polypropylene (PP) composites. Woodhains and his colleagues also showed that the use of a laboratory-sized thermokinetic mixer (K-mixer) greatly alleviated two serious problems with ONP. (a) difficulties in handling the ONP fiber–PP feed as a result of differences in bulk density of ONP fibers and PP and (b) difficulties in completely dispersing ONP fibers in PP.

Recently, a program was initiated by the FPL, the University of Toronto, and others in cooperation with the Wisconsin Department of Natural Resources to establish the commercial feasibility of extruded or injection-molded composite products from PP and ONP. The first phase of the program was to provide a data base for rationally selecting formulations for commercial evaluation by cooperating manufacturers of extruded and injection-molded products. For that purpose, composites with a range of ONP contents and PP melt flow index were blended in the University of Toronto laboratory-sized K-mixer, and their mechanical and rheological properties were measured. These composites were also compared with composites containing wood flour in place of ONP. This report summarizes the mechanical properties of the composites

## EXPERIMENTAL

A full-factorial experimental design was used to investigate the mechanical properties of PP–ONP composites (Although the data were statistically analyzed, that analysis is not presented here.) The matrix was designed to include the range of mechanical and rheological properties of interest to our cooperating commercial extruders and injection molders.

| Variable | Level |
|---|---|
| Melt flow index | 3, 12, or 30 g/10 min |
| Filler content | 32, 37, or 42 weight percent |

To compare ONP with the more conventional filler, wood flour (WF), composites were produced with 42 percent WF instead of ONP for each PP melt flow index. Because previous studies denionstrated that the maleated polypropylene Epolene E-43[2] (Eastman Chemical Products, Inc., Kingsport, TN) aided the dispersal of wood-based fillers in PP and improved some properties [5-7], all compositions contained 5 percent Epolene E-43 (weight percentage of filler) All ONP and WF runs were replicated to measure run-to-run variability.

The polypropylenes used were Fortilene 9101, 1602, and 1902 homopolymer spheres (Solvay Polimers Inc., Deer Park, TX) with melt flow indices of 3, 12, and 30 g/10 min (230°C/2.16 kg), respectively. Epolene E-43 has an acid number of 47 and an approximate molecular weight of 4,500. The wood flour (American Wood Fibers, Inc., Schofield, WI) was number 402, 420-$\mu$m (nominal 40–mesh) western pine. The newspaper was an overproduction issue from the Milwaukee Journal–Sentinel, Inc. (Milwaukee, WI) and was hammermilled to approximately 15-0mm flakes before use.

The blends were melt-compounded in a 1-L K-mixer, a high intensity kinetic mixer in which the sole source of heat is the kinetic energy of the high-speed blades [8]. The newspaper flakes were converted to fiber by mixing with the Epolene E-43 in the K-mixer at 3,300 rpm for approximately 90 s, after which the polymer was added. During continued mixing for approximately 60 s at 3,300 rpm, the polymer was melted, the fibers were dispersed into the melted polymer, and the resultant blend was heated to 185°C, at which point the blend was automatically dumped from the mixer. For each matrix trial (formulation), approximately twenty 0.25-kg batches were prepared; the batches were hammermilled, dry-blended, and stored over desiccant until molded.

Test specimens were prepared using a Frohring minijector model SP50 (Newbury Industries, Inc., Newbury, OH) plunger-type injection molding machine at 204°C with a residence time of approximately 1 min, ram pressure of 3.4–8.9 MPa, and mold temperature of 30°C. The specimens were stored over desiccant for at least 3 days before testing. Specimens and testing procedures followed ASTM specifications [9]. At least five specimens were tested for each composite blend from each replicate matrix.

## RESULTS AND DISCUSSION

Table I summarizes the mean coefficients of variation for each test. The high coefficient of variation for the impact tests limits any interpretation of relations between observed changes in impact behavior and the matrix variables.

### Filler Effects

For reference, Table II compares the properties of unfilled PP composites with those of composites filled with 42 percent ONP or WF. The major effects were the increase in tensile modulus and heat distortion temperature and the loss in unnotched impact energy.

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[2] 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.

Increasing the ONP content from 32 to 42 weight percent had little significant effect on tensile or flexural properties (Table 111, Figs. 1 and 2). (In these figures, as well as Figure 3, values were averaged for the three PP melt flow indices.) Considering the rather large coefficients of variation of the impact properties (Table I), the observed changes in those properties (Table III) may be more apparent than real. However, the approximately 10°C increase in heat distortion temperature at 1.83 MPa was undoubtedly real.

Substituting ONP for WF at the 42-percent level clearly increased both tensile and flexural strengths (Table III, Fig. 2), that is, overall increases of 29 and 24 percent, respectively. These improvements were undoubtedly brought about by the greater aspect ratio of the ONP fibers, which may have also introduced fiber orientation in the injection-molded test specimens. The same substitution of ONP for WF may also have benefited unnotched impact energy (Table III, Fig. 3) but not notched impact energy, indicating that the fibers introduced energy-absorbing processes that affected fracture initiation hut not fracture propagation. Again, an approximately 10°C improvement in heat distortion temperature was observed.

### Polypropylene Melt Flow Index Effects

A tenfold increase in PP melt flow index caused no significant changes in any of the measured properties (Table Ill, Figs. 4 and 5). This result is consistent with the small differences in properties of the unfilled polymers, for example, 5 percent in tensile strength and 25 percent in flexural modulus [10]. We conclude, therefore, that the absence of property changes in the composites indicates that the tenfold difference in polymer viscosity did not greatly alter the efficiency of fiber dispersion during melt blending in the K-mixer.

## SUMMARY

Polypropylene composites filled with wood flour (WF) or old newspaper (ONP) were blended in a laboratory thermokinetic mixer. Mechanical properties were measured on injection-molded specimens. The effects of filler content (32, 37, and 42 percent) and polypropylene (PP) melt flow index (3, 12, and 30 g/10 min) on mechanical properties were determined; properties of the 42-percent ONP composites were compared with those of comparable 42-percent WF composites. The major findings were as follows:

- Replacing WF with ONP fibers at 42 weight percent filler in PP composites increased flexural strength (24 percent), tensile strength (29 percent), unnotched impact energy (43 percent), and heat distortion temperature (12 percent at 1.83 MPa). These enhancements are attributed to the greater aspect ratio of the ONP fibers.

- Increasing ONP content from 32 to 42 weight percent had little effect on all properties except for an approximately 10°C increase in heat distortion temperature at 1.83 MPa.

- Mechanical properties of ONP-filled PP composites were insensitive to the tenfold change in PP melt flow index. Therefore, effectiveness of dispersing the ONP fibers in the polymers apparently was also independent of differences in melt viscosity.

These findings confirm both the ability to prepare ONP–PP composites with a laboratory thermokinetic mixer (K-mixer) and the mechanical property advantages to be achieved by using ONP fibers instead of WF. Within the range of ONP contents and PP melt flow indices examined, it is likely that the choice of ONP–PP formulations for commercial evaluation will be based almost solely on the melt rheological behavior of the composites at temperatures and shear rates specific to particular manufacturing operations.


## ACKNOWLEDGMENTS

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## REFERENCES


1. U.S. Environmental Protection Agency. 1988. "Characterization of municipal solid waste in the United States 1960 to 2000."
2. American Paper Institute. 1990. "1990 statistics of paper, paperboard, and wood pulp." New York.
3. R.T. Woodhams, G. Thomas, and D.K. Rogers. 1984. Polym. Eng. Sci. 24(15): 1166–1171.
4. R.T. Woodhams, S. Law, and J.J. Balatinecz. 1991. Proceedings, Wood Adhesives Symposium; 1990 May 16–18; Madison, WI: 177–182.
5. G.E. Myers, I.S. Chahyadi, C.A. Coberly, and D.S. Ermer. 1991. Intern. J. Polym. Mater. 15: 21–44.
6. G.E. Myers, I.S. Chahyadi, C. Gonzalez, C.A. Coberly, and D.S. Ermer. Intern. J. Polym. Mater. Accepted for publication.
7. D.J. Olsen, Preprints, ANTEC 91, 1886–1891.
8. S. Frenken, D. Lyons, and W.E. Baker. 1991. Preprints, ANTEC 91, 40–45.
9. American Society for Testing Materials. 1986. Annual Standards, Vol. 08.01, Sec. 8, Philadelphia, PA.
10. Solvay Polymers, Inc. 1990. Technical Brochure, Fortilene Polypropylene Product Line.


Table I--Approximate sample-to-sample coefficients of variation for composite properties

| Property | Coefficient of variation[a] (percent) |
|---|---|
| Tensile strength | 3 |
| Tensile modulus | 5 |
| Flexural strength | 2 |
| Flexural modulus | 4 |
| Notched impact energy | 19 |
| Unnotched impact energy | 14 |
| Heat distortion temperature | 2 |

[a] Mean value for each matrix variable.

Table II--Properties of filled (42% by weight) and unfilled composites

| Composite | Tensile modulus (GPA) | Tensile strength (MPa) | Impact energy, notched (unnotched) (J/m) | Heat distortion temperature (°C) 0.46[a] MPa | 1.83[b] MPa |
|---|---|---|---|---|---|
| Unfilled[c] | 1.2 | 33.8 | 21 (720) | 115 | 55 |
| ONP-filled[c] | 4.6 | 49.5 | 23 (140) | 146 | 114 |
| WF-filled[c] | 4.4 | 38.3 | 23 ( 87) | 141 | 102 |

[a] Determined by Solvay Polmers, Inc. (Deer Park, TX).
[b] Determined by University of Toronto.
[c] Values averaged over three polypropylene melt flow indices.

Table III -- Mean effects of composition on mechanical properties (percent change)[a]

| Change in composition | Tensile modulus | Tensile strength | Flexural modulus | Flexural strength | Impact energy, notched (unnotched) | Heat distortion temperature 0.46 MPa | 1.83 MPa |
|---|---|---|---|---|---|---|---|
| 32% to 42% ONP | +9 | < 2 | +12 | +4 | +20 (−18) | +3 | +13 |
| 42%WF to 42% ONP | +6 | +29 | +4 | +24 | < 2 (+43) | +5 | +12 |
| PP MFI 3 to 30 g/10 min | +4 | −4 | < 2 | −6 | −13 (−12) | 0 | 0 |

[a] Percent change of mean property value. Values for ONP content effect and change in WF to ONP effect were averaged for all PP melt flow indices (MFIs). Values for the effect of MFI were averaged for all filler contents.

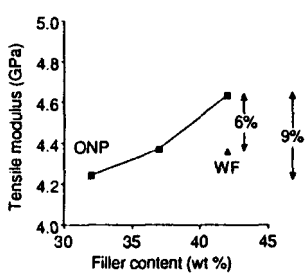


Figure 1–Composite tensile modulus as a function of filler content. ONP is old newspaper, WF is wood flour. Data averaged for three PP melt flow indices.

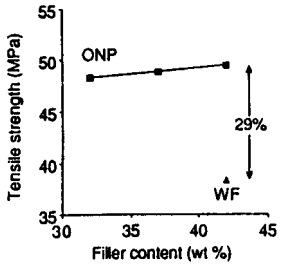


Figure 2–Composite tensile strength as a function of filler content. Data averaged for three PP melt flow indices.

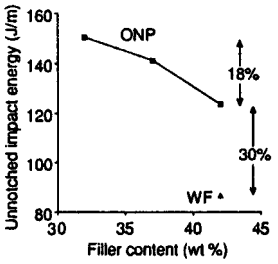


Figure 3–Composite unnotched Izod impact energy as a function of filler content. Data averaged for three PP mall Row indices.

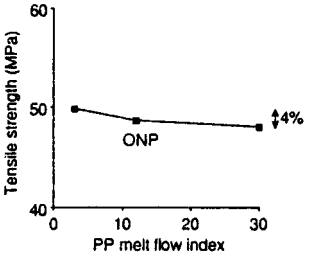


Figure 4-Composite tensile modulus as a function of polypropylene melt flow index (g/10 min, 230°C/2.16 kg) Data averaged for three levels of filler content.

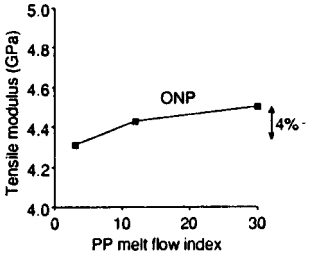


Figure 5–Composite tensile strength as a function of polypropylene melt flow index (g/10 min. 230°C/2.16 kg). Data averaged for three levels of filler content.



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