

# Mechanical Properties and Morphology of Impact Modified Polypropylene-Wood Flour Composites

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**ABSTRACT:** The mechanical properties and morphology of polypropylene/wood flour (PP/WF) composites with different impact modifiers and maleated polypropylene (MAPP) as a compatibilizer have been studied. Two different ethylene/propylene/diene terpolymers (EPDM) and one maleated styrene-ethylene/butylene-styrene triblock copolymer (SEBS-MA) have been used as impact modifiers in the PP/WF systems. All three elastomers increased the impact strength of the PP/WF composites but the addition of maleated EPDM and SEBS gave the greatest improvements in impact strength. Addition of MAPP did not affect the impact properties of the composites but had a positive effect on the composite unnotched impact strength when used together with elastomers. Tensile tests showed that MAPP had a negative effect on the elongation at break and a positive effect on tensile strength. The impact modifiers were found to decrease the stiffness of the composites. Scanning electron microscopy showed that maleated EPDM and SEBS had a stronger affinity for the wood surfaces than did the unmodified EPDM. The maleated elastomers are, therefore, expected to form a flexible interphase around the wood particles giving the composites better impact strength. MAPP further enhanced adhesion between WF and impact-modified PP systems. EPDM and EPDM-MA rubber domains were homogeneously dispersed in the PP matrix the diameter of domains being between 0.1-1  $\mu\text{m}$ . © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* **67**: 1503-1513, 1998

**Key words:** polypropylene-wood flour composites; mechanical properties; morphology; MAPP

## INTRODUCTION

The main purpose of adding cellulose-based filler to thermoplastics is to reduce the cost per unit volume and improve stiffness. Low price cellulose based fillers such as wood flour, wood fibers, and cellulose fibers have high stiffness, low density, and are recyclable. However, there is poor interfacial adhesion between the hydrophobic matrix and the hydrophilic filler, which usually results in decreased toughness.<sup>1-3</sup>

The toughness of filled polymers can be improved in several ways: 1) increase the matrix toughness; 2) optimize the interface (or interphase) between the filler and the matrix through the use of coupling agents, compatibilizer, and sizes; 3) optimize the filler-related properties such as filler content, particle size, and dispersion; 4) aspect ratio and orientation distributions also play a role in toughness of composites containing more fibrous materials.<sup>4</sup> This investigation focused on the first two approaches.

### Toughening of PP Matrix

The matrix material plays an important role in filled polymer systems. If the polymer matrix has

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high ductility, it can increase the toughness of the composite.<sup>4</sup> PP itself has poor impact properties, especially at low temperatures. The impact properties of PP can be improved through the use of block copolymers of polypropylene, addition of elastomers, or with copolymerization with ethylene. The most commonly used and most effective impact modifiers for PP are ethylene/propylene copolymers (EPM) or ethylene/propylene/diene terpolymer (EPDM).<sup>5-7</sup> Blends with 10-40% elastomers are called "impact polypropylenes"; the elastomeric phase being dispersed in the polymer matrix. Elastomer domain size and distribution also have an effect on the impact properties of PP. Uniform distribution and small domains, < 1  $\mu\text{m}$ , are expected to improve the impact properties of PP.<sup>8,9</sup> Jang et al.<sup>8</sup> studied the dependence of the impact behavior of the PP on the rubber particle size and found that small particles were more effective than larger particles for toughening PP. Dao<sup>9</sup> reported that the optimum particle size for impact improvement using EPDM rubber in the PP matrix is between 0.1-1  $\mu\text{m}$ .

#### Toughening of Filled Polymers

Many elastomers have been used as impact modifiers for filled/reinforced PP systems. The elastomeric phase improves the impact strength but reduces the E-modulus.<sup>10-14</sup> The mechanism of toughening is more complicated in a composite system than in a single-phase system because stress concentrations, interactions between components and heterogeneity, etc., provide additional complications. Generally, the role of the elastomer particles is to act as stress concentrators that initiate the local yielding of the matrix, which avoids brittle catastrophic failure of the material.<sup>4</sup> Impact modifiers can affect the filled PP morphology in three different ways: (a) the elastomer phase can exist as a third separate phase in the PP matrix, (b) the elastomer can partially or completely encapsulate the filler (form an interphase), or (c) a mixed condition of (a) and (b).<sup>10-13</sup> Stamhuis<sup>10,11</sup> used styrene/butadiene/styrene rubber (SBS), styrene-ethylene/butylene-styrene (SEBS), acrylonitrile/butadiene rubber (NBR), ethylene vinylacetate (EVA), and EPDM as impact modifiers in talc-filled PP composites. He reported improved impact properties with all additives but obtained the best results when the additives partially coated (encapsulated) the filler surfaces.

Earlier investigations have shown that the interaction between PP and cellulose-based fillers

can be improved with the addition of a maleated polypropylene (MAPP).<sup>3, 15-19</sup> Dalvåg et al.<sup>3</sup> reported improvements in tensile strength, elongation at break, and Charpy impact properties when MAPP was used as a compatibilizer in PP/WF composites. Felix and Gatenholm<sup>15</sup> and Gatenholm et al.<sup>16</sup> used MAPP in a PP/cellulose fiber (CF) system and reported increases in tensile strength and impact strength. Good adhesion between the cellulose fibers and PP matrix with MAPP was shown using scanning electron microscopy. The fracture surface of the composite showed that the failure occurred in the matrix and not in the interface between the CF and PP. Myers et al.<sup>17,18</sup> studied the influence of MAPP and extrusion temperature on PP/WF composites. MAPP had a positive effect on composite tensile strength and stiffness, but a negative effect on notched impact strength. They believed that the loss of impact strength may be caused by the increased reinforcement in the composites and also by increased wood filler brittleness at higher extrusion temperatures. Han et al.<sup>19</sup> used MAPP as a compatibilizer in different PP/wood composite systems. They reported improved mechanical properties with the addition of which depends upon the reactive size of hydroxyl groups on the filler surfaces. Cellulose kraft pulp fibers and dissolving pulp fibers had higher concentrations of hydroxyl groups on the surface and, therefore, demonstrated better mechanical properties than PP/WF composites.

Dalvåg et al.<sup>3</sup> tested several elastomer additives in cellulose filler/thermoplastic composite systems to improve the impact properties. The elastomers used were ethylene/vinylacetate (EVA), chlorinated polyethylene (CPE), polyisobutylene (PIB), ethylene/propylene thermoplastic elastomer (TPO), ionomer-modified polyethylene (syrlyn), and acrylonitrile/butadiene elastomer (NBR). They reported an impact strength for the PP/WF (30%) composite of about 27  $\text{kJ/m}^2$ , increasing to about 41  $\text{kJ/m}^2$  with the addition of 10% NBR, 30  $\text{kJ/m}^2$  with 10% PIB, and 29  $\text{kJ/m}^2$  with 10% TPO. The other additives did not improve the impact strength.

Scott et al.<sup>20</sup> reported that the addition of unmodified EPDM and maleated EPDM gave good Izod impact strength properties in calcium carbonate and oxidized silicon powder-filled polyethylene (PE) composite systems—the EPDM-MA showing superior impact properties. Oksman<sup>21</sup> used SEBS-MA in a PE/WF composite system and reported an improvement in impact properties and also a compatibilizing effect between the

wood filler and the PE. Long and Shank's<sup>12</sup> study of filled PP systems showed that SEBS-MA and EP-MA (ethylene-propylene) act not only as a toughening agent but also as compatibilizer between filler particles (talc, CaCO<sub>3</sub>, nylon-12) and the PP matrix. The maleated elastomers encapsulated the filler particles (core-shell), resulting in improved impact properties. Gupta et al.<sup>14</sup> studied glass fiber (GF)-reinforced PP/EPDM composite. They reported that GF increased impact strength, which was further increased with a high EPDM content (20–30 wt %). The reinforcing effects of GF was increased with the addition of EPDM. They explained that EPDM played a dual role in GF/PP composites. It helps maintain the alignment of the GF during tensile loading and by induces shear yielding of the matrix. Morphological study of the composites showed the fibers were coated, probably with EPDM. The GF was treated with a coupling agent compatible with PP, but the possible effects of this were not discussed.

The size and dispersion of filler particles in the matrix can effect the composites properties. Small, well-dispersed particles generally give better properties.<sup>5</sup> Small particles can block crack propagation, resulting in impact toughening. However, it is often difficult to disperse very fine particles because of their tendency to agglomerate. Particles with higher aspect ratios (e.g., wood fibers) have high stresses at the fiber ends that can cause failure under impact.<sup>22</sup> Stamhuis<sup>10</sup> found that smaller particles were coated more completely than larger ones.

An initial study was proposed to investigate the effects of elastomer-compatibilizer combinations on the morphology and mechanical properties of WF filled PP. The impact modifiers are believed to improve the PP impact strength and elongation at break but to decrease the E-modulus of the composites. Maleated impact modifiers are believed also to act as compatibilizers; MAPP may increase this effect further. Combinations of impact modifier and MAPP should result in good adhesion between the WF particles and PP matrix and increase the toughness of the PP matrix.

## MATERIALS AND METHODS

### Materials

The following commercially available materials were used:

### Matrix

Polypropylene PP, Fortilene PP 9200 (Solvay polymers), density 900 kg/m<sup>3</sup>, MFI 4 g/10 min, (230°C/2160 g).

### Impact Modifiers and Compatibilizer

Ethylene/propylene/diene terpolymer EPDM, Royalene IM-7200 (Uniroyal Chemical) density 870 kg/m<sup>3</sup>, ethylene to propylene (E/P) ratio 75/25. Maleated ethylene/propylene/diene terpolymer EPDM-MA, Royaltuf 465 (Uniroyal Chemical) total maleic anhydride/acid content of 1%, ethylene to propylene (E/P) ratio 55/45. Maleated styrene-ethylene/butylene-styrene triblock copolymer SEBS-MA, Kraton FG 1901X (Shell Chemicals), PS content 28% by weight, functionality 2% by weight as bound maleic anhydride, density 910 kg/m<sup>3</sup>. Maleated polypropylene MAPP, Unite MP880 (Aristech Chemical Corporation) average molecular weight of 90,000.

### Filler

Wood flour WF, number 402, Western pine, particle size 420 μm (nominal 40 mesh) (American Wood Fibers, Inc, Schofield, WI).

Compositions of various PP/WF composites are shown in Table I.

### Processing

The PP, WF, and additives were preblended in a Marion mixer and then compounded in a Davis-Standard (Pawcutuck, CT) corotating twin-screw extruder (screw diameter 32 mm). The barrel temperatures were: 193°C, for zones 1, 2, and 171°C for zones 3-7 and melt temperature at the die was 200°C. The screw speed was 220 rpm, pressure at the die was 4.5 MPa, and material output was 24 kg/h. The extruded strands were cooled in a water slide system, pelletized, and dried at 105°C. The compounded pellets were injection molded using a conventional Cincinnati Milacron (Batavia, OH) 33 ton reciprocating screw injection molder into standard ASTM test specimens.

### Mechanical Testing

Tensile testing of the specimens were performed according to ASTM D 638 on a MTS 810 material test machine using an model 632, 12F-20 strain gauge (MTS, Systems, Minneapolis, MN). Cross-head speed was 5 mm/min. The E-modulus and

**Table I Composition of the Various PP/WF Composites**

Sample Code	Composite Compositions for Polymers (%-by Weight), Impact Modifiers and Compatibilizer					
	PP	WF	MAPP	EPDM	EPDM-MA	SEBS-MA
1	60	40				
2	58	40	2			
3	50	40		10		
4	48	40	2	10		
5	50	40			10	
6	48	40	2		10	
7	50	40				10
8	48	40	2			10

maximum tensile strength and elongation at break were calculated from the tensile test data. At least 10 test specimens of every composition were tested.

Impact testing was performed using the ASTM D 256 Izod Impact method. Both notched and unnotched impact energies were determined for at least 10 test samples of every composition.

Conditions during testing were 23°C and 50% relative humidity.

### Scanning Electron Microscopy

The fracture surfaces from room temperature and liquid nitrogen Izod impact test specimens were examined using JEOL JSM-840 and CamScan S 4-80DV scanning electron microscopes (SEM) at an acceleration voltage of 15 kV. The elastomer particles on the fracture surfaces were revealed by etching with *n*-heptane vapour for 20 s. Several authors have used *n*-heptane as an etching medium for EPDM rubber in PP.<sup>23,24</sup> Particle size distributions were determined using an image analysis system for scanning electron microscopes (SemAfore, JEOL). All specimens were sputter coated with gold.

## RESULTS AND DISCUSSION

### Mechanical Properties

Table H summarizes the mean and standard deviation of the mechanical properties of PP/WF composites with different impact modifiers and MAPP as compatibilizer. The results are also presented in separate figures.

Figure 1 shows the tensile strength of the PP/WF composites (bar 1) compared with composites

with different impact modifiers and MAPP. The tensile strength is highest for the PP/WF composite with MAPP as a compatibilizer, but the composites with both SEBS-MA and SEBS-MA with MAPP also have increased tensile strength. The MAPP compatibilizer has a positive effect on the tensile strength in all composites. An increase in tensile strength means that the stress has been transferred from the PP matrix to the WF particle. The tensile strength of many filled polymers can be improved using adhesion promoters (compatibilizers), which improve the adhesion and the nature of the filler/matrix interface.<sup>25</sup> It is, therefore, expected that MAPP and SEBS-MA will improve the interracial bonding between WF and PP resulting in improved tensile strength. Both grades of EPDM had a negative effect on the tensile strength but were improved slightly with MAPP.

Figure 2 shows the elongation at break for modified PP/WF composites. Bar 1 is the reference PP/WF composite. The elongation at break is greater for both composites with EPDM and EPDM-MA rubber (bar 3, 5) but decreases when the MAPP compatibilizer is added (bar 4, 6). The SEBS-MA and SEBS-MA with MAPP composite systems (bar 7, 8) have the highest elongation at break. Combining the data from Figures 1 and 2 shows that the addition of MAPP makes PP/WF composites stronger but less tough, except composites with SEBS-MA.

Fillers with higher stiffness than the matrix can increase the modulus of the composites, but generally fillers cause a dramatic decrease in the elongation at break. Almost all of the elongation occurs in the matrix if the filler is rigid. If there is good adhesion between the filler and the matrix, a decrease of the elongation at break, even with

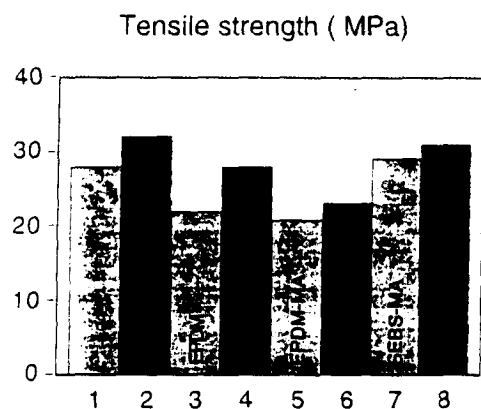
**Table II Mechanical Properties of WF/PP Composites ( $\pm$  Values are Standard Deviations)**

Sample Code	Tensile Properties		Izod Impact Properties		
	Strength (MPa)	Modulus (GPa)	Elongation at break (%)	Notched (J/m)	Unnotched (J/m)
1 —	27.9 $\pm$ 0.3	2.4 $\pm$ 0.2	3.1 $\pm$ 0.3	26.1 $\pm$ 1.1	85.8 $\pm$ 9.4
2 MAPP	32.4 $\pm$ 0.2	2.4 $\pm$ 0.1	2.6 $\pm$ 0.2	23.5 $\pm$ 1.3	86.1 $\pm$ 9.4
3 EPDM	22.0 $\pm$ 0.3	1.9 $\pm$ 0.1	3.8 $\pm$ 0.3	38.2 $\pm$ 1.2	97.3 $\pm$ 9.6
4 EPDM + MAPP	27.7 $\pm$ 0.2	1.9 $\pm$ 0.1	3.1 $\pm$ 0.2	34.4 $\pm$ 1.4	105.3 $\pm$ 10.0
5 EPDM-MA	21.0 $\pm$ 0.3	1.6 $\pm$ 0.1	3.8 $\pm$ 0.3	47.7 $\pm$ 2.2	144.6 $\pm$ 7.1
6 EPDM-MA + MAPP	22.9 $\pm$ 0.6	1.8 $\pm$ 0.2	3.6 $\pm$ 0.2	47.9 $\pm$ 2.1	146.0 $\pm$ 16.9
7 SEBS-MA	29.3 $\pm$ 0.5	1.9 $\pm$ 0.1	3.9 $\pm$ 0.2	51.2 $\pm$ 2.6	153.6 $\pm$ 16.4
8 SEBS-MA + MAPP	30.5 $\pm$ 0.5	1.9 $\pm$ 0.1	4.2 $\pm$ 0.2	54.1 $\pm$ 2.2	167.4 $\pm$ 16.7

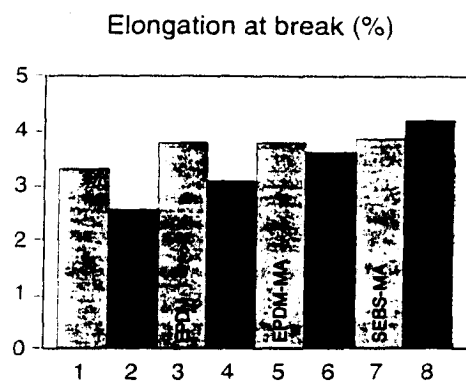
small amounts of filler, can be expected. If the adhesion is poor, the elongation at break may decrease more gradually.<sup>25</sup> This seems to agree with the results obtained. The PP/WF composite elongation at break is dramatically decreased compared to unfilled PP (590% at a crosshead speed of 2 in/min, reported by the manufacturer). The addition of MAPP decreases the elongation at break further while impact modifiers increased it. The positive effect of impact modifiers may depend on more a flexible matrix (addition of EPDM) and maybe further affected by the development of a flexible interphase around the wood particles (addition of EPDM-MA and SEBS-MA).

Figure 3 shows that the stiffness of the WF filled PP without impact modifiers is 2.4 GPa (bar 1) compared with 1.4 GPa (bar 0) for unfilled PP (reported by the manufacturer). All impact modifiers decrease the stiffness of the composites, as expected due to the low  $E$ -modulus of the elastomer. The  $E$ -modulus of elastomer-filled PP can be

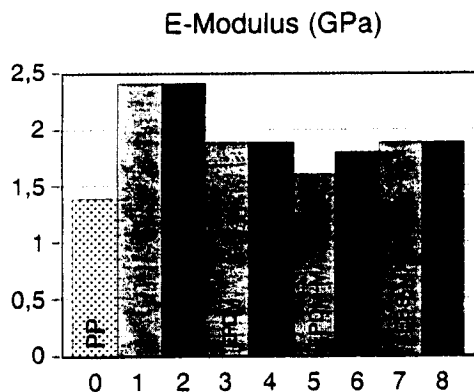
calculated using the Lewis and Nielsen equation,<sup>25</sup> as a result, a reduction of  $E$ -modulus about 19% with elastomer content of 10% can be expected. The  $E$ -modulus of the PP/WF composite (2.4 GPa) decreases with the addition of EPDM and SEBS-MA to 1.9 GPa (21%) and with EPDM-MA to 1.6 GPa (33%). The results from the addition of EPDM and SEBS-MA agree quite well with the theoretical calculations, but for the addition of EPDM-MA, they do not. If the stiffness of the composites containing EPDM and EPDM-MA are compared, it can be seen that both elastomers are very soft, but that the EPDM-MA decreased the stiffness more. Because of the greater affinity of the EPDM-MA, we can speculate that the elastomer form a interphase around the WF particles to some degree. This type of morphology has been shown to cause a greater reduction in  $E$ -modulus than a morphology where the elastomer exists as discrete domains in the matrix.<sup>11,12</sup> The composites containing SEBS-MA do not show as large reduc-



**Figure 1** Tensile strength of the composites with different impact modifiers and MAPP as a compatibilizer. Tested in the dry condition.



**Figure 2** Elongation at break of the composites with different impact modifiers and MAPP as a compatibilizer.



**Figure 3** Stiffness of the composites, pure PP compared with composites with different additives.

tions as composites containing EPDM-MA, possibly as a result of the higher modulus of SEBS-MA. However, because of the complexity of the interphase, further study is necessary to fully explain the composite properties.

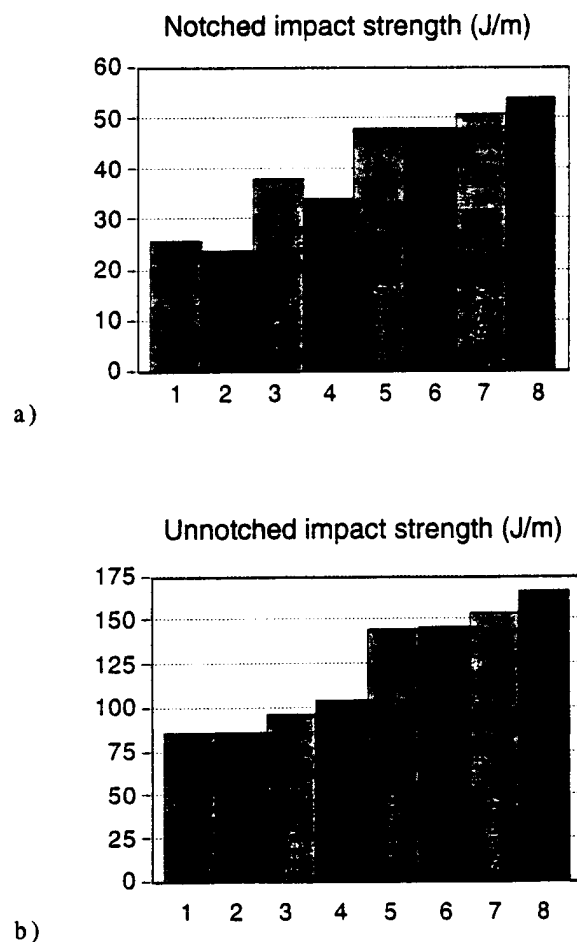
A better balance of composite properties may be achieved at lower concentrations of the impact modifier, depending upon the required product performances, for example, higher E-modulus while retaining good impact and tensile strength. Oksman<sup>21</sup> reported that the addition of 5 wt % of SEBS-MA to PE/WF composites improved the E-modulus by almost 10%, the tensile strength 28%, and impact strength 58%.

Figure 4 shows the impact strength of each composition. Improvements in impact strength were seen in all impact-modified systems. The addition of EPDM-MA resulted in greater improvements in impact performance but a lower E-modulus when compared to composite systems containing unmodified EPDM. Stamhuis<sup>10,11</sup> noted similar findings in talc-filled PP systems using SBS as an impact modifier. Impact modifiers with an affinity for the particles partially encapsulated or coated the talc particles rather than simply existing as separate domains in the bulk matrix. This encapsulation reduces stress concentrations at the particle-polymer interface, leading to better impact performance, but also resulting in lower E-moduli. The composite systems containing SEBS-MA gave the highest impact energies. The MAPP compatibilizer had little or no effect on the notched impact strength but seems to increase the unnotched impact strength of the composites when used together with elastomers. Myers et al.<sup>17,18</sup> reported that the MAPP did not improve the impact strength of PP/WF composites but instead reduced it with increasing MAPP content. Dalvåg et al.<sup>3</sup> reported increased impact

strength using MAPP in PP/WF composites. The addition of 2% MAPP increased the unnotched impact strength about 30%.

A summary of the effects of elastomers and MAPP on the WF filled PP is shown in Table III ("+" is positive, "-" negative, and "O" is no effect).

Table III shows that both composites containing SEBS-MA gave the best properties. The notched Izod impact strength of PP/WF/SEBS-MA + MAPP composites were increased about 100% and the unnotched impact strength 95%, the elongation at break was increased by 35%, and tensile strength was improved 9% over the PP/WF composite. The E-modulus decreased 35% compared to PP/WF composites (see Table II) but was still 35% greater than unfilled PP. The tensile strengths was increased in composites with the addition of MAPP or SEBS-MA. The reasons for the absence of this behavior in composites with EPDM and EPDM-MA with and without MAPP



**Figure 4** Impact strength of notched and unnotched samples of different blends.

**Table III Summary of the Effects of Additives on the PP/WF Composites**

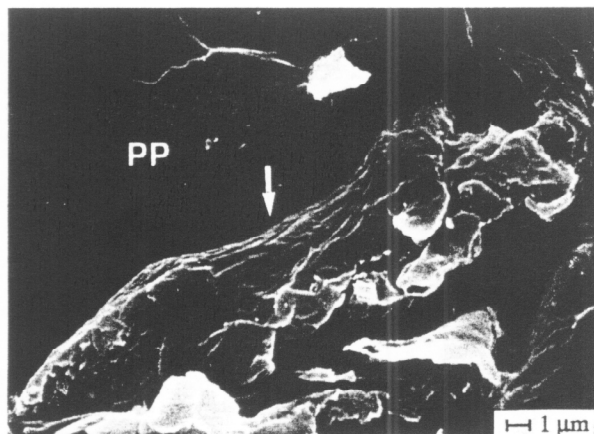
Composite Code	$\sigma_{\max}$	E	$\epsilon$	Izod Notched	Izod Unnotched
2 MAPP	+	○	-	-	○
3 EPDM	-	-	+	+	○
4 EPDM + MAPP	○	-	○	+	+
5 EPDM-MA	-	-	+	+	+
6 EPDM-MA + MAPP	-	-	+	+	+
7 SEBS-MA	+	-	+	+	+
8 SEBS-MA + MAPP	+	-	+	+	+

Comparisons made using Student's *t*-test, 5% significance level. "+" positive, "-" negative and "○" no or little effect.

are difficult to explain. It is possible that EPDM and EPDM-MA do not form an interphase strong enough for stress transfer from the matrix to the filler to take place.

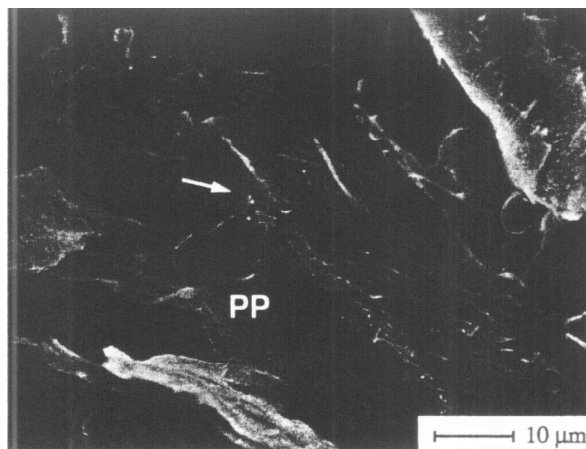
### Morphology

Examination of the fracture surfaces of the composites by scanning electron microscope gave information about how impact modifiers and MAPP affect the morphology of the composite. The rubber particle sizes and the interracial region between the PP matrix and the wood filler were investigated. Figure 5 shows the microstructure of the composite without impact modifier and compatibilizer (blend 1) showing a wood particle embedded in the polymer matrix. The wood particle is not broken and there are voids around the particle indicating poor interaction between the wood surface and the PP matrix. The WF particles were well dispersed on the PP matrix.

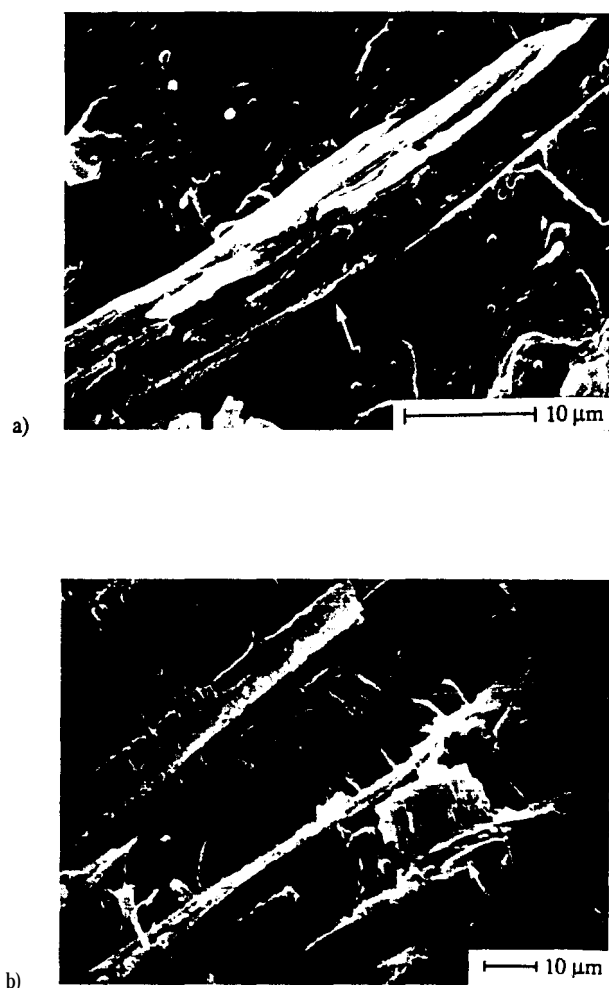


**Figure 5** SEM micrograph of room-temperature fractured specimen. Interface/interphase region between the wood filler and the PP matrix.

Figure 6 shows microstructure of the composite with MAPP as compatibilizer (blend 2). The microstructure is different to that in Figure 5. Generally, it is more difficult to differentiate wood particles from the PP matrix. This may suggest that the wood particles are coated, probably by the matrix, and that the failure most commonly occurs in the matrix and not at the filler surfaces. There are places where the adhesion between the PP and the WF is good, as broken wood particles can be seen with no gap between the PP matrix and WF surfaces. When adhesion is not so good, there are voids around the wood particles and places where WF particles have pulled out. Good adhesion between the filler and the matrix was expected because composite 2 had improved tensile strength, which means that loads can be transferred from the matrix to the filler. This suggests that there is some kind of interracial contact between the WF and the PP. The majority of the studies of compatibilizing using MAPP were on the PP/CF composite system. Other authors have



**Figure 6** SEM micrograph of room-temperature fractured specimens. Composite with MAPP as a compatibilizer.



**Figure 7** SEM micrograph of fractured specimen. (a) PP/WF composite with EPDM, (b) PP/WF with EPDM and MAPP.

shown<sup>3,15-19</sup> that MAPP has a compatibilizing effect on PP/CF and PP/WF composites. The composite system in Figure 6 showed the highest stiffness and tensile strength, which suggest improved adhesion between the WF filler and the PP matrix, but the lowest impact properties and elongation at break, which signify embrittlement of the filler or the matrix. The MAPP may also act as a dispersing agent between polar fillers and unpolar matrix, resulting in better dispersion of the filler.<sup>19</sup> In this investigation a good dispersion of WF particles in PP matrix was found. Differences in particle dispersion between the different composite microstructure with addition of MAPP were not observed.

Figure 7 shows micrographs the PP/WF composite with (a) EPDM and (b) with both EPDM and MAPP. Figure 7(a), there is poor adhesion between the PP matrix and the WF particle sur-

face, as there are voids around the wood particle. In Figure 7(b), better adhesion between WF particle and PP matrix can be seen. Fracture paths often passed through the wood particles (broken WF particles are visible), which indicates good adhesion between the WF filler and the PP matrix and also that the interphase is stronger than the WF particle. Tensile strength is improved in the latter case from 22 to the 27.7 MPa, which suggests improved adhesion. The E-modulus is unaffected, but elongation at break and the notched impact strength are decreased. It could be expected that the better interface is only formed in the composite containing the MAPP, which explains the decreases in elongation at break and impact strength. The improved impact strength compared to composites without elastomer additives can be explained by better toughness of the matrix itself. In both composites, the EPDM elastomer is uniformly distributed in the PP matrix and has a domain size between 0.1 – 1 μm.

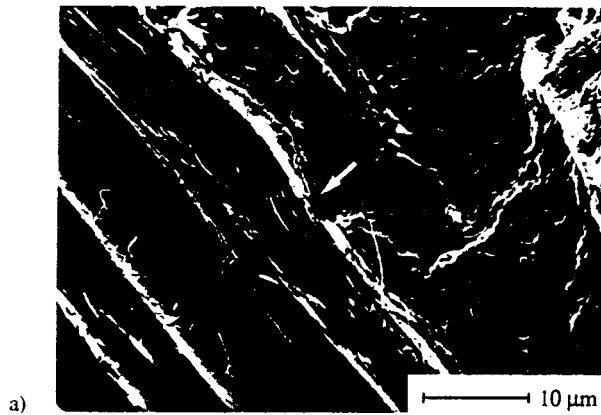
Figure 8 shows the morphology of the PP/WF composite with (a) EPDM-MA and (b) EPDM -MA with MAPP. There is good adhesion between the PP matrix and the wood particle surfaces in both composites; fracture paths have passed through the wood particles in both cases, which indicates good adhesion. The adhesion between the rubber particles and PP matrix is improved when compared to composites containing unmodified EPDM. As far as the mechanical properties are concerned, the tensile strengths and E-modulus are the lowest of all composites but elongation at break and impact strength are better. The micrographs and mechanical properties indicate that the EPDM-MA forms an interphase that effectively increases the impact strength and decreases the E-modulus. but that the interphase is too soft for stress transfer from the matrix to the filler. It is difficult to see the rubber particles because of the good interaction between the matrix and EPDM-MA, which causes the failure in the PP matrix. The EPDM elastomer particles are uniformly distributed in the PP matrix and have a size between 0.1 to 1 μm.

Figure 9 shows the morphology of the composite with (a) SEBS-MA and (b) the same with MAPP. There is good adhesion between the wood particle and PP matrix in both micrographs. No voids around the wood particle surfaces are present, and the fracture paths have often passed through the wood particles. Almost all mechanical properties were improved compared to composites with EPDM and EPDM-MA elastomers. The tensile strength of (a) was 29.3 MPa, which is second



highest after PP/WF with MAPP and is further increased to 30.5 MPa when MAPP was added. Elongation at break and impact strength were the highest of all composites tested and the E-modulus was the same as the composites with EPDM. It is difficult to measure the particle size of the elastomer phase because of the good adhesion between the matrix and the SEBS-MA.

Figure 10 shows EPDM and EPDM-MA elastomer particles in the PP matrix. The fracture surfaces have been etched with *n*-heptane vapor; the holes in these micrographs represent the EPDM particle size that was between 0.1 and 1  $\mu\text{m}$  with an average of 0.3  $\mu\text{m}$ . A more accurate determination of the particle size distribution from the fracture surfaces is very difficult, especially if there is strong interracial adhesion be-

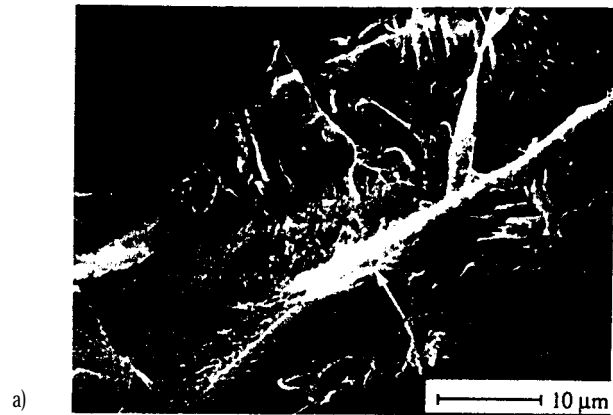


a)

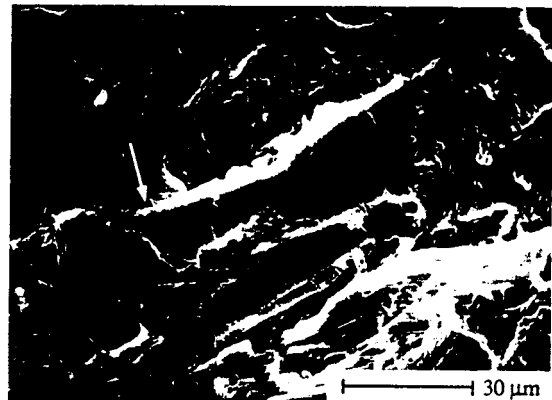


b)

**Figure 8** SEM micrograph of fractured specimens. (a) PP/WF composites with EPDM-MA as a impact modifier, and (b) same composite but with MAPP added as a compatibilizer.



a)



b)

**Figure 9** (a) PP/WF composite with SEBS-MA as impact modifier, and (b) the same blend when MAPP is added as a compatibilizer.

tween the matrix and the elastomer. The fracture propagates through the bulk matrix and the elastomer particles do not pull out. The quality of the EPDM-MA particle size micrographs can be due to particle/matrix adhesion and etching conditions.

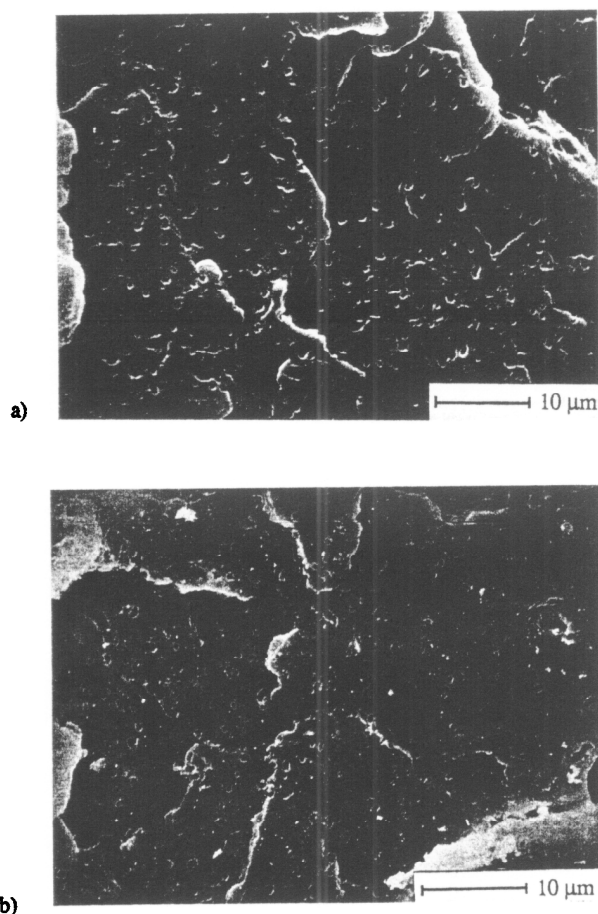
Figure 11 shows schematic representations of different microstructures showing unmodified PP/WF composite and different modifications. In Figure 11( a) the unmodified PP filled with WF shows poor adhesion between the WF and PP surfaces, and there are voids around the WF particle. In Figure 11(b) the composite with MAPP as a compatibilizer, the MAPP is expected to enhance interracial adhesion between the WF and PP. Figure 11(c) shows the microstructure for the composite with EPDM elastomer. There is poor adhesion between the WF particle and the PP matrix. When MAPP is added as a compatibilizer [ Fig. 11(d) ], the interracial adhesion between the WF particle and the PP matrix is improved. The MAPP improves interracial adhesion but does not

form a soft interphase around the WF particle. Figure 11(e) EPDM-MA shows good adhesion with WF particles and is expected to form a soft interphase around them. A similar morphology is found when MAPP is added. Figure 11(f) shows the microstructure when SEBS-MA is added. Good adhesion between the surfaces is expected, and the SEBS-MA forms a strong interphase between the WF and PP. When MAPP is added, the adhesion is even stronger.

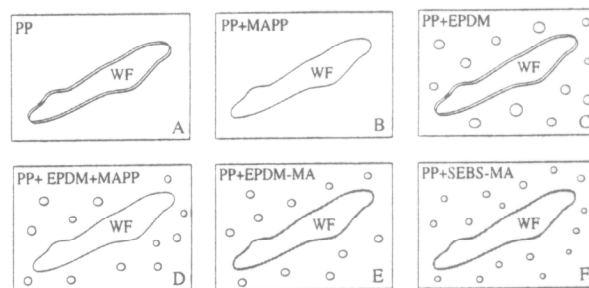
## CONCLUSIONS

The objective of this study was to investigate the effects of different combinations of impact modifiers and MAPP as compatibilizer on the mechanical properties and morphology of WF-filled PP.

The E-modulus was improved by 70% when WF was added to pure PP but reductions were found



**Figure 10** Elastomer particles in PP matrix. (a) EPDM and (b) EPDM-MA. Fractured surfaces etched with *n*-heptane vapor for 20 s.



**Figure 11** Schematic representations of modified and WF filled PP. (a) and (c) shows no adhesion between WF and PP. (b) Interfacial adhesion; (d)–(f) an expected formation of interphase around the WF particle.

when elastomers were added to these composite systems.

Addition of MAPP did not affect the notched impact strength but improved the unnotched impact strength when added with elastomers. MAPP had a positive effect on the tensile strength forming an interphase between the WF and PP, but a negative effect on the elongation at break. MAPP generally had a positive effect on the mechanical properties when it used together with SEBS-MA and EPDM.

EPDM increased toughness, elongation at break and impact strength were improved compared to unmodified the composites.

The SEBS-MA and the EPDM-MA both have higher affinity for wood flour than EPDM, and gave the largest increases in impact performance. The results indicate that SEBS-MA and EPDM-MA act both as compatibilizer and impact modifier, resulting in better interracial adhesion, better impact strength, and higher elongation at break. However, the lower E-modulus and a inability to transfer stress born the matrix to the WF particle confirm that the EPDM-MA interphase between the WF and PP is softer than the interphase that SEBS-MA forms.

Composites containing both SEBS-MA and MAPP show 107% better notched impact strength compared to PP/WF composites, but the stiffness was reduced by 35% compared to unmodified composites. The addition of both EPDM-MA and MAPP to PP/WF composites increased the notched impact strength by 83%, but decreased the composites stiffness by 28%.

Scanning electron microscopy showed that maleated EPDM and SEBS had a stronger affinity for the wood surfaces than did the unmodified EPDM, and that the MAPP further improved the adhesion between WF and PP.

The particle size distributions were between

0.1 to 1  $\mu\text{m}$  for both EPDM and EPDM-MA. The maleated EPDM particle sizes were more difficult to measure because of good adhesion between the impact modifier and the PP matrix.

These results can be summarized as follows: Maleated elastomers make effective impact modifiers in the PP/WF composite systems and the addition of MAPP compatibilizer has a positive effect on composite stiffness, tensile strength, and unnotched impact strength. Morphological study showed that the MAPP-enhanced adhesion between the WF and the impact-modified PP systems. Composites with SEBS-MA showed the greatest impact strength, elongation at break, and tensile strength compared to composite systems with EPDM and EPDM-MA. SEBS-MA shows the best potential to be used as an impact modifier in the PP/WF composite system, and the use of MAPP can further increase mechanical performance.

This work was carried out during K. Oksman's stay as a visiting scientist at the USDA Forest Service, Forest Products Laboratory in Madison, WI.

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VOLUME 67 / NUMBER 9 / FEBRUARY 28, 1998

*C O N T E N T S*

- Mechanical Properties and Morphology of Impact Modified Polypropylene–Wood Flour Composites**  
*K. Oksman and C. Clemens* 1503
- Use of Chitosan to Improve Dyeability of DP-Finished Cotton (II)**  
*Y. Shin and D. I. Yoo* 1515
- Amphiphilic Beechwood Glucuronoxylan Derivatives**  
*A. Ebringerová, I. Sroková, P. Talába, M. Kačuráková, and Z. Hromádková* 1523
- Morphology and Mechanical Properties of Nylon 6 Toughened with Waste Poly (vinyl butyral) Film**  
*Y.-J. Cha, C.-H. Lee, and S. Choe* 1531
- Fracture Energy Release Rate in Nylon Fibers**  
*S. Michielsen* 1541
- A GC-MS Study of the Addition Reaction of Aryl Amines with Acrylic Monomers**  
*M. Farahani, J. M. Antonucci, and L. R. Karam* 1545
- Simulations of Primary and Secondary Gas Penetration for a Gas-Assisted Injection-Molded Thin Part with Gas Channel**  
*S. C. Chen, N.-T. Cheng, and S.-Y. Hu* 1553
- Impact Behavior of Glass-Epoxy Composites Containing Foam Material**  
*Kishore and S. Venkatraman* 1565
- Modeling of Industrial Nylon-6,6 Polycondensation Process in a Twin-Screw Extruder Reactor. I. Phenomenological Model and Parameter Adjusting**  
*R. Giudici, C. A. Oller Do Nascimento, I. C. Beiler, and N. Scherbakoff* 1573
- Reaction Rate Enhancement During Swollen-State Polymerization of Poly (ethylene terephthalate)**  
*M. K. Parashar, R. P. Gupta, A. Jain, and U. S. Agarwal* 1589
- Differential Scanning Calorimetry and Wide-Angle X-ray Scattering Studies of Bread Staling**  
*J. H. Jagannath, K. S. Jayaraman, S. S. Arya, and R. Somashekar* 1597