# Influence of Type and Concentration of Maleic Anhydride Grafted Polypropylene and Impact Modifiers on Mechanical Properties of PP/Wood Sawdust Composites

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**ABSTRACT:** Surface treatments of natural fibers in polymer-wood composites (PWC) are still one of the most interesting subjects for research among technologists, especially with inclusion of other property modifiers in the composites. This work investigated the effects of incorporating different types and contents of maleic anhydride grafted polypropylene (MAPP) and impact modifiers, as property promoters, on the mechanical properties of PP/wood sawdust composites. The concentration of wood sawdust was also varied. The experimental results revealed that increasing the wood fiber into the PP matrix reduced the overall strength and toughness of the composites. It was observed that the higher the impact modifier concentration, the greater the improvement in toughness of the PP/wood composites, and therefore an impact modifier with high comonomer content and high melt flow rate is recommended. This work recommends a MAPP concentration of 2.0 wt % (of sawdust) to be introduced into the PP/wood composites containing 11.1 wt % selected impact modifier, for optimization of the overall composite mechanical strength. The recommended MAPP coupling agent should possess a high wt % MAH-grafting level and high molecular weight of PP copolymer. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 97: 475–484, 2005

**Key words:** polypropylene (PP); surface treatments; reinforcement; impact modifiers; mechanical properties

## INTRODUCTION

Composite products manufactured from blends of thermoplastics and natural fibers have increasingly attracted a number of researchers and manufacturers because of cost savings, good mechanical properties, better dimensional stability, and environmental issues. Various thermoplastics, including polyethylene,[1,2] polypropylene (PP),[3–5] and poly(vinyl chloride) (PVC),[6,7] have been used for research and development, both on laboratory and industrial scales. Examples of polymer-wood composite products include decking, window and door profiles, automobile paneling, panel inserts, flower pots, and so on, mostly being manufactured through extrusion and injection molding processes.[8]

Since polymers and natural fibers are incompatible, some surface modifications, usually by the use of coupling agents, are required for the improvement of the service performances of the composite products.[5,7] Surface treatments of natural fibers in polymer-natural fiber composites are very essential for research, especially in the case of nonpolar and hydrophobic polymers (polypropylene and polyethylene).[2,9–14] Thwe and Liao[9] used maleic anhydride polypropylene (MAPP) as a coupling agent in bamboo fiber-PP composites and found that the tensile strength and stiffness of the composites greatly increased due to improved interfacial bonding between the polymer and the fiber. Similar behavior was also observed by Joseph and coworkers,[10] who introduced various chemical treatments including MAPP to sisal fibers composited with polypropylene. The changes in mechanical properties of the polymer-natural fiber composites are usually explained in terms of adhesion mechanisms at the interface, the mechanisms including physical interactions (interdiffusion, adsorption-wettability, and molecular entanglement) and chemical bonding.[2] Biagiotti and colleagues[11] examined the effects of various coupling agents, such as maleic anhydride (MA), Maleic anhydride grafted polypropylene (MAPP), and vinyltrimethoxy silane, on the properties of flax fiber-polypropylene composites. They recommended that MAPP produced the best surface energy between the polymer and the fiber. Mohanty and coworkers[12] used MAPP as a coupling agent in sisal-polypropylene composites at different fiber loadings

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**TABLE I**
**The Chemical Structures and Descriptions of Impact Modifiers Used**

| Item | Type of impact modifier | | |
|---|---|---|---|
| | Engage 8480 | Engage 8407 | Elvaloy 1820AC |
| Co-monomer content, wt % | 20 | 40 | 20 |
| Melt index, 190°C/2.16 kg, dg/min | 1 | 30 | 8 |
| Density, g/cm$^3$ | 0.902 | 0.870 | 0.942 |
| Vicat softening point, °C | 86 | 41 | 54 |
| DSC melting point, 10°C/min rate, °C | 100 | 60 | 92 |
| Tensile modulus, MPa | 34 | 3 | 35 |
| Ultimate tensile strength, MPa | 26 | 3 | 11 |
| Elongation at break (%) | 630 | 1010 | 780 |
| Chemical structure | —[$CH_2$—$CH_2$]$_m$—[$CH_2$—CH]$_n$— | $C_6H_{13}$ | [—$CH_2$—$CH_2$]$_m$—[$CH_2$—CH]$_n$— $COOCH_3$ |

and MAPP concentrations, and suggested that for composites with 21 volume percent of sisal fiber, a MAPP of 1% gave an optimum mechanical strength with decreased damping properties.

Although the use of coupling agents improves the stiffness and strength of the thermoplastic-natural fiber composites, the toughness of the composites tends to exhibit the opposite behavior. One of the conventional methods to regain composite toughness is the use of elastomeric additives; however, this method lowers the strength of the composite. Rana and colleagues[13] examined the effects of different impact modifiers in short jute-fiber reinforced polypropylene composites and found that the addition of the impact modifiers decreased the tensile and flexural modulus, but enhanced the impact strength. They also suggested that olefinic-based impact modifiers were more effective than elastomeric-based impact modifiers. Hristov and coworkers[14] investigated the mechanical properties of polypropylene/wood fiber composites modified with maleated polypropylene as a compatibilizer and styrene-butadiene rubber (SBR) as an impact modifier. They found that the tensile modulus, yield strength, and impact strength increased by the addition of maleated polypropylene and SBR. They also suggested that if good interfacial adhesion between the polymer and the fiber was obtained, the addition of SBR would not reduce the tensile modulus and yield strength of the composites.

As stated earlier, the surface treatments of natural fibers in polymer-natural fiber composites are still one of the most interesting subjects for research and discussion, especially with the presence of other property modifiers in the composites. In this article, the effects of incorporating both coupling agents and impact modifiers, as property promoters, into PP/sawdust composites were studied and reported, not only to complement information in the literature, but also to improve the interfacial bonding between the PP and the fibers, and thus enhance the mechanical performance. By varying the types and concentrations of the property promoters, the effects of the molecular structure of the coupling agents and impact modifiers on the mechanical and morphological properties of the PP-wood composites can be discussed.

## EXPERIMENTAL

### Raw materials

- Polypropylene: Supplied by Thai Petrochemical Industry Co., Ltd (Bangkok, Thailand), in the form of pellets. The as-received MFI is 11 g/10 min.
- Wood sawdust particles: Obtained from carpentry and wood-working processes and supplied by V. P. Plastics Products (1993) Co., Ltd (Bangkok Thailand). The average size of the sawdust particles used in this work was in the range of 100–300 $\mu$m. At the initial stage of the work, the contents of wood sawdust particles added into the PP were ranged from 0.0 to 30.0 wt %. The selected content of the sawdust particles, giving the optimum mechanical properties, was then used for further investigations (effects of additions of coupling agents and impact modifiers).
- Impact modifiers: Three impact modifiers were utilized in this work, these being Ethylene Octene copolymer (Engage polyolefin elastomer) 8480, 8407, and Ethylene methyl acrylate (Elvaloy®) 1820 AC by DuPont. All impact modifiers were supplied in granule form. The details of the materials are given in Table I. By using these impact modifiers in varying concentrations, the influence of the chemical structure of the impact modifiers on the properties of PP/sawdust composites could be monitored.
- Chemical coupling agents: Three maleic anhydride grafted polypropylene coupling agents were used in this work, namely; MZ203D,

**TABLE II**
**The Property Descriptions of MAPP Coupling Agents Used**

| Item | Type of coupling agent | | |
|---|---|---|---|
| | MZ 203 D | MD 353 D | MD 411 D |
| Base resin | PP impact copolymer | Random copolymer PP | Random copolymer PP |
| Density, g/cm$^3$ | 0.94 | 0.90 | 0.90 |
| MAPP Graft Level, wt % | 0.85 | 1–1.4 | 1–1.4 |
| Melt flow rate, g/10 min | 102 | 450 | 290 |
| Melting temperature, °C | 160 | 136 | 136 |

MD353D, and MD411D, which were supplied by DuPont. All coupling agents were supplied in granule form. The details of all coupling agents are given in Table II. By using these coupling agents in varying concentrations, the influence of the chemical structure of the coupling agents on the properties of PP/sawdust composites could be discussed.

### Blending of PP, wood sawdust, coupling agents, and impact modifiers

To minimize the moisture content, the wood sawdust particles were carefully dried prior to use through heat treatment in an oven at 80°C for 24h until the sawdust weight was constant (< 5% moisture content in wood sawdust in this work). PP pellets were dry-blended with desired amounts of sawdust particles, coupling agents, and impact modifiers using a high speed mixer for 2 min before being melt-blended in a twin screw extruder (Haake Polylab-Rheomex CTW 100P, Germany). The blending temperature profiles on the extruder were 170, 180, 190, and 200°C from hopper to die zones. The screw rotating speed was 50 rpm. The head of the extruder contained a three-strand die (each strand having a diameter of 3mm), producing three extrudates at the die exit. The extrudates were then passed through a water bath via the use of a pull-off unit before undergoing a pelletizer to produce composite granules, and then held in an oven for 24 h at 80°C.

The composite extrudates produced from the twin screw extruder were specimen-shaped using an injection molding machine (E-lite E-80, Italy), the size and shape of the specimens depending on the tests as will be indicated in the characterization section. The barrel temperature profiles on the injection molding machine were 180, 190, and 200°C from hopper to die zones. The screw rotating speed used was 160 rpm, and the injection speed and pressure were 40 cm$^3$/s and 850 kg/cm$^2$, respectively.

### Characterizations

#### Mechanical properties

The mechanical properties of the PP/sawdust composites were assessed via tensile, flexural, and impact properties, and hardness. All the mechanical property results reported in this work were averaged from ten determinations.

- The tensile tests were performed on a SHIMADZU tensile tester (Tokyo, Japan) at a crosshead speed of 50 mm.min$^{-1}$, following test procedure BS 2782: Part 3 (1996) Methods 320A.
- The flexural property was determined according to ASTM D790 (1990) (specimen dimensions of 6.4 $\times$ 120 mm$^2$, support span of 90 mm, and a crosshead speed of 2.8 mm.min$^{-1}$).
- Izod impact tests, following ASTM D256 (1990), were performed on a YASUDA Impact Tester (Osaka, Japan) with the notched side facing the pendulum.
- The hardness of the composites was evaluated using a Durometer Shore D (Model 473) by PTC® Instruments Co., Ltd. The test procedure followed was as specified by ASTM D 2240 (1990).

#### SEM investigations

Failure mechanisms were investigated using a JEOL (JSM-6301F) SEM machine at 20 kV accelerating voltage. The composite fracture surfaces for examination were obtained after 2-min immersion in liquid nitrogen. The details of the experimental procedure and sample preparations were discussed elsewhere.[7]

## RESULTS AND DISCUSSION

### Strength and toughness of PP-wood composites (untreated and unmodified)

Figures 1 and 2 show the tensile modulus and elongation at break, and the ultimate tensile strength (UTS) of PP/wood composites containing different wood sawdust contents, respectively. It can be clearly observed in Figure 1 that increasing the wood sawdust content increased the composite modulus, but decreased the values of elongation at break and UTS. This indicated that the overall strength **(**Fig. 2**)** of the PP/wood composites reduced with the presence of wood sawdust fiber. Similar behavior was also found by Colom and colleagues.[2] The changes in the tensile

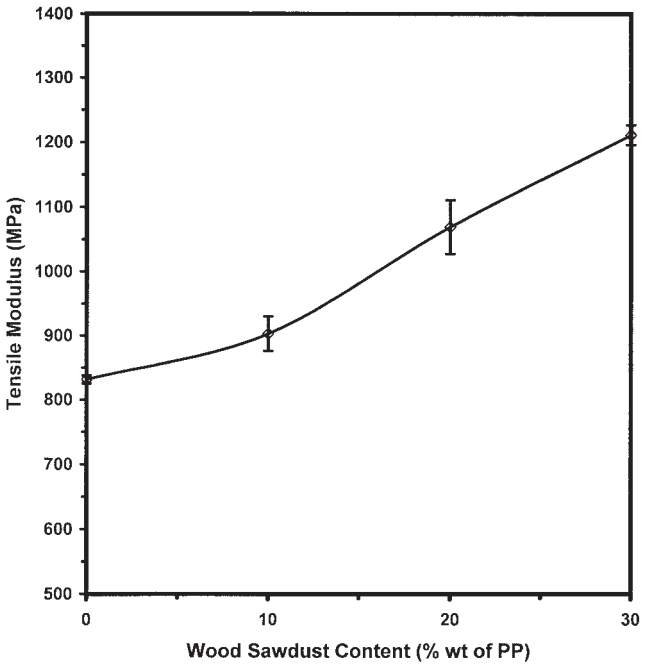



**Figure 1** Effect of sawdust content on tensile modulus for PP/sawdust composites.

properties of the PP-sawdust composites, due to the addition of sawdust, can be considered as follows.

- Modulus of the composites: The tensile modulus progressively increased with wood sawdust content, probably caused by the fact that the wood sawdust is more rigid than the polymer.

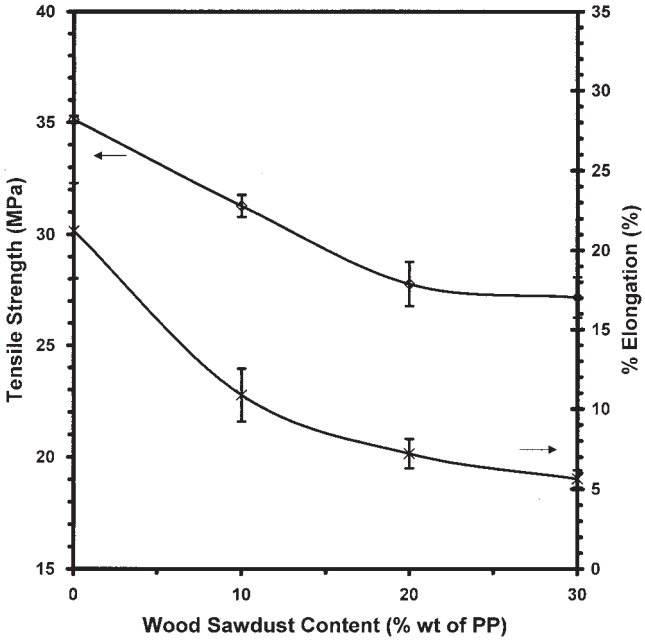



**Figure 2** Effect of sawdust content on tensile strength and elongation at break for PP/wood sawdust composites.

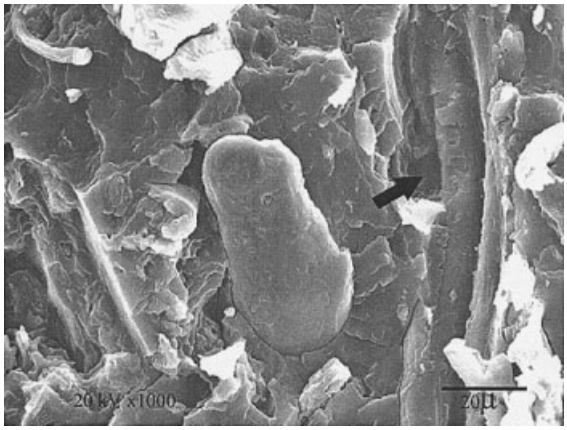


**Figure 3** SEM micrographs for PP/wood sawdust composites containing 30 wt % wood sawdust.

- Strength of the composites: The decreases in elongation at break and tensile strength were probably caused by a number of reasons including: (i) poor dispersion of the fibers in the matrix, (ii) moisture pick-up, (iii) increases of interfacial defects or debonding between PP and wood sawdust fibers due to use of untreated fibers. As for the first reason, the sawdust fibers tended to cling together, due to strong interfiber hydrogen bonding, and resisted dispersion of the individual fiber as the fiber content was increased. In the second case, since the wood sawdust is hydrophilic in nature and was chemically untreated (in this part of the work), the fiber may have picked up moisture during storage, processing, and testing. It was thought in this work that the moisture may have interfered with the adsorption effects by reducing the effect of physical bonding and potentially acting as a lubricant between the fiber surface and PP phases. Information on the mechanism of the interfering moisture in the polymer-wood composites can be obtained elsewhere.[15] The last reason for interfacial defects and debonding between rubber and fibers can be proven using a SEM micrograph, which is shown in Figure 3. A clean fiber surface is seen from the fractured surface, which indicates no interfacial interaction between the polymer and the fiber (see the arrow).
- Figure 4 shows the impact strength and hardness of PP/wood composites containing different sawdust amounts. It can be seen that the presence of wood sawdust fiber greatly reduced the toughness of the PP/wood composites with an increase in the composite hardness, although the effect became less pronounced as the wood content was increased. Work by Karger-Kociss[16] suggested that the toughness of a PP-fiber composite can be increased by incorporating long glass fiber into polypropylene. However, this was not the case for the wood sawdust particles used in this work, the

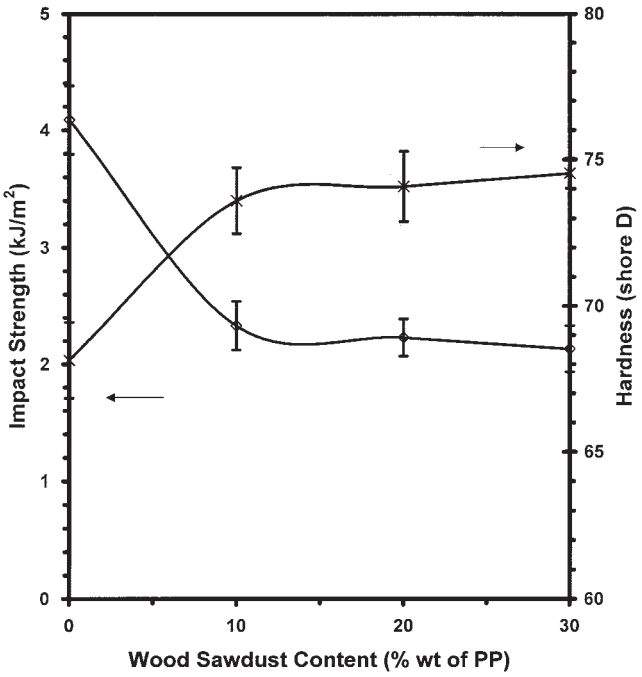



**Figure 4** Plots of impact strength and hardness against wood sawdust content for PP/wood sawdust composites.

sawdust being more like particulate filler. The decrease in impact strength of PP by inclusion of wood sawdust particles could have two possible explanations, one being the presence of wood particle agglomerates and the other being a dilution effect in which the PP matrix was interfaced by rigid wood sawdust particles.[7,15] The former explanation is also given in the work of Levita and colleagues.[17]

### Improvements of strength and toughness of the composites

The mechanical property results discussed in Figures 2 and 4 clearly indicate that introducing the wood sawdust fiber resulted in decreases in strength and toughness of the PP/wood composites. Therefore, this part of the work aimed to improve the strength and the toughness of the composites, the former being achieved by using coupling agents and the latter being obtained by including impact modifiers. After having considered the magnitude of the changes in the strength and toughness of the composites as a result of varying wood sawdust content, and considering economic benefits, 30 wt % wood sawdust was selected for further investigation.

#### Effects of types and contents of impact modifiers

Figure 5 shows the effects of types and concentrations of impact modifiers on the tensile modulus of PP/sawdust composites. In general, it can be seen that the tensile modulus of the composites for all impact modifiers decreased to a minimum value around an impact modifier content of 2.0–11.1 wt % of PP. After that, the tensile modulus tended to stabilize. The decrease in the modulus of the composite was expected due to the presence of elastomeric impact modifiers, which caused a reduction of the composite stiffness. Similar behavior was found for the tensile strength of the composites, and results are shown in Figure 6. The decrease in tensile strength clearly indicated the absence of interfacial bonding between PP molecules and wood sawdust by the presence of the impact modifiers. This statement can be supported by Sui and coworkers,[18] who reported that *if* the presence of a rubbery phase (or impact modifier) has formed an interfacial bonding between a polymer and glass-fiber, the strength of the composite would greatly increase. However, this was not the case for the impact modifiers used in this paper. This would lead us to focus on improving the interfacial bonding between the PP matrix and the sawdust particles by introducing coupling agents into the composites, which is discussed in the next section. It should also be noted from Figures 5 and 6 that the type of impact modifier had a very small effect on the changes in tensile modulus and tensile strength of the PP/wood composites.

The toughness of the composites was observed to greatly improve as required by increasing the impact modifiers, these results being shown by values of elongation at break (Fig. 6) and the impact strength (Fig. 7).

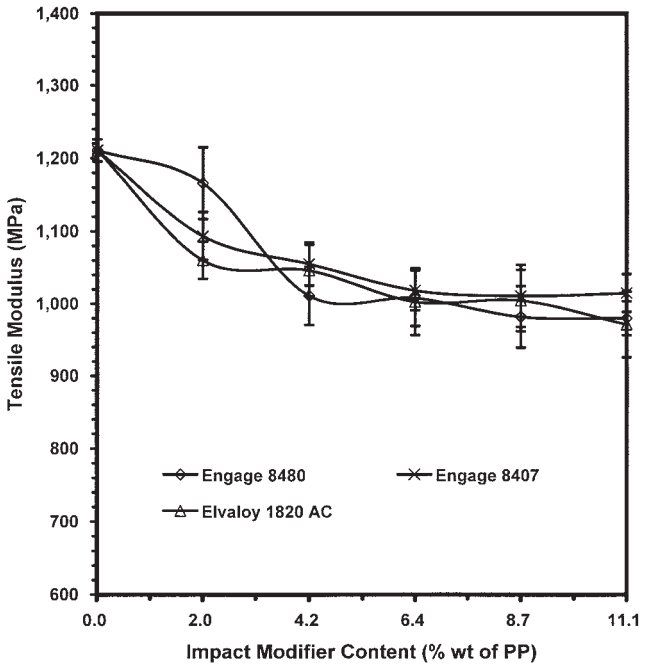



**Figure 5** Influence of impact modifier type and content on tensile modulus of PP/wood sawdust composites.

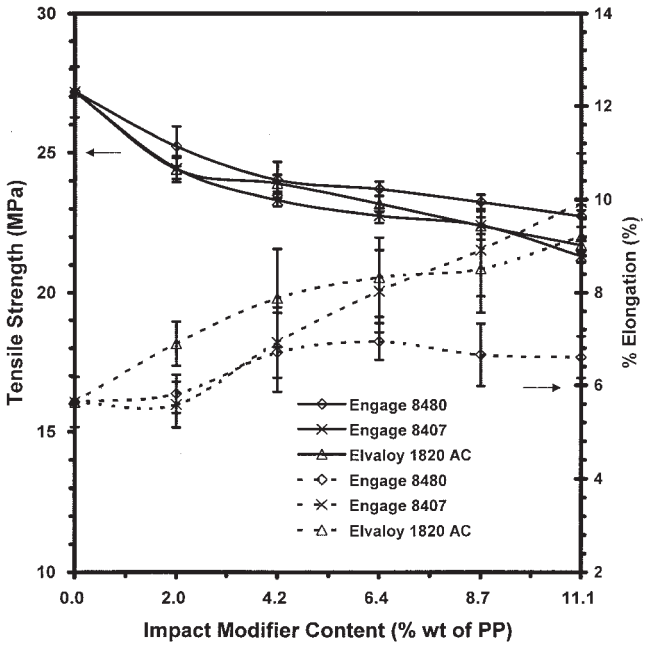



**Figure 6** Effect of impact modifier type and content on tensile strength and elongation at break for PP/wood sawdust composites.

The toughening mechanism of PP/wood composites by adding the elastomeric impact modifiers was caused by the improved ability of the rubber phase to deform plastically and triaxially.[14] The triaxial stresses built in the rubber phases would then relieve to the matrix phase, and facilitate local shear yielding in the matrix phase, thus increasing the toughness of the composite. This view was also suggested by Lovell.[19] This explanation can be substantiated by considering the SEM micrographs in Figure 8, which show the fracture surfaces of the PP/wood sawdust composites obtained from the impact tests. It can be seen that the composites with high impact strength (11.1 wt % impact modifier of Engage8407) exhibited elongated PP matrix on the fracture surface and along the fiber, which indicates a ductile deformation, while this did not occur at the composite with low impact strength (2.0 wt % Engage8407).

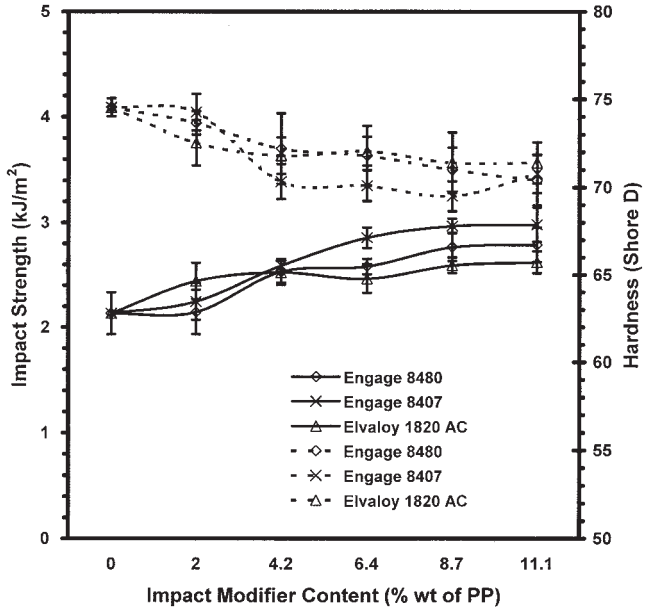
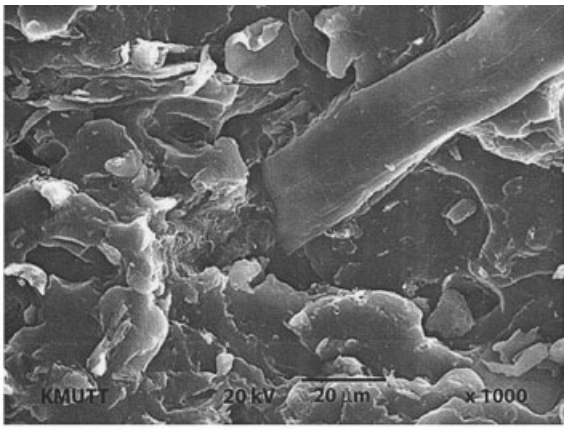
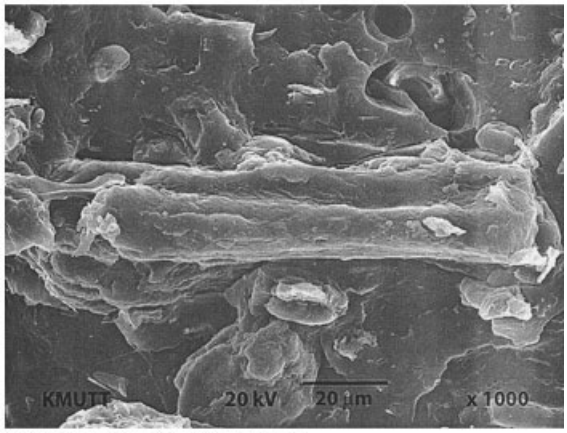



**Figure 7** Variations in the impact strength and hardness of PP/wood sawdust composites for various types and contents of impact modifiers.




(Figure 8a) (2.0%wt Engage8407)




(Figure 8b) (11.1%wt Engage8407)

**Figure 8** SEM micrograph of PP/wood composite: (a) low impact strength (2.0 wt % Engage8407), (b) high impact strength (11.1 wt % Engage8407).

The differences in the toughness results (elongation at break and impact strength), as a result of different impact modifiers, should be considered using their physical and mechanical properties and molecular characteristics, which are shown in Table I. It can be seen that the composite with Engage8407 gave the highest toughness properties, the effect being more pronounced at high concentrations of the impact modifier (about 6.4–11.1 wt % of the PP). This was expected since the Engage8407 had higher values of comonomer content, melt flow rate index, and elonga-

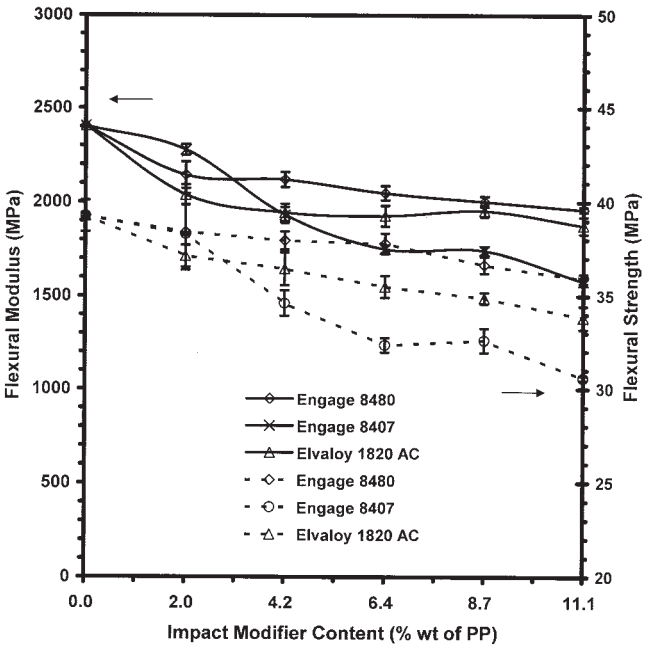



**Figure 9** Changes in flexural modulus and flexural strength of PP/wood sawdust composites for various types and contents of impact modifiers.

tion at break, coupled with low values of softening and melting temperatures. The hardness results of PP/wood composites containing various types and concentrations of impact modifiers are also given in Figure 7. The results were in line with the elongation at break and impact strength in terms of composite toughness. That is, the hardness of the composites decreased when toughened by the elastomeric materials.

Flexural modulus and strength of the PP containing 30 wt % wood sawdust with varying types and concentrations of impact modifiers are shown in Figure 9. It was found that both flexural modulus and strength of the PP/wood composites decreased progressively with increasing impact modifiers. This was because the modulus and the strength of the impact modifiers themselves were less than those of the unmodified PP/wood composites. For a given impact modifier content, the composites containing Engage8480 exhibited the highest flexural modulus and strength as expected, the lowest being given by the use of Engage8407. The reasoning for this can be explored using the flexural property results of the impact modifiers themselves, as listed in Table I.

## Effects of types and contents of coupling agents

As stated earlier, to improve the mechanical properties of the toughened PP-wood sawdust composites, some coupling agents may need to be introduced, this accounting for the formation of interfacial bonding between the PP and sawdust particles. The PP-wood composite containing 30 wt % wood sawdust and 11.1 wt % impact modifier (Engage8407) was selected for investigating the effects of the types and concentrations of coupling agents on the properties. Figure 10 shows the effect of the MAPP coupling agent on the tensile modulus of PP/wood sawdust composites. In general, it was observed for all coupling agents that the tensile modulus increased to a maximum value around a coupling agent content of 2.0 wt % of wood sawdust fibers. After that, the tensile modulus tended to stabilize. The tensile strength of the composites was also found to increase, accompanied by a decrease in elongation at break, with increasing MAPP coupling agent content (Fig. 11). The decrease in elongation at break was expected since the composites with MAPP coupling agents now were stiffer and had higher strength. The optimum MAPP coupling agent content of around 2.0 wt % added in the composites seemed to agree with a number of related works on PP-natural fiber composites.[12,20] The optimum concentration (around 2.0 wt % MAPP), giving the maximum tensile modulus and tensile strength in the composites, indicates high interfacial interactions between the sawdust and the PP matrix. This was associated with the ester linkages formed by chemical reactions of the MAPP coupling agent and sawdust particles, and the physical entanglement of PP molecules from the MAPP coupling agent and the matrix; the descriptions of this mechanism can be found elsewhere.[21]

This explanation can be substantiated by considering the SEM micrographs in Figure 12, which show the fractured surfaces of the PP/wood sawdust compos-

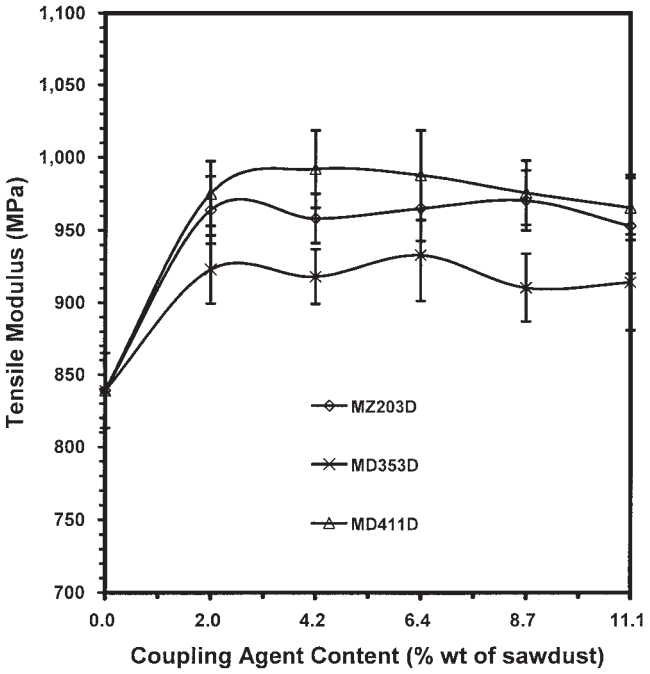



**Figure 10** Influence of coupling agent type and content on tensile modulus of PP/wood sawdust composites.

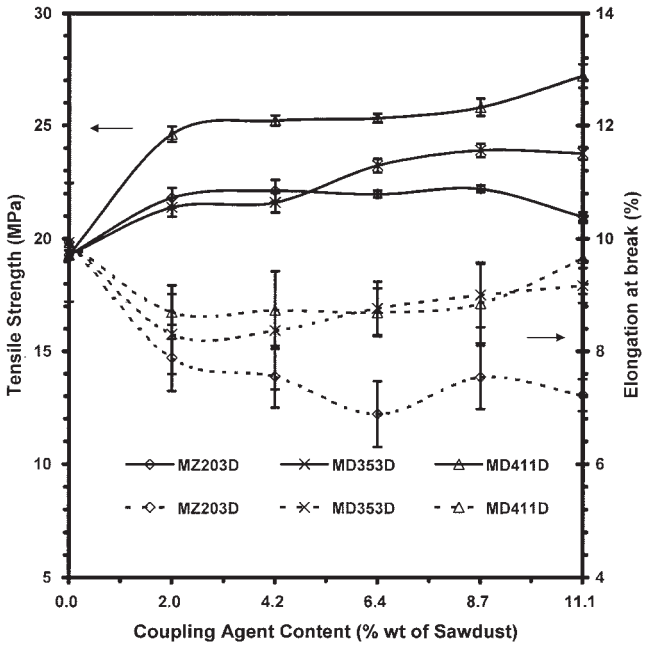
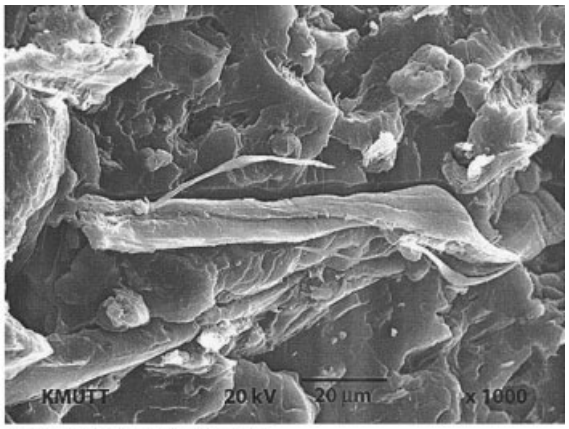



**Figure 11** Effect of coupling agent type and content on tensile strength and elongation at break for PP/wood sawdust composites.

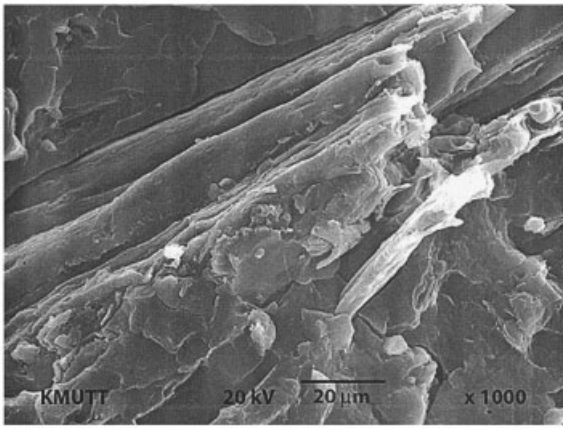
ites at low (Fig. 12a for untreated composite) and high (Figs. 12b and 12c for 2.0 wt % and 11.1 wt % MD411D-MAPP treated composites, respectively) tensile strength. It can be seen that the composite without a MAPP coupling agent exhibited some interfacial de-bondings (and voids) between the wood sawdust particles and the PP matrix, whereas this did not occur when introducing the MAPP coupling agent into the composite. The stabilization of the tensile modulus and tensile strength at high coupling agent concentrations (4.2–11.1 wt %) may be because excess concentrations of MAPP higher than 2.0 wt % in the composites played a minor part as a polymer in the bulk materials, and did not participate any further in the interfacial adhesion between the PP and the wood sawdust in the composites, as the wood concentration was kept constant.

Considering the effect of the type of MAPP coupling agent, it was noticeable that the tensile strength of the composite with MD411D was the highest. This may be because, using the material property results of all coupling agents in Table II, MD411D had the higher wt % MAH-grafting level (as compared with MZ203D) and higher molecular weight of PP copolymer (as compared with MD353D, which was indicated by the melt flow rate value). Therefore, the composite with MD411D had more chances to form chemical bonding (ester linkages) between the coupling agent and the wood sawdust, and to enhance the physical entanglement of the PP molecules, as discussed earlier.

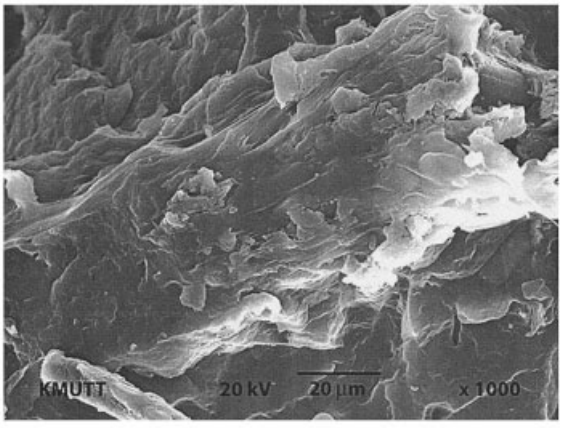
Figure 13 shows the results of the flexural modulus and strength of the PP/wood sawdust composites with different types and concentrations of MAPP coupling agents. The changing trends of the flexural results affected by types and concentrations of MAPP coupling agents, including their explanations, are very similar to those of the tensile strength results.




(Figure 12a) (0.0%wt MAPP)




(Figure 12b) (2.0%wt MAPP)




(Figure 12c) (11.1%wt MAPP)

**Figure 12** SEM micrographs of PP/wood composites: (a) untreated, (b) treated with 2.0 wt % MAPP, and (c) treated with 11.1 wt % MAPP.

Figure 14 illustrates the impact strength of the PP/wood sawdust composites with different types and concentrations of MAPP coupling agents. One would expect that the impact strength of polymer/wood composites should either be unaffected or increase with addition of the MAPP coupling agent since the interfacial bonding between the polymer and fiber phases has formed. Work by Colom and coworkers[2] suggested that the impact strength of the polyolefin-cellulosic fiber composites increased after treating the fiber surface with maleated-ethylene, but this could be beneficial at low fiber content. At high fiber content (about 30–40 wt %), the impact properties of the treated composite could be lower than those of the untreated. This was agreed to by the results given in Figure 14, as this part of the work used 30 wt % wood sawdust content, which shows a decrease of the impact strength of the composites with the presence of the MAPP coupling agent. However, it was found that the impact strength was unaffected by the coupling agent content. The explanation for this (stabilized impact strength) is probably similar to that given for tensile properties as discussed earlier.

Figure 14 also shows the hardness results of PP/wood composites containing various types and concentrations of MAPP coupling agents. The hardness results did not change with varying concentrations of the coupling agents, but were affected by the type of coupling agent. It seems that the composites containing coupling agents with higher MAPP-graft levels and higher molecular weights of PP copolymer gave higher hardness values.

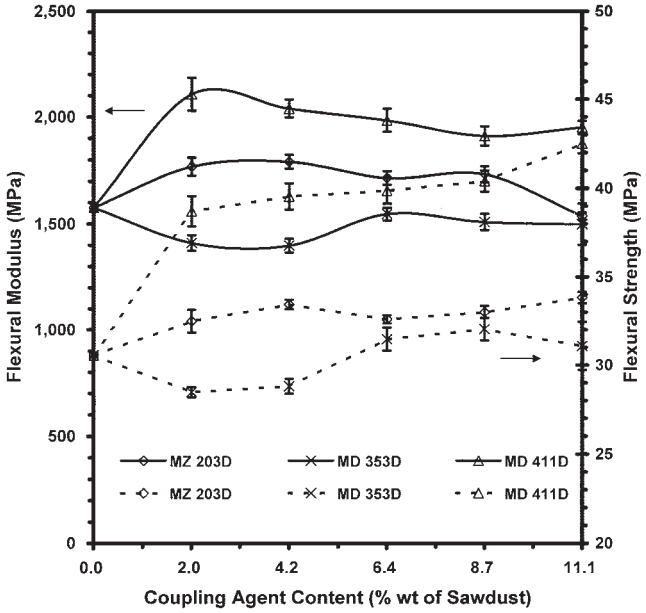



**Figure 13** Changes in flexural modulus and strength of PP/wood sawdust composites for various types and contents of coupling agents.

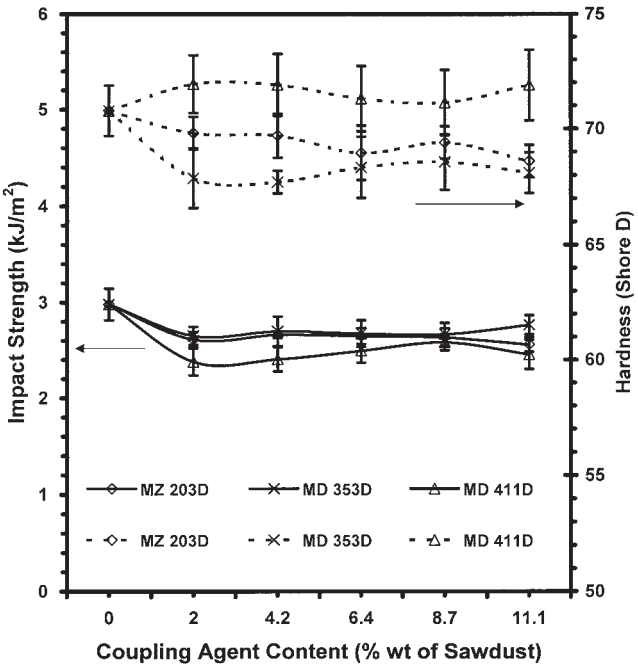



**Figure 14** Variations in the impact strength and hardness of PP/wood sawdust composites for various types and contents of coupling agents.

## CONCLUSIONS

Three types of MAPP and impact modifiers, used as property promoters, were introduced into PP/wood sawdust composites, and the mechanical properties investigated. The results suggest that as the wood fiber content was increased, the overall strength and toughness of the composites decreased. The toughness of the PP/wood composites could be regained by adding an impact modifier, especially one with high comonomer content and high melt flow index. Increasing impact modifier content enhanced the toughness of the composites. An addition of 2.0 wt % MAPP concentration into the PP/wood composites, containing 11.1 wt % selected impact modifier, is suggested for optimization of the overall mechanical properties. To more effectively improve the strength of the composites, a MAPP coupling agent with high wt % MAH-grafting level and high molecular weight of PP copolymer should be selected.

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