ORIGINAL ARTICLE

Influence of Meranti sawdust aspect ratios and amount of loadings on mechanical and morphological properties of composites from polypropylene and Meranti sawdust

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Abstract

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Influence of Meranti sawdust aspect ratios and amount of loadings on mechanical and morphological properties of composites from polypropylene and Meranti sawdust
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This research focused on the mechanical and morphological properties of polypropylene composites reinforced by Meranti hardwood sawdust. The effects of sawdust aspect ratio (L/D ratio), the amount of the sawdust and the application of maleic anhydride-grafted-polypropylene (MAPP) coupling agent on the composite properties were also investigated. Composites were prepared by compounding in a single-screw extruder, then, injection moulding technique was used to obtain testing specimens. It was found that Young's modulus and flexural modulus of the sawdust-polypropylene composites were increased prominently with the increasing of not only the sawdust aspect ratio but also the % sawdust content. Besides, impact strength was depended on the sawdust aspect ratio. In addition, the use of the MAPP coupling agent had a profound effect on the mechanical properties of the composites. Micrographs from scanning electron microscope (SEM) revealed the improvement of adhesion in the interfacial boundary between the sawdust and the polypropylene matrix.

Key words : Meranti sawdust, polypropylene composite, mechanical properties

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บทคัดย่อ

จุฑารัตน์ ปรัชญาวรากร และ กนิษฐา อังกุรรัชต์
ผลของอัตราส่วนระหว่างความยาวต่อความกว้างและปริมาณของขี้เลื่อยจากไม้เต็งต่อสมบัติเชิงกลและสัณฐานวิทยาของคอมโพสิตจากพอลิพรอพิลีนและขี้เลื่อยจากไม้เต็ง
ว. สงขลานครินทร์ วทท. 2546 25(5) : 595-606

งานวิจัยนี้มุ่งเน้นไปที่สมบัติเชิงกลและสัณฐานวิทยาของพอลิพรอพิลีนคอมโพสิตเสริมแรงด้วยขี้เลื่อยจากไม้เนื้อแข็งชนิดไม้เต็ง โดยศึกษาผลของอัตราส่วนระหว่างความยาวต่อความกว้างของขี้เลื่อย ปริมาณขี้เลื่อยและการใช้สารคู่ควบชนิดมาลิอิกแอนไฮไดรด์-กราฟท์-พอลิพรอพิลีนที่มีต่อสมบัติต่าง ๆ ของคอมโพสิต คอมโพสิตสามารถเตรียมได้โดยการผสมในเครื่องอัดรีดชนิดเกลียวหนอนเดี่ยว จากนั้นทำการขึ้นรูปเป็นชิ้นงานที่ใช้ในการทดสอบโดยใช้เทคนิคการฉีดขึ้นรูป ซึ่งพบว่าค่ามอดุลัสและมอดุลัสโค้งงอของคอมโพสิตจากพอลิพรอพิลีนและขี้เลื่อยนั้นเพิ่มขึ้นอย่างเด่นชัดเมื่ออัตราส่วนระหว่างความยาวต่อความกว้างของขี้เลื่อยและปริมาณขี้เลื่อยเพิ่มขึ้น สำหรับความทนทานต่อแรงกระแทกนั้นขึ้นอยู่กับอัตราส่วนระหว่างความยาวต่อความกว้างของขี้เลื่อย นอกจากนี้การใช้สารคู่ควบชนิดมาลิอิกแอนไฮไดรด์-กราฟท์-พอลิพรอพิลีนส่งผลอย่างเด่นชัดต่อสมบัติของคอมโพสิต ภาพจากกล้องจุลทรรศน์อิเลคตรอนแบบส่องกราดแสดงถึงการปรับปรุงแรงยึดเหนี่ยวระหว่างวัฏภาคของขี้เลื่อยและพอลิพรอพิลีน

ภาควิชาเคมี คณะวิทยาศาสตร์ สถาบันเทคโนโลยีพระจอมเกล้าเจ้าคุณทหารลาดกระบัง ลาดกระบัง กรุงเทพฯ 10520

Nowadays, plastic products are receiving more and more attention due to their versatile applications. However, disadvantages of the plastic arrive when they are required to accept high forces or be stiffer. This is the origin of the production of plastic composites that contain both polymer matrix and organic or inorganic reinforcement. The popular inorganic reinforcing component, such as glass fibres, is highly costly; therefore, one possible option to reduce the composite production cost is to use organic wood fibres because of their abundancy, favourable cost and high stiffness.

In wood industries such as the furniture industry or paper industry, a large amount of wood flakes and wood flours in the form of sawdust are always found as wastes. These unused materials are usually applied as a fuel source or for the production of plywoods. Because of high availability and low cost of the sawdust, it can be more versatile and valuable by compounding the sawdust with plastics in order to improve plastic properties, for example, high strength and easy processing that are better than wood or plastics.

In the related literature, it has been reported that most polymer composites involve fibre reinforcement, for instance, bamboo fibres (Chen *et al*., 1998), fibres from oil palm empty fruit bunch (Rozman *et al*., 2003), aspen fibres (Coutinho *et al*., 1997 and 1998) or eucalyptus wood (Marcovich *et al*., 1998). These composites bring about greater mechanical properties than those of polymers alone, especially the Young's modulus and flexural modulus (Chen *et al*., 1998; Rozman *et al*., 2003; Coutinho *et al*., 1997 and 1998; Marcovich *et al*., 1998 and Angles *et al*., 1999). Furthermore, the application of coupling agents such as maleic anhydride-grafted-polypropylene (MAPP) (Chen *et al*., 1998; Rozman *et al*., 2003; Coutinho *et al*., 1997 and 1998 and Marcovich *et al*., 1998) or silane coupling agent (Coutinho *et al*., 1997 and Maldas and Kokta, 1991), have received much attention on the account of their effectiveness in intensifying phase compatibility between the polymer matrix and the reinforcement, leading to preferred mechanical properties of the composites.

In this study, polymer composites were prepared using polypropylene, a type of commodity plastics that can be used for numerous applications, and the reinforcement from Meranti hardwood sawdust, easily found in furniture manufacturing factories, especially in Thailand. The effects of sawdust aspect ratio (L/D ratio) and the amount of the sawdust on the composite mechanical and morphological properties were investigated. In addition, the dependence of MAPP coupling agent on the composite properties were compared to the properties of composites without the use of MAPP.

Materials and Methods

1. Materials

The plastic used in this study was Profax Z30S homopolymer from HMC Polymers, Thailand. The density and melt flow index were 0.90 g/cm^3 and 22 g/10 min, respectively. Meranti hardwood (*Shorea foxworthyi*) sawdust was collected from furniture factories and it was used as reinforcement. The collected sawdust was ground and seived to 4 different mesh sizes of 20, 35, 50 and 100, giving 4 different sawdust aspect ratios. MAPP coupling agent with 0.5% maleic anhydride was Polybond 3150 from TOA Uniroyal Chemicals.

2. Mixing and Processing Procedures

The ground and sieved Meranti sawdust was mixed with polypropylene at different loadings as follows: 10 20 and 30 php (parts per hundred polymer by weight) using a high-speed mixer with a speed at 1500 rpm for 5 minutes. Then, the mixture was passed though a single-screw extruder for compounding using a screw speed of 40 rpm. The obtained extrudate was ground and injected into testing specimens using an injection moulding machine. The temperature range between 170-190°C was selected from the feeding section to the metering section.

It should be noted that the addition of MAPP was performed at the first mixing stage only for the MAPP-treated composites. The amount of MAPP was kept at 5% by weight of the sawdust. In addition, the sawdust loading was maintained at 10 php in order to study the influence of sawdust aspect ratios, and the sieved sawdust of 35 mesh was selected to examine the dependence of sawdust contents.

3. Testing

3.1 Optical Microscopy

Optical microscopy was the technique used for the determination of sawdust aspect ratios (length to diameter ratios). The calibration was performed using a 1 mm graticule and at least 100 particles of sawdust per each sieve sizes (20, 35, 50 and 100 meshes) were examined to obtain average values of the sawdust aspect ratios.

3.2 Mechanical Testing

Tensile strength, Young's modulus and % elongation at break of the PP and its composites were obtained using a Universal Testing Machine following ASTM D-638. The gauge length of the specimens was 50 mm and the crosshead speed was 5 mm/min.

For flexural strength and modulus (ASTM D-790), the speed test was 10 mm/min and the span length was 40 mm. Impact strength was the Izod type as ASTM D-256.

For each test, at least 10 specimens were used in order examine the average values of the mechanical properties.

3.3 Morphology

Morphology of the prepared composites was performed by using LEO 1455 VP Scanning Electron Microscopy, SEM. The specimens were first kept in N_2 atmosphere, then, they were broken. After that, the broken surfaces were coated with a thin layer of gold to prevent an electrical charging during the SEM observation.

3.4 FTIR Spectroscopic Study

Composite films with the thickness of 10-100 mm were used for FTIR measurements. The FTIR spectra were recorded on a Spectrum 2000 GX spectrometer (Perkin-Elmer) using the KBr disc technique for 16 scans. The resolution was 4 cm^{-1} and the spectral scanned range was 4000-650 cm^{-1}.

Results and Discussion

1. Mechanical Properties

1.1 Effect of Sawdust Aspect Ratio

1.1.1 Tensile Properties

The sawdust aspect ratios were

examined using the optical microscopy technique from 100 measurements for each sieved size. It can be seen in Figure 1 that the distinct sawdust particle sizes, from the sawdust sieving through different sizes of 20, 35, 50 and 100 meshes, give deviant values of average aspect ratios of 10.5, 8.6, 6.4 and 4.3, respectively. In other words, the smaller the sieve sizes, the higher the value of aspect ratios.

The influence of the aspect ratio of the Meranti hardwood sawdust on the mechanical properties of the sawdust-PP composites was carried out using a constant amount of 10 php of the sawdust. The relationship between the tensile strength and aspect ratio of the composites is presented in Figure 2(a). It is found that at low aspect ratio the addition of the sawdust into the composites creates the phase discontinuity and heterogeneity inside the composite specimens, resulting in a drop in tensile strength. When the value of L/D is higher, the tensile strength of the composite seems to be increased compared to the lower aspect ratio of the sawdust. This is due to the fact that, with good interfacial adhesion between fibre and matrix, higher sawdust aspect ratio or longer length of the sawdust makes the reinforcement transfer forces along the length of the reinforcement more efficiently (Hull and Clyne, 1996).

With the application of MAPP coupling agent, the tensile strength of the composites shows an increasing trend (Figure 2(a)), compared to the composites without the coupling agent. The increment is caused by maleic anhydride from the MAPP molecule that can react with the hydroxyl groups of cellulose or hemicellulose, two main components in the sawdust (Tsoumis, 1991), giving esterification reaction between the reinforcement and the matrix phases (Felix and Gatenholm, 1991). Furthermore, the long continuous chains in the MAPP molecules are compatible with the PP matrix chains via physical entanglement (Felix and Gatenholm, 1991). The combination of both the chemical and physical bondings causes the improvement of the composite tensile strength. The chemical formation between the MAPP and sawdust structure from the results of FTIR spectra will be discussed later.

Figure 2(b) shows the relationship between the composite Young's modulus and the aspect ratio of the sawdust. When the sawdust is introduced into the PP matrix, the composite

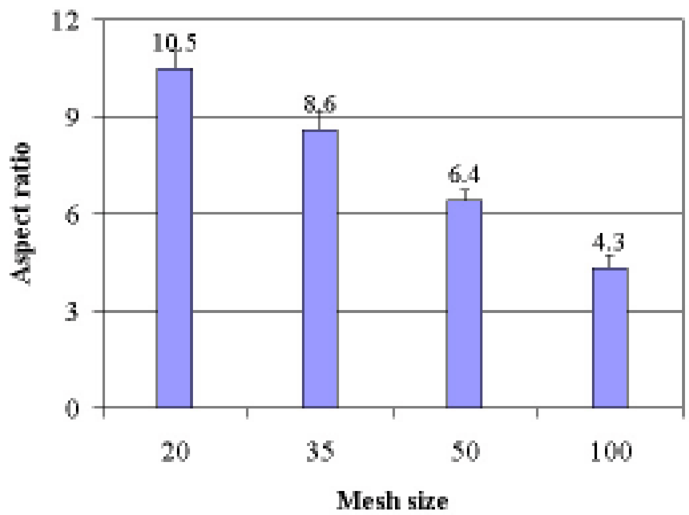


Figure 1. Avearge values from 100 measurements of sawdust aspect ratio obtained from different sieve sizes (a) 20 mesh, (b) 35 mesh, (c) 50 mesh and (d) 100 mesh, using optical microscopy technique.

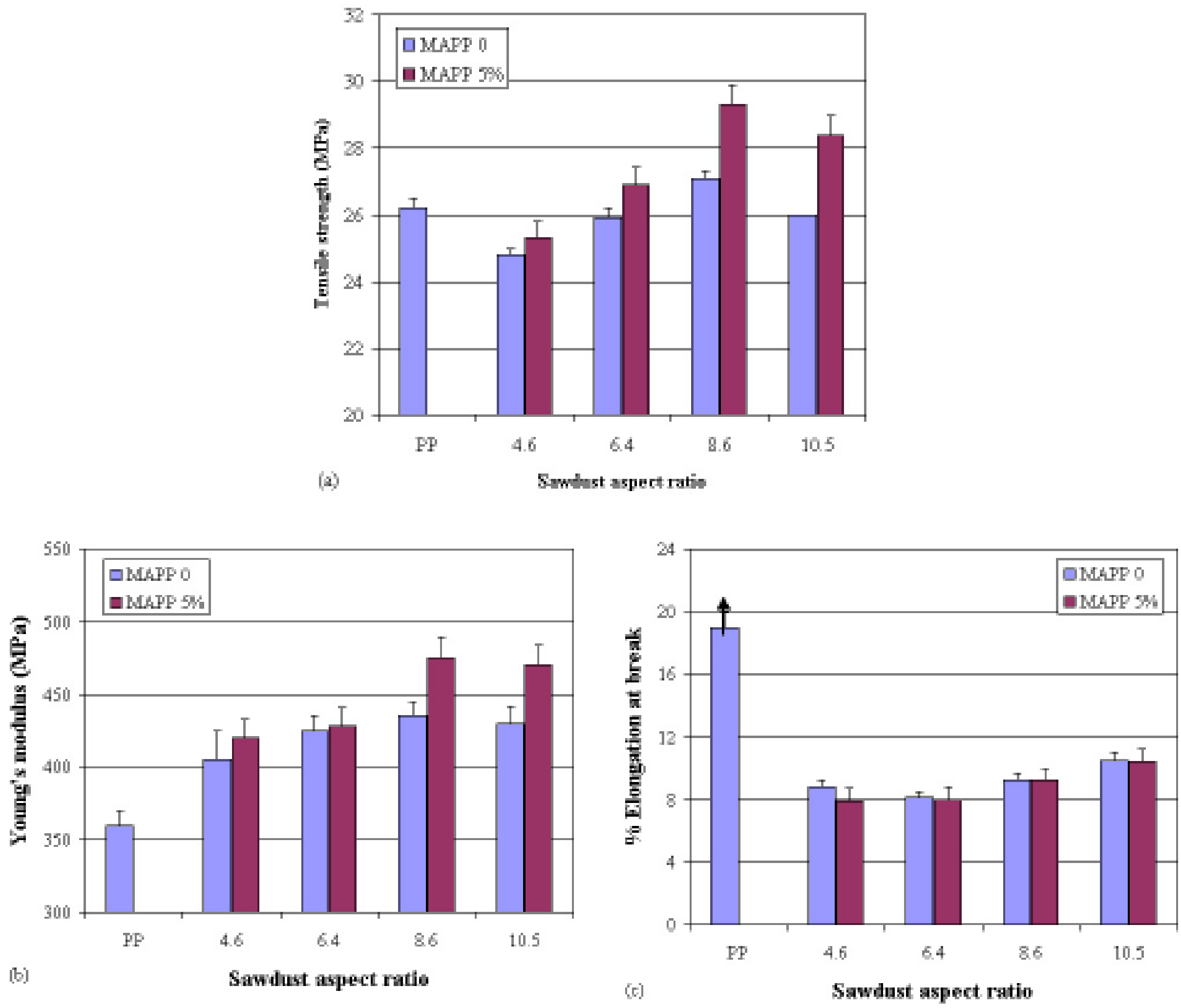


Figure 2. The effect of sawdust aspect ratio on the (a) tensile strength (b) Young's modulus and (c) % elongation at break of the Meranti sawdust reinforced PP composites.

Young's modulus rises due to the stiff phase of the hardwood sawdust component. Besides, greater values of the aspect ratio lead to higher values of composite Young's modulus, especially when the L/D ratio is 10.5 and the MAPP coupling agent is used. This can be explained by the fact that high L/D ratio of the sawdust results in the sufficient stress transfer along with the length of the sawdust (Hull and Clyne, 1996). The enhancement in the tensile strength and the Young's modulus with the sawdust aspect ratio leads to the reduction of % elongation at break of the composites with and without the application of MAPP coupling agent, in comparison to the PP polymer (Figure 2(c)).

1.1.2 Flexural Properties

Flexural properties of the composites can be accomplished by three-point bending test using the universal-testing machine. From Figures 3(a) and 3(b), it can be clearly seen that the flexural strength and modulus of the composites are enlarged, notably in the MAPP treated composites that leads to approximately 80% increment, in comparison to PP. The effect of the MAPP coupling agent on the flexural properties of the

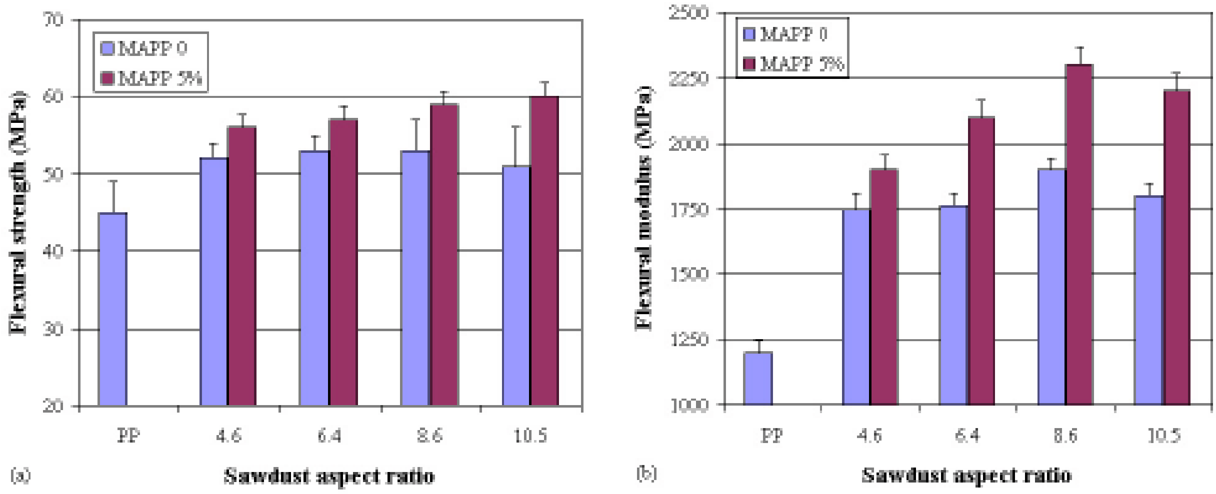


Figure 3. Comparison of flexural properties (a) flexural strength and (b) flexural modulus of the composites on the sawdust aspect ratio.

composites is similar to that of the composite tensile properties as observed in Section 1.1.1, due to the improvement of the interfacial boundary by the chemical and physical interactions. Moreover, higher aspect ratio of the sawdust also gives greater composite flexural properties by reason of the satisfactory stress transfer in longer length of sawdust as mentioned earlier.

1.1.3 Impact Strength

Impact strength of the composites between PP and Meranti sawdust with the different aspect ratios is illustrated in Figure 4. It can be seen that the impact strength seems to be enhanced with the increase in sawdust aspect ratio. It can be explained that when the impact force is given into the composite specimens, stress concentration occurs, consequently, the specimen fracture appears at the interphase area between the reinforcement and the matrix. Therefore, longer reinforcing component can receive greater impact forces than can the shorter L/D ratio reinforcing component. Lawrence and Robert, (1994) stated that Izod impact strength increased with the length of reinforcing fibres and this is in accordance with this present study. In addition, the MAPP coupling agent makes good inter-boundary bridges between the reinforcement and the matrix and that results in greater impact strength of the MAPP-treated composites.

1.2 Effect of Sawdust Loading

1.2.1 Tensile Properties

The effect of the amount of sawdust on the mechanical properties of the sawdust-PP composites can be examined by varying the sawdust loadings as follows: 10 20 and 30 php (parts per hundred polymer) when the 35 mesh sawdust is maintained. When the amount of the sawdust is extended, the tensile strength of the composite is increased (Figure 5(a)), as reported in the PP composite reinforcing with paper pulp (Woodhams and Law, 1993). Besides, the usage of MAPP coupling agent leads to greater composite tensile strength as shown in Figure 5(a).

For the Young's modulus of the composites reinforced by the Meranti sawdust in Figure 5(b), it shows the increased values with the raised amount of the sawdust. It can be seen that the Young's modulus at 30 php sawdust loading of the untreated and treated composites with MAPP coupling agent presents an increment of approximately 25% and 40%, respectively. This is due to the solid particle of sawdust reinforcement leading to greater stiffness in polymer composites. Because of the composite shows greater stiffness than the

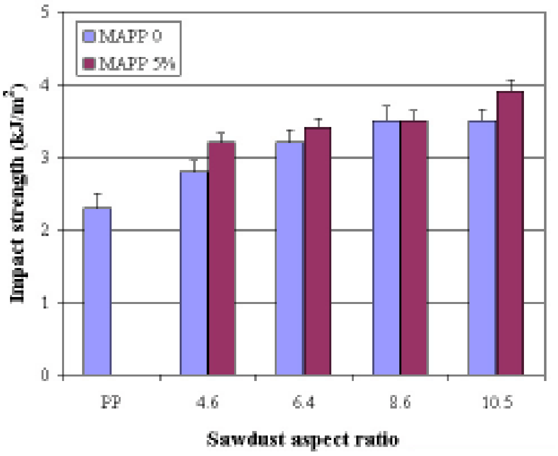


Figure 4. Impact strength of Meranti sawdust and PP composites with different sawdust aspect ratios.

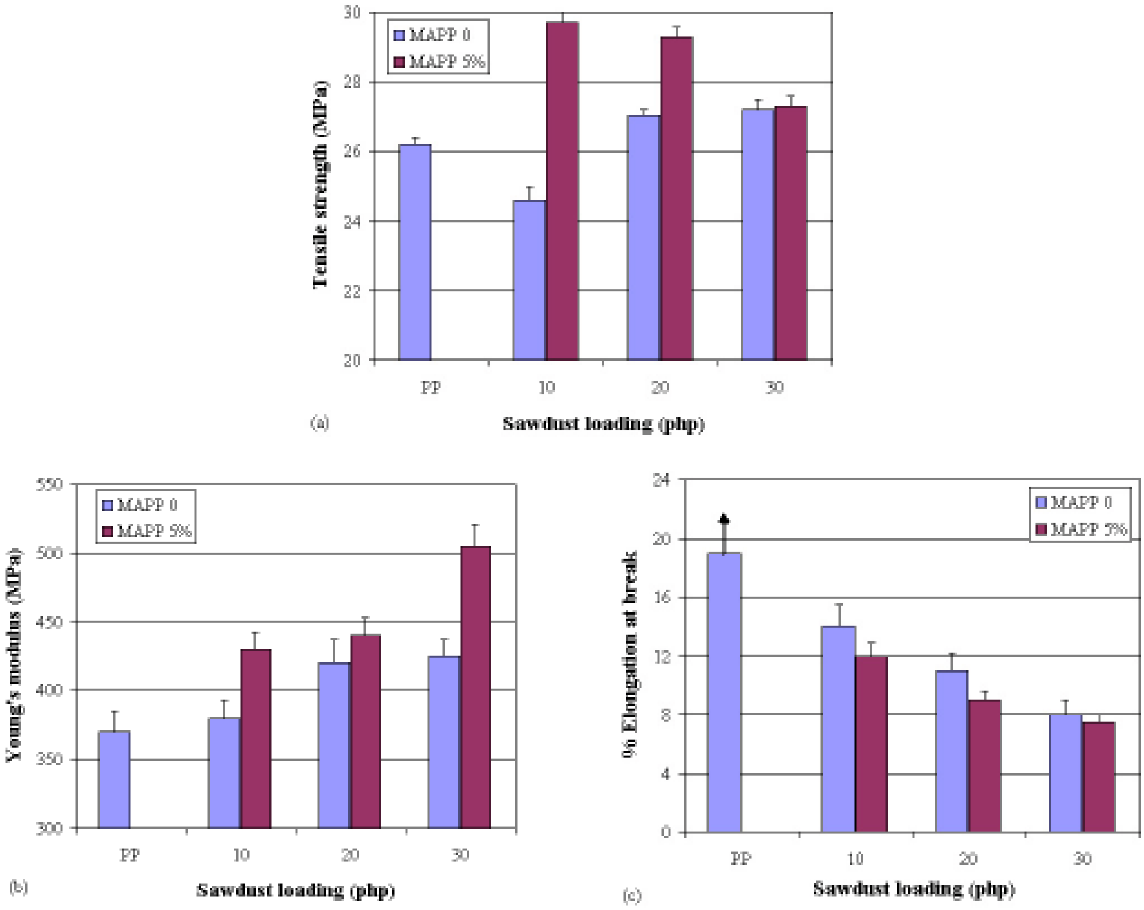


Figure 5. The relationship between (a) tensile strength (b) Young's modulus and (c) % elongation at break with the sawdust loadings for the PP-sawdust composites.

PP polymer, it causes a decline in % elongation at break, particularly when the amount of the sawdust is 30 php (Figure 5(c)).

1.2.2 Flexural Properties

An expanding tendency in flexural properties of the composites with the increasing in sawdust content is illustrated in Figure 6. From Figure 6(a), it can be seen that the flexural strength of the composites increases inconsiderably, whereas the composite flexural modulus enhances enormously (Figure 6(b)). The improvement in the composite flexural strength and modulus was found to be about 15% and 55%, respectively, comparing between the PP and composite of 30 php sawdust. Likewise, the application of MAPP coupling agent obviously improves the PP-Meranti sawdust composite flexural properties. The flexural modulus of the MAPP treated composite at 20 php demonstrates the flexural modulus that is about twice the value of the PP polymer.

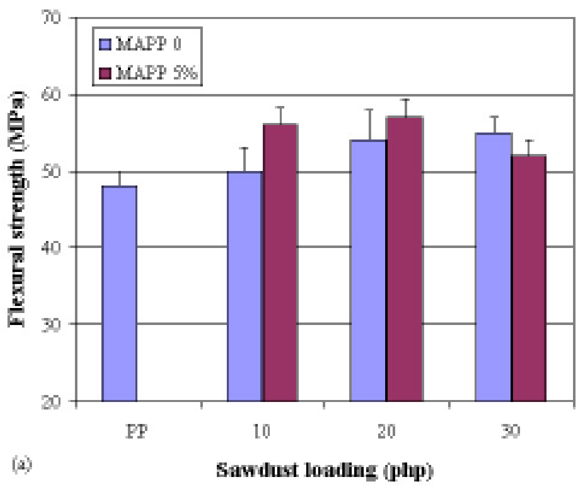
1.2.3 Impact Strength

The effect of sawdust loading on the composite impact strength is presented in Figure 7. The nearly constant value of more or less 2.5 kJ/m^2 was obtained for the composites without the application of MAPP so it can be said that the impact property of the composites is not influenced by the amount of the sawdust. However, the MAPP treatment causes the enhancement in the composite impact strength due to the improved interfacial adhesion between the two different phases. It helps absorbing impact energy from the reinforcement to the matrix; as a result, higher impact strength can be obtained.

2. Morphology

Morphology of the PP composites reinforced by Meranti sawdust is represented in Figure 8. Figure 8(a) shows the dispersion of sawdust phase into the PP matrix phase. It can be seen that the reinforcement agglomeration can be obtained, and this observation can also be easily found in some composites reinforced by longer fibres (Chen *et al.*, 1998 and Felix and Gatenholm, 1991).

In the composites without the use of MAPP coupling agent for adapting the phase compatibility, the long crack and the resulting void between the sawdust reinforcement and the PP matrix can be seen clearly, as presented in Figure 8(b). Nonetheless, the utilisation of the MAPP coupling agent makes the crack or voids between the interphase boundary disappeared as can be noticed in Figure 8(c). From the fucntion of the MAPP coupling agent as found in the SEM micrographs, it indicates the



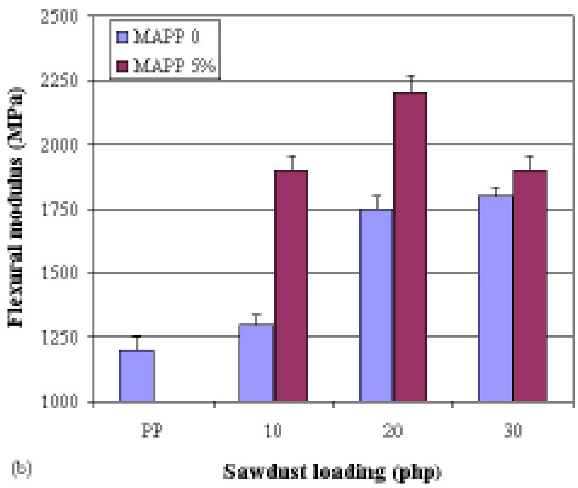


Figure 6. The variation of the different amounts of sawdust on the (a) flexural strength and (b) flexural modulus of the composites.

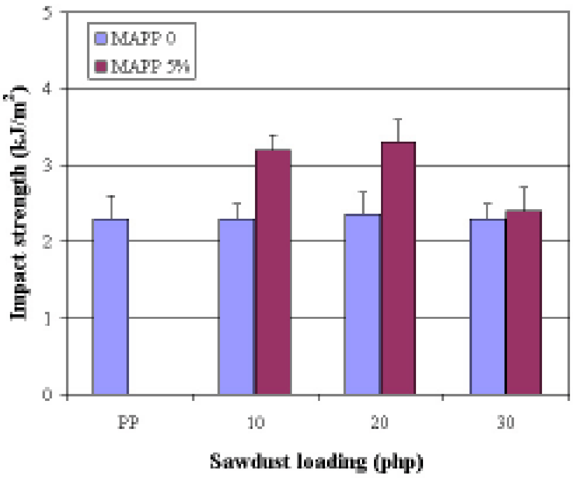


Figure 7. Impact strength of Meranti sawdust and PP composites obtained from the application of different sawdust contents.

phase compatibility between the sawdust and the PP. The results are in good accordance with the enhancement in the composite mechanical properties as observed in Section 1.

3. FTIR Spectroscopic Study

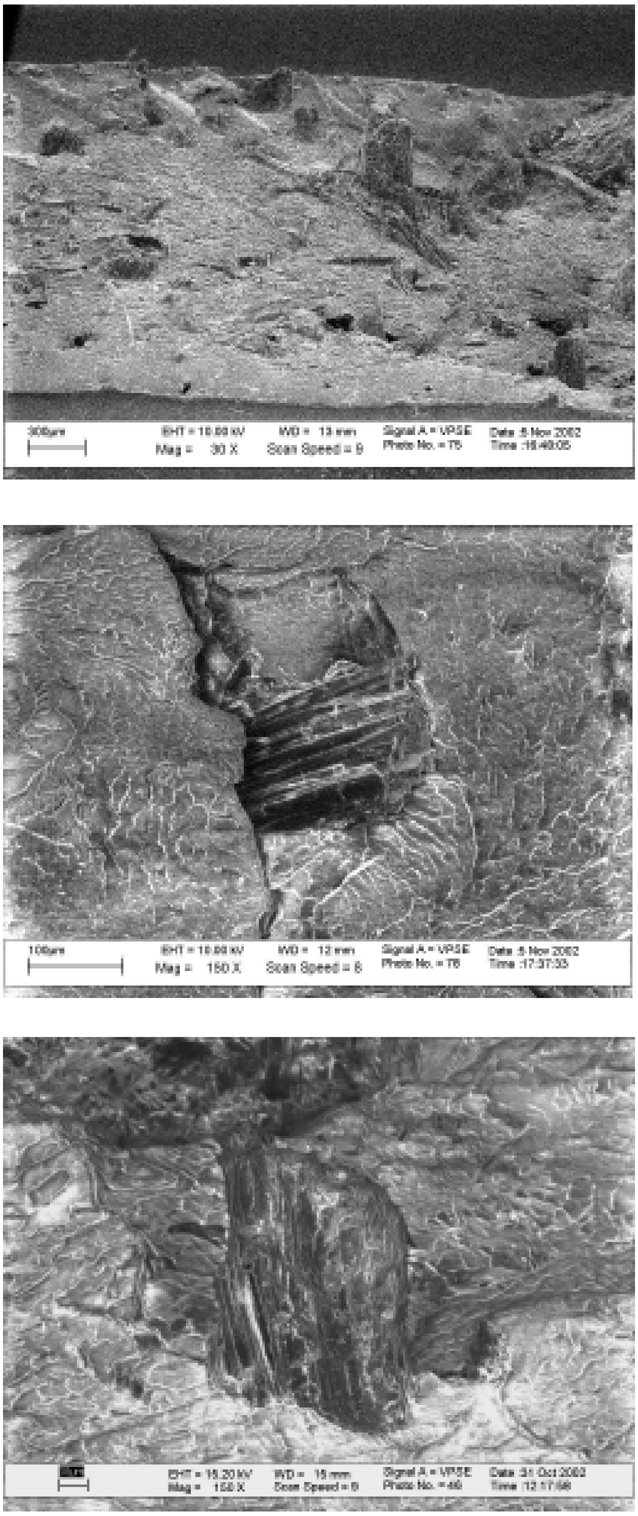
Functional group analysis of the PP, untreated and treated sawdust-PP composites can be performed using FTIR spectroscopy. PP spectrum in Figure 9 shows a broad peak in the range of 2600-3100 cm^{-1} referring to C-H stretching of CH_2 or CH_3 of PP chain (Bower and Maddams, 1989). Other vibrational peaks appear in the region of 1300-1400 cm^{-1} assigned for C-H deformation of CH_2 and/or CH_3 (Bower and Maddams, 1989; Kazayawako *et al*., 1997 and Matias *et al*., 2000) of cellulose and hemicellulose.

It can be seen in Figure 8(b) that the untreated composite spectrum, composed of only PP and sawdust, indicates an extra prominent peak position at 1600 cm^{-1} (Bower and Maddams, Kazayawako *et al*., 1997 and Matias *et al*., 2000), compared to the PP spectrum in Figure 8(a), due to the benzene ring of lignin molecules in sawdust. Vibrational peaks appear in the region of 1300-1400 cm^{-1} also assigned for C-H deformation of CH_2 and/or CH_3 as found in PP. Another broad band found in the wavenumber range of 1000-1300 cm^{-1} refers to C-O stretching and C-O-C stretching of the cellulose in the composites (Bower and Maddams, 1989; Kazayawako *et al*., 1997 and Matias *et al*., 2000).

When the composite is treated with MAPP, the IR spectrum as illustrated in Figure 8(c) presents C=O stretching of carbonyl group and ester linkage at the wavenumber in the region of 1700-1770 cm^{-1} regarding to the interaction between the MAPP coupling agent and the hydroxyl group of the cellulose composite (Bower and Maddams, 1989; Kazayawako *et al*., 1997 and Matias *et al*., 2000). The ester linkage from the IR spectrum also leads to improvement in mechanical properties of the MAPP treated composites as reported in the earlier section.

Conclusions

Polypropylene composites reinforced by Meranti sawdust showed greater mechanical properties than PP polymer. When the influence of the sawdust aspect ratios and the amounts of the sawdust were considered, it was found that both factors resulted in a significant improvement of the composite Young's modulus and flexural modulus. Tensile, flexural and impact strengths were found to be slightly increased. Moreover, the



(a)

(b)

(c)

Figure 8. SEM micrographs for (a) untreated composites (30×) (b) untreated composites (150×) and (c) composite treated with MAPP (150×).

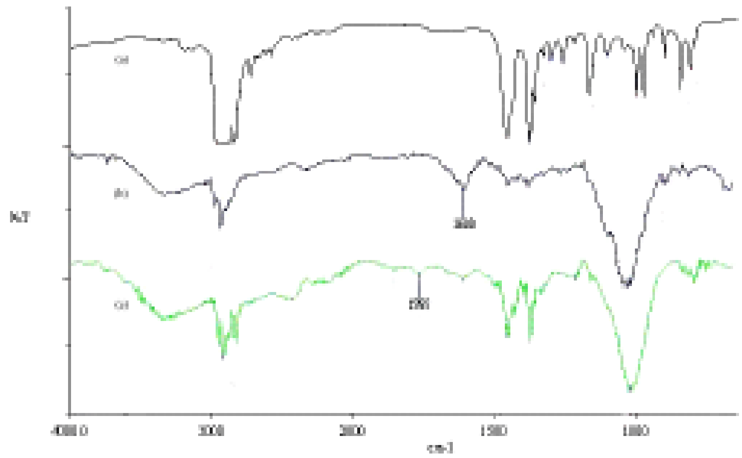


Figure 9. FTIR spectra for (a) PP, (b) composite of PP and sawdust and (c) composite of PP and sawdust with MAPP coupling agent.

addition of the MAPP coupling agent caused the enhancement in the phase compatibility that could be observed from the SEM micrographs. The evidence of the formation of ester bond between the sawdust and the MAPP could be obtained from IR spectra. Consequently, the MAPP-treated composites represented greater mechanical properties than those of the untreated composites.

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