

Effect of Silane-Based Coupling Agents and Acrylic Acid Based Compatibilizers on Mechanical Properties of Oil Palm Empty Fruit Bunch Filled High-Density Polyethylene Composites

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ABSTRACT: The mechanical properties of composites consisting of high-density polyethylene (HDPE) and oil palm fibrous wastes—that is, empty fruit bunch (EFB)—have been investigated. Tensile modulus showed an increase, whereas tensile strength, elongation at break, and impact strength decreased with increasing filler loading. The strong tendency of EFB to exist in the form of fiber bundles and the poor filler–matrix interaction is believed to be responsible for the poor strength displayed by the composites. Attempts to improve these properties using two types of coupling agents, that is, 3-aminopropyltrimethoxysilane (3-APM) and 3-aminopropyltriethoxysilane (3-APE) and two types of compatibilizers, poly(propylene–acrylic acid) (PPAA) and poly(propylene–ethylene–acrylic acid), (PPEAA), are described. While almost all chemical treatments increased the stiffness of the composites, limited improvement has been observed in the case of tensile strength. This has been attributed to the presence of fiber bundles that remain intact even after several types of chemical treatment have been carried out. Thus, the role of EFB as reinforcing agent is not fully realized. Scanning electron microscopy (SEM) micrographs revealed that the main energy-absorbing mechanisms contributing towards toughness enhancement is through the fiber bundle pull-out process. © 1998 John Wiley & Sons, Inc. *J Appl Polym Sci* 68: 2189–2203, 1998

Key words: high-density polyethylene; oil palm empty fruit bunch; filler; coupling agent; compatibilizer; mechanical properties; scanning electron microscope

INTRODUCTION

The use of lignocellulosic materials, such as wood and cellulose, in the production of thermoplastic composites is becoming more attractive, at least judging from the increasing amount of literatures in that areas.^{1–13} There are several factors that contribute to the observed trend. Lignocellulosic-derived fillers have many advantages compared to inorganic fillers, such as lower density, greater deformability, flexibility during processing with no harm to the equipment, and, of course, lower

cost per unit volume. Moreover, these materials are derived from a renewable resource.

Despite of the above-mentioned advantages, the use of lignocellulose fillers, in general, has not been extensive mainly due to a few reasons, such as poor dispersion characteristics in the thermoplastic melt, limited compatibility with the matrix, and limited thermal stability at the temperatures typically encountered during processing. These factors will obviously contribute to unsatisfactory final properties of the composites.

One of the lignocellulosic materials that is of great relevance to the Malaysian scenario is the large quantity of biomass generated by oil palm industries. Since the chemical composition of the oil palm is similar to that of wood, these wastes

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could be turned into new raw materials with expanding potentials. Recent investigations have shown that valuable products, such as thermoplastic composites,¹⁴ sheet molding compounds,¹⁵ and pulp and paper,¹⁶ could be produced from various biofibers of oil palm tree (*Elais guineensis*). The impetus to utilize the materials is further dictated by environmental pressure groups and stringent environmental laws and regulations.

Previously, the potential utilization of fillers that were derived from oil palm trees, namely, empty fruit bunch (EFB) and oil palm frond (OPF), for the production of high-density polyethylene (HDPE) composites have been reported.^{17,18} The investigation has been focused on the effect of filler loadings and filler particle size distribution on the mechanical properties of the composites. In general, it has been found that the incorporation of fillers into HDPE matrix has, to a certain extent, reduced both tensile and impact strength of the composites. This has been attributed to the poor filler dispersion and also poor filler–matrix interfacial bonding. Several studies on other lignocellulosic-filled thermoplastics have indicated that some property enhancements could be achieved via the incorporation of coupling agents and/or compatibilizers.^{1–13} Thus, it is the objective of the present work to investigate the effect of types and amount of coupling agents and/or compatibilizers on the mechanical properties of the EFB–HDPE composites. In this study, emphasis has been given to EFB instead of OPF for two main reasons. From a microstructure–property relationship point of view, both EFB–HDPE and OPF–HDPE composites displayed similar trends, and their overall mechanical properties are more or less comparable.^{17,18} Secondly, EFB is preferable in terms of availability and cost. EFB is readily available at a typical token price of USD 10.00 per tonne as compared to USD 30.00 per tonne for OPF. In addition, the amount of EFB waste generated by the palm oil industries in Malaysia is very high, that is, it is estimated to be about 8 million tonnes per year. Thus, considerable research and development efforts have to be undertaken in finding useful utilization of the EFB. This will also surely help in solving the environmental problems related to the disposal of the oil palm fibrous wastes.

EXPERIMENTAL

Materials

Oil palm empty fruit bunch (EFB), which consists of about 65% holocellulose and 25% of lignin (with

particle size distribution in the range of 270–500 μm), was supplied by Sabutek Sdn. Bhd., Teluk Intan, Perak, Malaysia. The polyethylene used was a high-density polyethylene (melt index of 0.7 g/10 min and density of 0.96 g/cc) from Thai Polyethylene Co. Ltd. (Bangkok, Thailand). In the present study, attempts were made to choose a suitable coupling agent and/or compatibilizer for the EFB–HDPE composite system. Two types of coupling agents, that is, 3-aminopropyltrimethoxysilane (3-APM) and 3-aminopropyltriethoxysilane (3-APE), were used. In the case of compatibilizers, poly(propylene–ethylene–acrylic acid) (PPEAA) and poly(propylene–acrylic acid) (PPAA) have been chosen. All coupling agents and compatibilizers used were supplied by Polysciences Incorporation (California).

Filler Treatment

Applications of the coupling agents (3-APM and 3-APE) and compatibilizers (PPEAA and PPAA) to the EFB–HDPE composites differ from one another due to the nature of the chemicals themselves. Both coupling agents were delivered in liquid form, and, prior to application, they were diluted in ethanol to make up to a 10% solution. Surface treatment of the EFB was carried according to the manufacturers' instruction. The EFB filler was charged into a bench-top tumbler mixer, and the 3-APM and 3-APE solution were added slowly to ensure uniform distribution of the coupling agent. After completion of the silane addition, the filler was continuously mixed for another 30 min. The treated filler was then dried at 100°C for about 5 h to allow complete evaporation of ethanol. Both PPAA and PPEAA compatibilizers came in ready-to-use powders and were added directly to the EFB–HDPE mix.

Compounding and Processing

The compounding of untreated and treated EFB–HDPE was carried out in a single-screw extruder (Betol Extruder Model 116) at a screw speed of 20 rpm, with barrel temperatures of 150, 160, and 170°C from feeding zone to the die zone, respectively. The compounds were extruded through a single 3-mm rod die and pelletized. The loading of the EFB was varied from 0 to 60% by weight of the filler.

The extrudates were hot-pressed in a mold of internal dimensions of 18.5 \times 13.5 \times 0.5 cm under a pressure of 4 MPa. Hot-press procedures in-

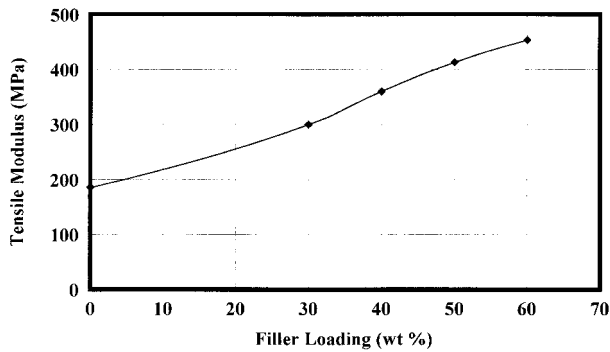


Figure 1 The effect of filler loading on the tensile modulus of EFB-HDPE composites.

involved preheating at 100°C, followed by heating at 170°C, and subsequent cooling under pressure. The total molding time was 15 min.

Testing

The sheet produced was cut into 2 types of test samples, that is, tensile and impact tests. Tensile tests were carried out according to ASTM procedure D-1708 using a universal testing machine (Instron model 1114) at a crosshead speed of 5 mm/min. The Izod impact tests were performed according to ASTM D256-88 on unnotched samples with dimensions of $6.5 \times 1.5 \times 0.5$ cm, using an impact pendulum tester (Zwick Model 5101). A minimum of 6 samples were tested in each case. All mechanical tests were carried out at room temperature. All samples were conditioned at $23 \pm 2^\circ\text{C}$ and $55 \pm 5\%$ RH for approximately 72 h before being tested.

Morphological Study

Studies on the morphology of the EFB-HDPE composites' tensile fracture surfaces were carried out using a scanning electron microscope (SEM), model Leica Cambridge S-360. The objective is to get some information regarding filler dispersion and bonding quality between filler and matrix and to detect the presence of microdefects, if any. The fracture ends of the specimens were mounted on aluminum stub and sputter-coated with a thin layer of gold to avoid electrostatic charging during examination.

RESULTS AND DISCUSSION

Effect of Filler Loadings

Figure 1 shows the effect of filler incorporation on the tensile modulus of untreated EFB-HDPE

composites. The increase in modulus with filler loading clearly indicates the ability of EFB fillers to impart greater stiffness to the HDPE composites. This is in agreement with the trend observed in other lignocellulosic-filled thermoplastics.⁵⁻¹³ On the contrary, tensile strength decreased steadily as the concentration of EFB increased in the composites (Fig. 2). This is not surprising since other studies have also indicated that the incorporation of filler into thermoplastic matrix may not necessarily increase the tensile strength of a composite.^{19,20} Unlike fibers that have a uniform cross section and relatively high aspect ratio (that is, the length-to-diameter ratio, or l/d), for irregular-shaped fillers such as EFB, their capability to support stress transmitted from the polymer matrix is rather poor. Thus, the stiffness and strength enhancement in the filled composites are, in general, much lower than that of fiber-reinforced systems.

The incorporation of EFB into HDPE matrix has resulted in a dramatic reduction in the elongation at break (EB) (Fig. 3). The ductility of HDPE, as indicated by its high EB, that is, about 130%, has been suppressed by the presence of EFB fillers. Similar observations have been reported by several workers for other lignocellulosic thermoplastic composites.⁵⁻⁹ This may be attributed to the decreased deformability of a rigid interface between the filler and the matrix material. At higher filler loading, the domination of filler-matrix interaction can be expected to diminish and is being replaced by filler-filler interaction. This claim is supported by the qualitative evidences obtained from the morphological studies of the fractured surfaces using SEM. This subject will be discussed further later.

Figure 4 illustrates the effect of filler loading

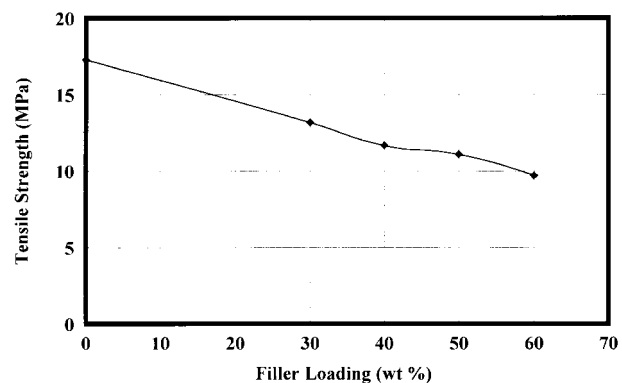


Figure 2 Variation of tensile strength with filler loading for EFB-HDPE composites.

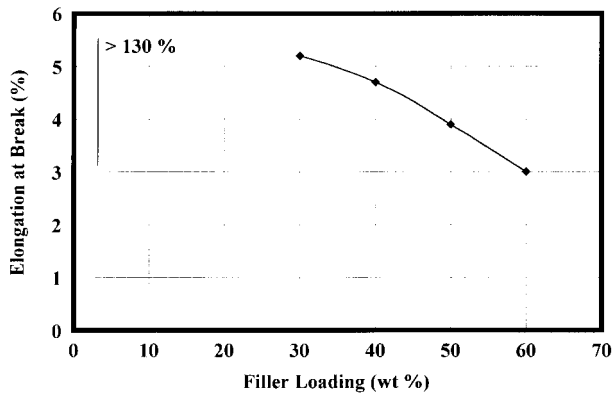


Figure 3 Elongation at break versus filler loading for EFB-HDPE composites.

on the impact strength of HDPE composites. As expected, the impact strength decreases in more or less the same manner as EB with increasing EFB loading. This clearly indicates that the presence of EFB has reduced the energy-absorbing capabilities of the composites. A similar trend has been reported by Myers et al.¹⁰ in the case of wood-flour-filled HDPE. In the present study, the rather poor impact properties of the EFB-HDPE composites may be related to the microstructures of the composites. As indicated earlier, the irregular shape of the EFB fillers play a significant role in determining the properties of the composites. Although energy-absorbing mechanisms, such as pull-out and debonding, do operate in the composite system, as indicated from SEM study,^{17,18} their efficiencies in contributing towards the toughness enhancement of the composites were restricted by the existence of fiber bundles. In addition to that, EFB with polar hydroxyl groups (contributed by lignin, hemi cellulose, and cellulose) could not form a good interfacial bonding with nonpolar HDPE matrix. This type of incompatibility produce an adverse effect on the toughness of the composite as it would ease the crack propagation throughout EFB-HDPE interfacial region. The discussion on the mode of failure in the forthcoming section will reveal that the extensive fiber bundle (of diameter in the range of 50–250 μm) pull-out, instead of individual fiber pull-out, has resulted in the embrittlement of the composites.

In the following section, the discussion will focus on the effect of chemical treatments on the mechanical properties of EFB-HDPE composites. The application of 2 types of coupling agents and compatibilizers on HDPE composites with 40 wt % EFB fillers will be reported. This composites

was chosen for two main reasons, as follows: first, to provide a general trend on the effect of chemical treatment on the mechanical properties; second, from our previous investigation,^{17,18} EFB-HDPE composites with 40 wt % EFB loading have displayed a reasonably good balance of processability and mechanical properties.

Effect of Coupling Agents

Figure 5 depicts the effect of coupling agent loading on the tensile modulus of EFB-HDPE composites. It can be seen that for both 3-APM and 3-APE, the modulus increased significantly with the addition of 1% coupling agent. Riley et al.²¹ have noted that the three main factors affecting the composites' modulus were filler modulus, filler loading, and filler aspect ratio. High stiffness composite requires filler particles of high modulus and high aspect ratio (the ratio of the major to the minor dimension of a particle), and preferably at high filler loading. Since all of those factors have been kept more or less constant in the present study, it can be inferred that the presence of coupling agents have led to a significant improvement in the filler-matrix interfacial bonding. This will obviously results in an increase in the efficiency of stress transfer from the matrix to the filler, which consequently gives rise to higher modulus. However, further addition of both coupling agents has not produced an upward trend in the modulus. While with the modulus values stabilized in the case of 3-APM, some reduction in tensile modulus was observed for 3-APE. The slightly higher modulus values for the 3-APM treated EFB-HDPE may be attributed either to a better interaction between the reactive groups in 3-APM with the constituents phases or higher

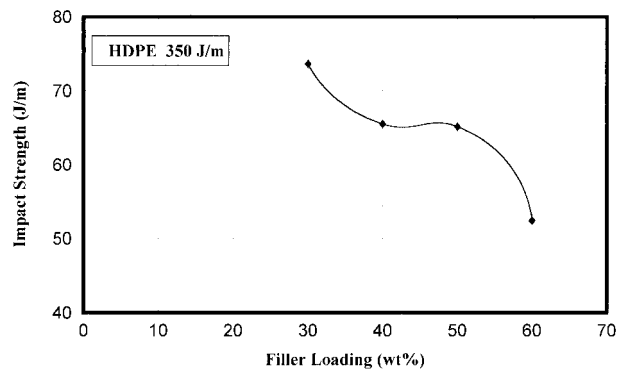


Figure 4 Impact strength as a function of filler loading for EFB-HDPE composites.

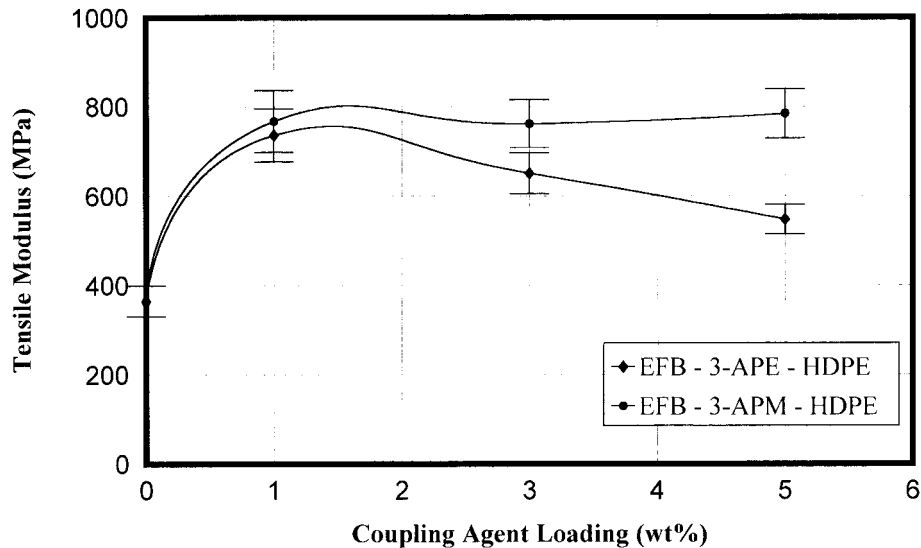


Figure 5 Effect of silane coupling agent on the tensile modulus of EFB-HDPE composites.

flexibility incurred by ethoxy groups (which consists of 2 carbon atoms) for 3-APE as compared to 3-APM, which consists of methoxy groups (1 carbon atom).

The effect of 3-APM and 3-APE on the tensile strength of HDPE composites is illustrated in Figure 6. Unlike tensile modulus, the incorporation of both types of coupling agents has not produced any significant effect on the tensile strength. This seems to indicate that the improvement in the filler-matrix interaction is still not capable of

overcoming the main problem associated with the filler geometry, as discussed earlier, with the untreated EFB-HDPE composites. The irregular shapes of the EFB coupled with its strong tendency to bundle together have suppressed the ability of the filler to support stresses transmitted by the HDPE matrix, even in the presence of coupling agent. This will be exemplified further by the evidence from the morphological study later.

Application of both 3-APM and 3-APE have deteriorated the ductility of the EFB-HDPE com-

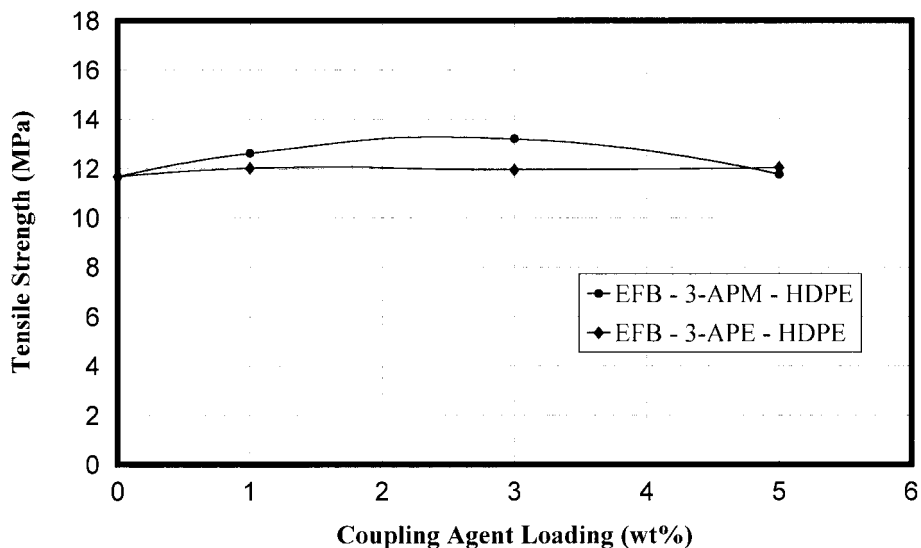


Figure 6 Variation of tensile strength with coupling agent concentration for EFB-HDPE composites.

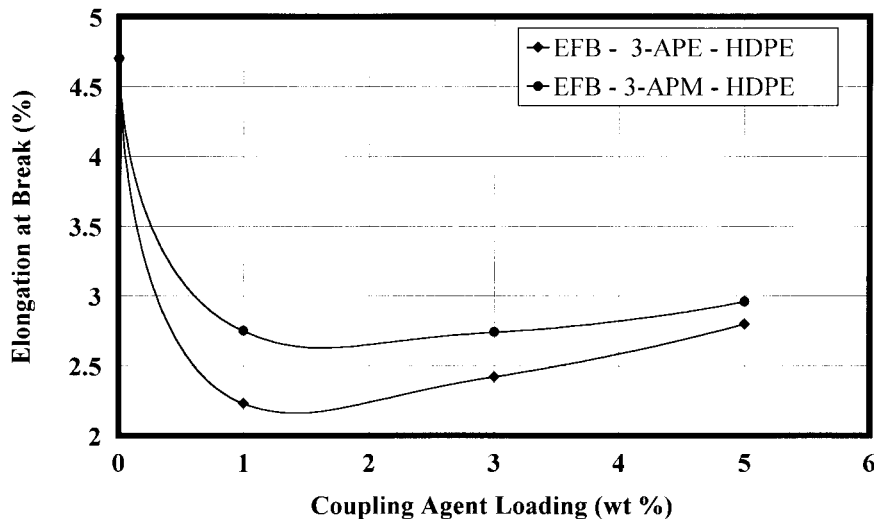


Figure 7 Elongation at break as a function of silane coupling agent for EFB–HDPE composites.

posites, as shown in Figure 7. The considerable reduction in EB from about 5% (untreated system) to 2.5% with the incorporation of just 1% coupling agent clearly indicates that the improvement in the filler–matrix adhesion is giving a positive contribution for the enhancement of stiffness but not strength and ductility of the EFB–HDPE composites. This may again be related to extensive fiber bundles, which reduced the deformability of the HDPE matrix and, hence, embrittled the composites. A similar effect has been reported by Raj et al.⁷ in their study on wood fiber-filled polyethylene composites.

The effect of coupling agents on the impact properties of EFB–HDPE composites is depicted in Figure 8. Both 3-APM and 3-APE treated composites seem to display a similar trend in their impact strength. A significant enhancement in the impact strength was initially observed with 1% concentration of coupling agent, followed by a stable impact strength at a higher coupling agent concentration. The improvement in the filler–matrix adhesion, which has produced a pronounced effect on the tensile modulus shown earlier, has resulted a similar effect on the impact property of EFB–HDPE composites.

Effect of Compatibilizers

Figure 9 shows the variation in tensile modulus with the incorporation of two types of compatibilizers, that is, PPAA and PPEAA. As expected,

the modification of the filler–matrix interfacial bonding through the presence of both compatibilizers has produced a significant effect on the tensile modulus of EFB–HDPE composites. In both cases, the modulus increases steadily with the increasing concentration of compatibilizers. At 5% loading for instance, the modulus of treated composites has doubled that of untreated composites. This may be attributed to the improvement in the filler–matrix bonding, which leads to an increase in the efficiency of stress transfer from the matrix to the filler phases. The slightly higher modulus of the PPEAA-treated EFB–HDPE composites, as compared to the PPAA-treated counterpart, seems to indicate that the presence of 19% of non-polar ethylene groups in PPEAA has resulted in a better compatibility between HDPE and PPEAA.

The effect of PPAA and PPEAA on the tensile strength of EFB–HDPE composites is shown in Figure 10. In spite of the improvement in the modulus as discussed above, the incorporation of both compatibilizers fail to impart any positive effect on the tensile strength of the composites. This again supports our earlier claim that with the irregular-shaped filler geometry of EFB, any improvement in the filler–matrix interaction, even in the presence of coupling agent or compatibilizer, is not capable of enhancing the ultimate property, such as tensile strength of the composites. In addition, the incorporation of compatibilizer also fails to improve the fiber dispersion. The existence of fiber bundles, whereby the individual

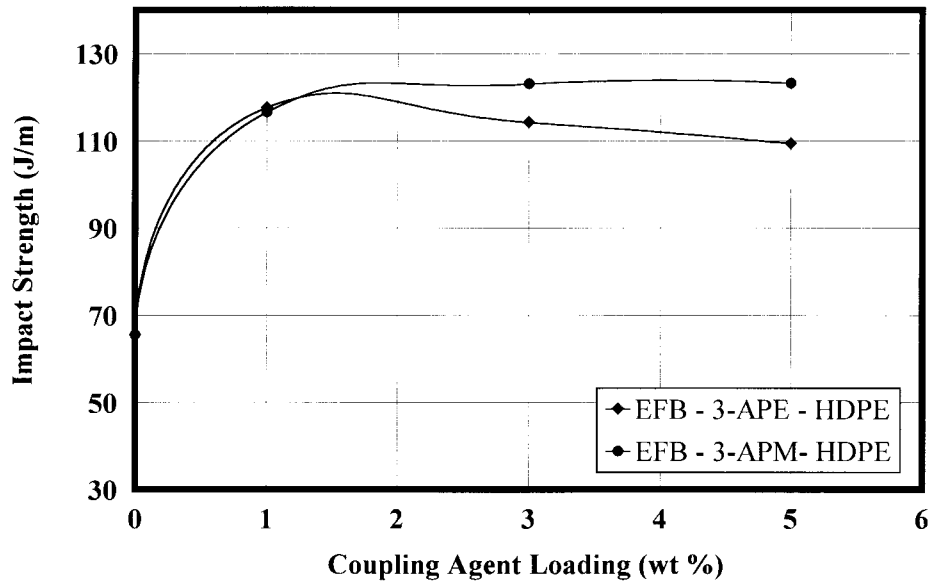


Figure 8 Impact strength versus coupling agent concentration for EFB-HDPE composites.

fiber is still banded together, is prevalent, as indicated by the SEM micrographs. This subject will be elaborated later in this article.

Figure 11 shows the relationship between elongation at break (EB) with the loading of compatibilizers. The slight reduction in the values of EB with increasing compatibilizers loading again reflect the fact that the improvement in the filler-matrix interaction cannot be exploited to enhance

the ductility or EB of the EFB-HDPE composites. A similar trend has also been reported by Ahmad Fuad et al.¹⁹ in the case of rice-husk-ashes-filled polypropylene.

The effect of PPAA and PPEAA on the impact strength of HDPE composites is shown in Figure 12. The incorporation of both types of compatibilizers has no significant effect on the impact strength of the composites. Thus, it may be in-

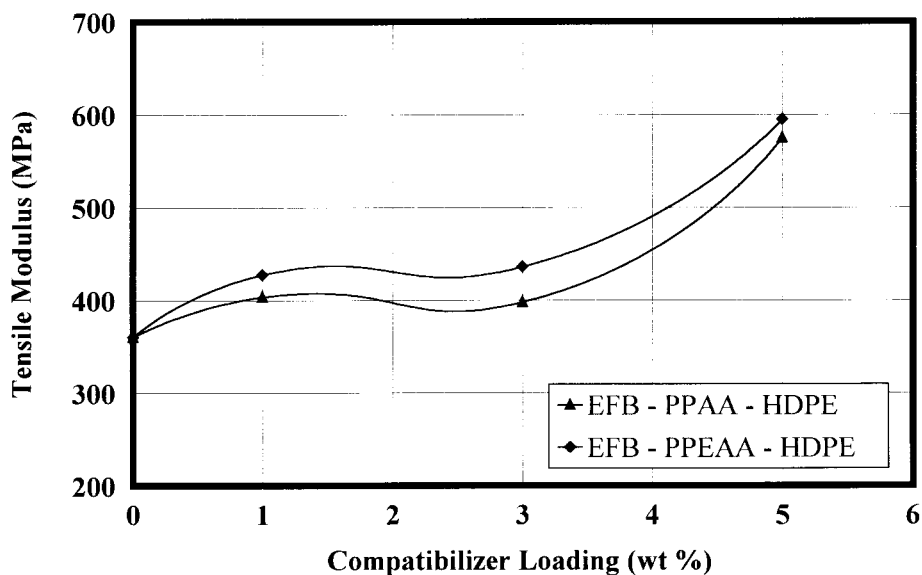


Figure 9 Effect of acrylic acid based compatibilizers on the tensile modulus of EFB-HDPE composites.

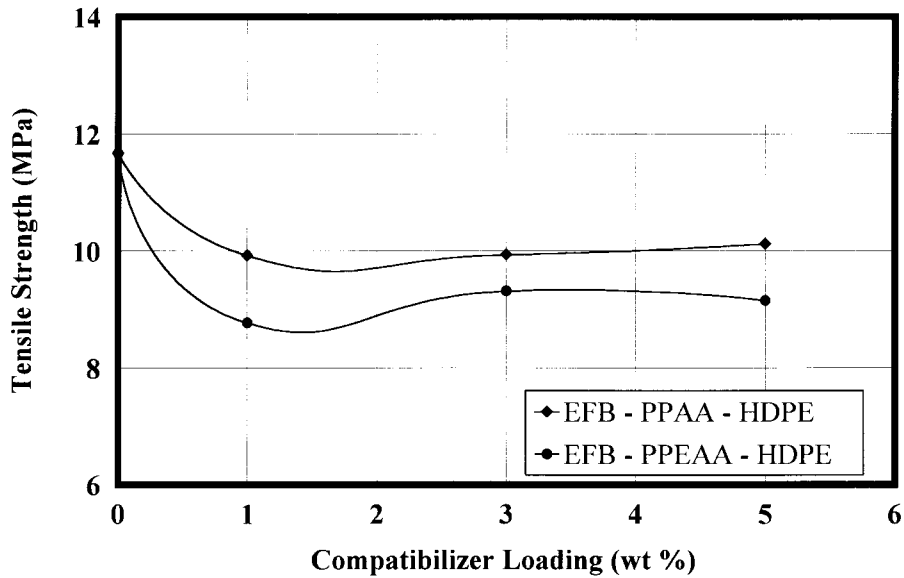


Figure 10 Tensile strength as a function of acrylic acid based compatibilizers for EFB-HDPE composites.

ferred that the improvement in the interfacial bonding between EFB and HDPE through compatibilization is not capable of creating better resistance against crack propagation during the fracture process. On the contrary, the trend observed with the PPAA compatibilized composite system seems to indicate that the presence of PPAA has embrittled the composites.

Comparison Between the Effect of Coupling Agent and Compatibilizer

In the preceding section, discussion has been focused on the effect of 2 types of coupling agents, 3-APE and 3-APM, and 2 types of compatibilizers, PPAA and PPEAA. From the various mechanical properties investigated, it can be concluded that

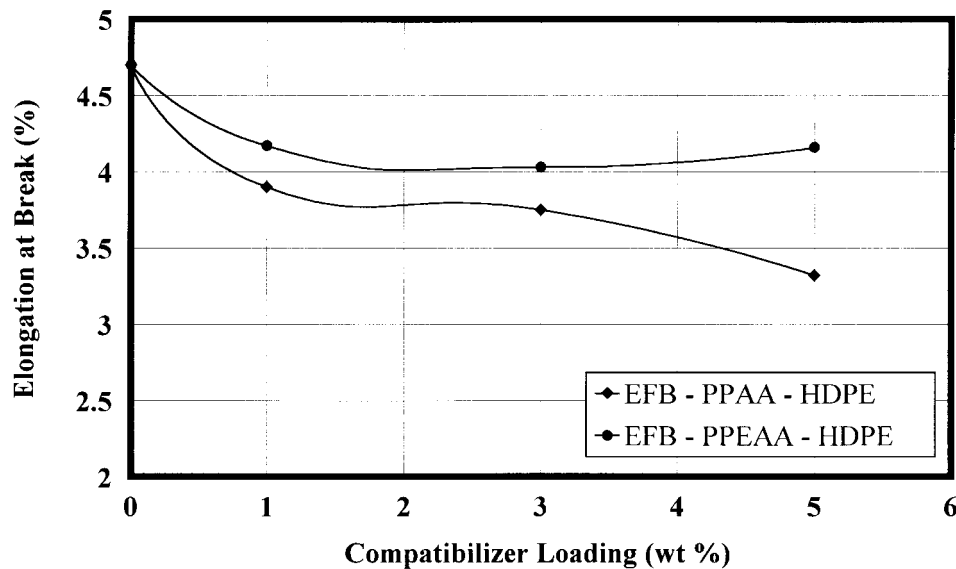


Figure 11 Elongation at break versus compatibilizer concentration for EFB-HDPE composites.

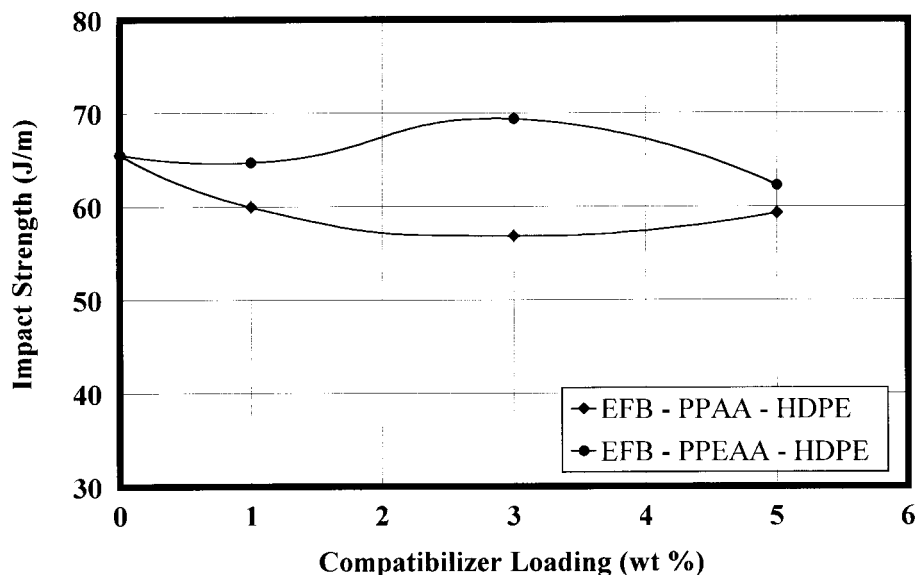


Figure 12 Effect of compatibilizer concentration on the impact strength of EFB-HDPE composites.

in the case of coupling agent, the incorporation of 3-APM has produced composites with superior properties as compared to 3-APE. This may be attributed to the better interaction between the chemical reactive groups in the 3-APM with both EFB and HDPE. As for the compatibilized composite systems, the enhancement in the mechanical properties of the PPEAA-treated composites gives a clear indication that the presence of ethylene group has promoted a better interaction and compatibilization between PPEAA and HDPE matrix.

In this section, discussion will be emphasized on the comparison between EFB-PPEAA-HDPE and EFB-3-APM-HDPE composites. The purpose is to identify the relative efficiency of two different types of chemical treatment in improving the mechanical properties of the composites.

Figure 13 shows the dependence of tensile modulus on the loadings of both 3-APM and PPEAA. Initially, the incorporation of 1% of both chemicals has increased the modulus of HDPE composites, albeit higher enhancement was observed in the case of 3-APM-treated composites. While further addition of 3-APM does not seem to increase the modulus, a different trend was displayed by the PPEAA-treated composites. The tensile modulus increases steadily with increasing PPEAA concentration. This may be related to the different nature of interaction between PPEAA with EFB and HDPE phases. These observations indicate that the efficiency of chemical treatment in improving

the stiffness of EFB-HDPE composites is dependent on the types and loading of the chemicals used. Unlike 3-APM, which is relatively simple, and low-molecular-weight chemicals, PPEAA consists of long-chain polymeric materials. The presence of ethylene group is believed to play a significant role in promoting a good interaction and compatibility with the HDPE matrix. The acrylic acid groups in PPEAA, on the other hand, is capable of forming chemical linkages with the lignocellulosic components, that is, the hydroxyl groups in the EFB. Thus, as the amount of PPEAA is increased, there are more chemical groups, that is, acrylic acids and ethylene groups, to interact with EFB and HDPE phases, respectively. Figure 14 illustrates the hypothetical reaction of lignocellulosic EFB filler with PPEAA. The condensation reaction taking place between the carboxyl groups in PPEAA and hydroxyl groups in EFB is believed to result in the formation of ester linkages between EFB and PPEAA.

The comparative effect of 3-APM and PPEAA on the tensile strength of EFB-HDPE composites is shown in Figure 15. In spite of the slightly different trend exhibited by both treated composites, it is quite clear that both types of chemical treatments have failed to produce a profound effect on the tensile strength of the composites. These results provide a good indication that as far as the microstructure of the composites is concerned, the contribution of filler geometry is far more prominent than the interfacial bonding between the EFB and HDPE.

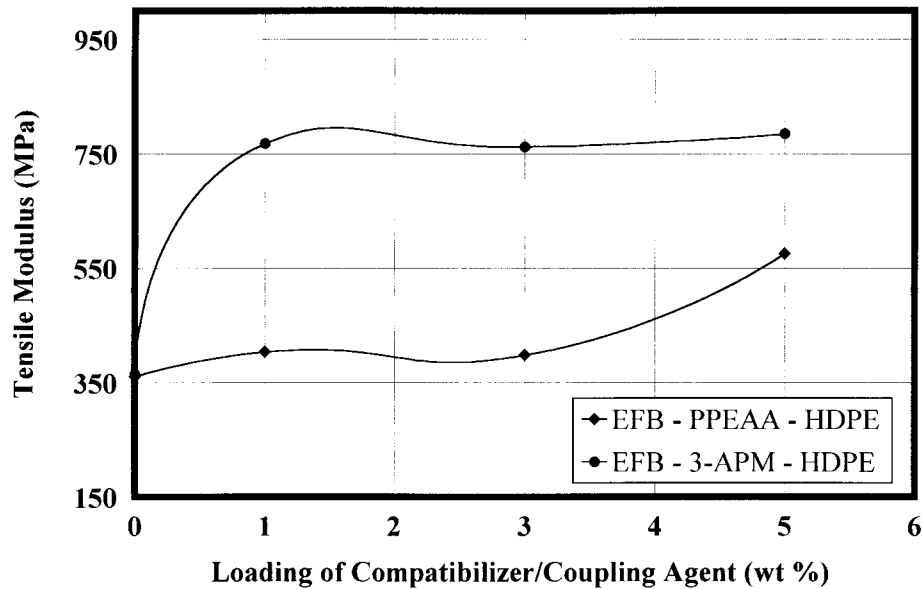


Figure 13 Comparative effect of 3-APM and PPEAA on the tensile modulus of EFB-HDPE composites.

In spite of some improvement in the bonding, which was achieved via the chemical treatment, the irregular filler geometry has upset the efficiency of stress transfer from the matrix to the filler. This has obviously resulted in the poor tensile strength of the treated EFB-HDPE composites.

In the case of the elongation at break (EB), both treated composites seem to display a similar downward trend with increasing loading of either 3-APM or PPEAA (Fig. 16). The slightly higher EB of PPEAA-treated composites can be related to the long-chain nature of PPEAA, which may

impart some flexibility to the composites. This agrees well with the lower modulus values of EFB-PPEAA-HDPE composites as compared to that of EFB-3-APM-HDPE composites.

Figure 17 shows the dependence of impact strength on the loading of both 3-APM and PPEAA. While the impact strength remains constant, regardless of the amount of PPEAA used, a different trend was observed in the case of 3-APM-treated composites. A sharp increase in the impact strength was noted with 1% 3-APM, followed by more or less constant values at higher loading. The superior impact properties of the EFB-3-APM-HDPE composites indicates that higher forces are needed to fracture the composites. This gives a clear indication that the significant improvement in the interfacial bonding has increased the energy-absorbing capabilities of the composites. Although fiber bundle pull-out and debonding is believed to be still prevalent in the material, a greater resistance to crack propagation can be expected to occur during the fracture process.

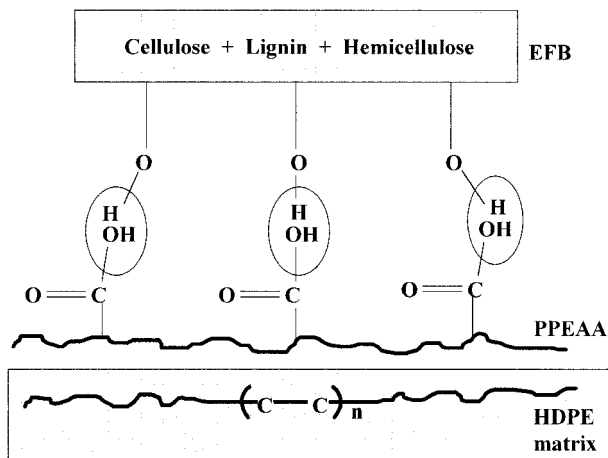


Figure 14 Hypothetical condensation reaction, which leads to the esterification of the lignocellulosic components of EFB.

Modes of Failure

SEM was employed to obtain some qualitative evidences on the bonding quality between the EFB and HDPE and the dispersion of EFB in the HDPE matrix. This information is essential since the microstructure of the composites is known to

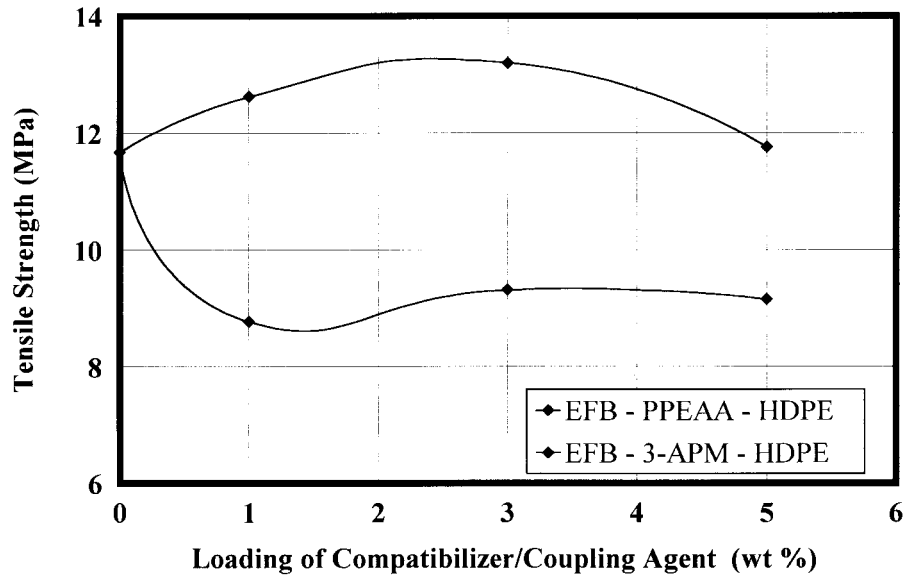


Figure 15 Tensile strength versus 3-APM and PPEAA loading for EFB-HDPE composites.

play a significant role in determining the mechanical properties of filled composites.²² As the EFB were cut and ground using a laboratory-scale grinder without any heat, pressure, or chemical treatment, as in the thermomechanical pulping process, EFB has a strong tendency to exist in the form of fiber bundles. In the present EFB-HDPE composites, these fiber bundles have been observed by SEM to be distributed unevenly throughout the matrix. Figure 18 shows a typical

SEM micrograph of HDPE filled with 40 wt % EFB. At a higher magnification, fiber bundles of diameter as high as 200 μm can be observed on the crack plane, as shown in Figure 19. The presence of such fiber bundles can be expected to give a detrimental effect to the tensile and impact strength of the HDPE composites. The small-scale plastic deformation process, which resulted in the matrix tearing, as shown by SEM micrograph in Figure 20, also failed to retain the ductility of the

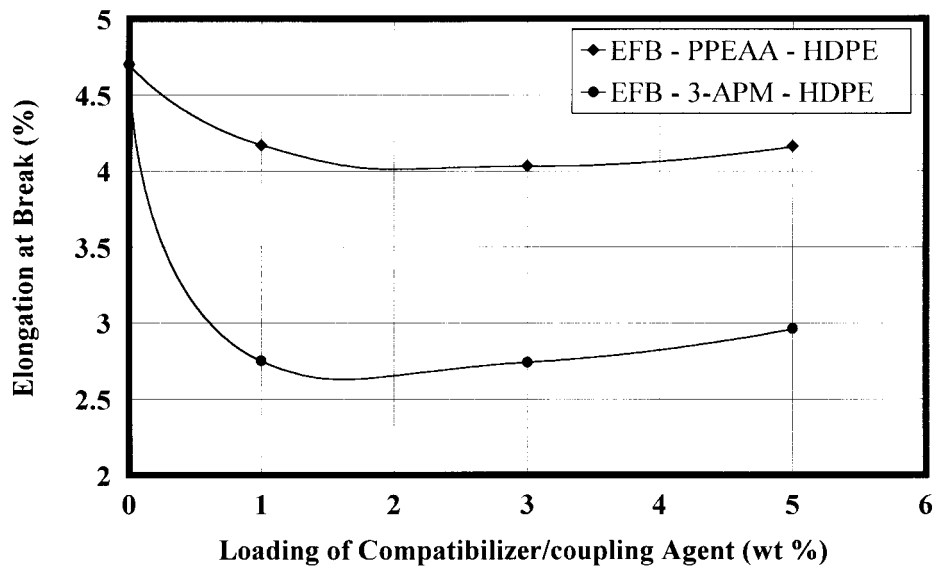


Figure 16 Comparative effect of 3-APM and PPEAA on elongation at break for EFB-HDPE composites.

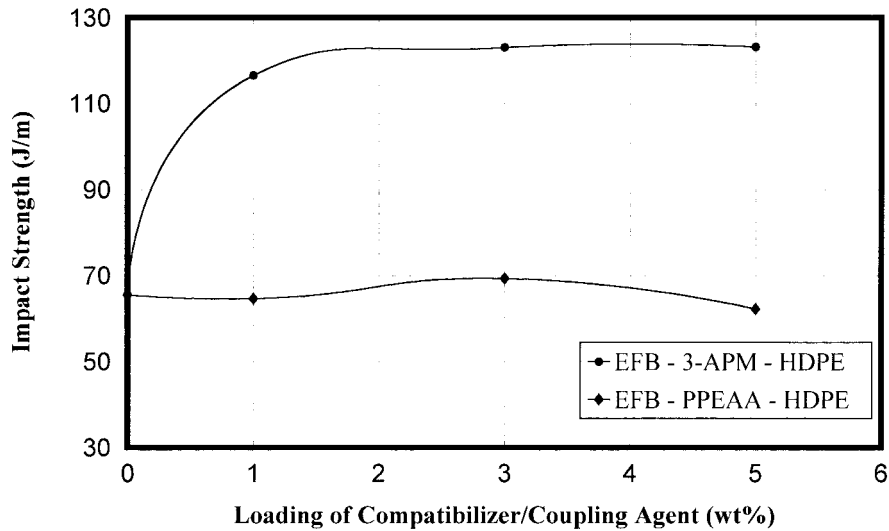


Figure 17 Variation of impact strength with 3-APM and PPEAA loading for EFB-HDPE composites.

EFB-HDPE composites. A significant improvement in strength could be achieved if the fibers were to be dispersed uniformly (i.e., much smaller in diameter, and hence a higher aspect ratio) in the matrix. The holes shown by the SEM micrograph in Figure 21 provide a good indication for the occurrence of fiber bundle pull-out. The amount of energy associated with the fiber bundle pull-out could be expected to be much lower than those related to the individual fiber pull-out, as normally observed in the case of short glass or carbon-fiber-reinforced thermoplastic composites.^{23,24} This may explain the poor impact properties of the EFB-HDPE composites. A similar observation has been encountered by Folkes²⁵ in the case of short carbon-fiber-reinforced nylon 6.6

when both nylon 6.6 and carbon fibers were directly injection-molded without undergoing the compounding stage.

As mentioned in the introduction, one of the major problems associated with the lignocellulosic thermoplastic composites is the poor compatibility or interaction between the constituent phases. The polarity of lignocellulose EFB fillers is obviously not capable of forming a good filler-matrix interaction with the nonpolar HDPE. A trace of a fiber surface with its bordered pits printed in the matrix, depicted in Figure 22, clearly indicates that the EFB fillers were merely imbedded in the HDPE matrix without a good bonding at the interface. Some evidence of fiber bundle debonding

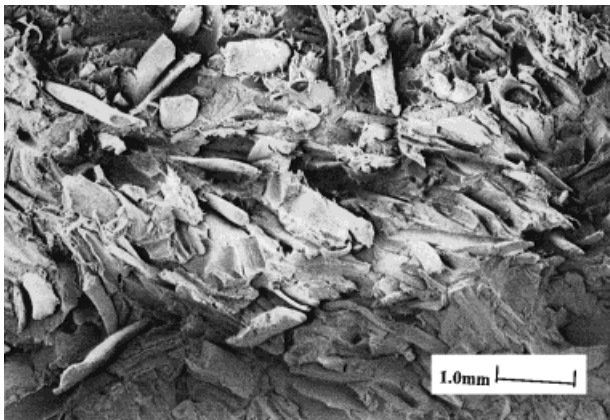


Figure 18 SEM micrograph of untreated EFB-HDPE composites (40 wt % filler loading).

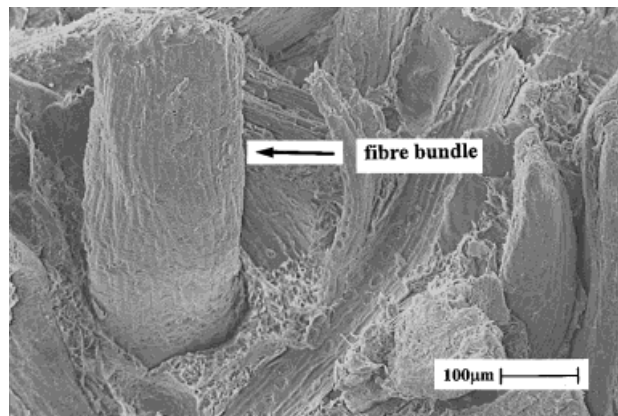


Figure 19 SEM micrograph indicating the presence of fiber bundles in untreated EFB-HDPE composites.

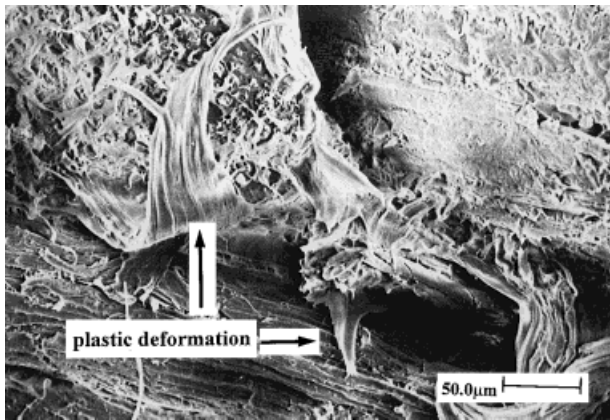


Figure 20 SEM micrograph showing plastic deformation in the form of matrix tearing observed in untreated EFB-HDPE composites.

could also be observed from the SEM micrograph depicted in Figure 23.

Figure 24 shows the SEM micrographs of EFB-HDPE treated with coupling agent, that is, 3-APM. It can be seen that the poor dispersion and the wide range of particle size distribution of EFB fillers still exist in the composite product. In the case of PPEAA-treated EFB-HDPE composites, the SEM micrograph shown in Figure 25 provides a good indication on the improvement of interfacial bonding between the constituents phases. The EFB filler can be seen to be quite well bonded to the HDPE matrix. This is in contrast to the previous SEM micrograph (Fig. 23) of untreated EFB-HDPE composites. However, the extensive fiber

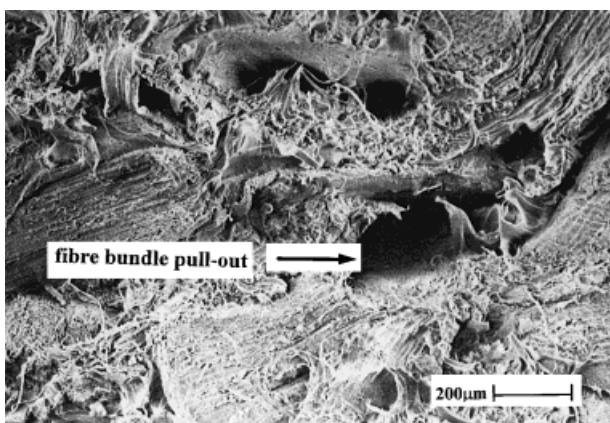


Figure 21 SEM micrograph indicating the occurrence of fiber bundle pull-out in untreated EFB-HDPE composites.

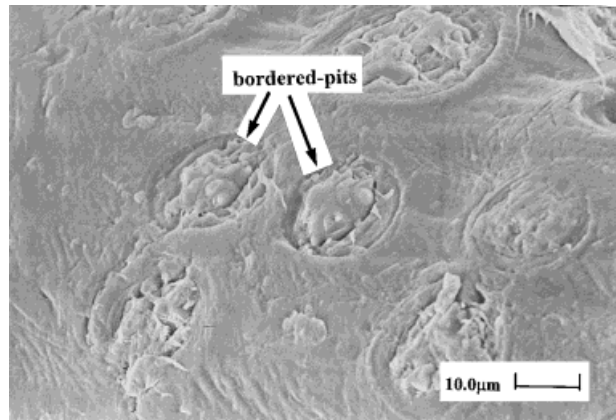


Figure 22 A trace of a fiber surface, with its bordered pits, indicating poor interfacial bonding between EFB and HDPE.

bundle pull-out, which is dominant on the fracture plane, as illustrated in Figure 24, is believed to be more detrimental to the mechanical performance of the composites. The integrity of the fiber bundle is still intact, that is, the holocellulose that constitutes about 65% of the EFB is well binding by the lignin. This will provide hinderance for the coupling agent or compatibilizer to form an efficient interaction with the holocellulose fibers. Thus, it is not suprising to observe a poor enhancement in the tensile strength of the composites, even after surface treatment of the EFB filler has been carried out.

Figure 26 shows a schematic representation of the mode of failure of a typical short fiber-reinforced thermoplastic composite. During the frac-

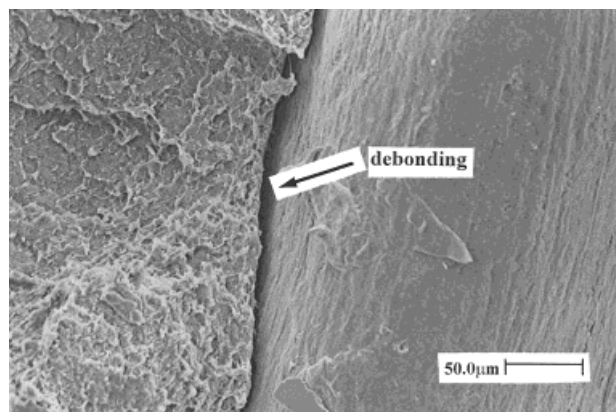


Figure 23 SEM micrograph indicating poor interfacial bonding between EFB and HDPE for the untreated composites.

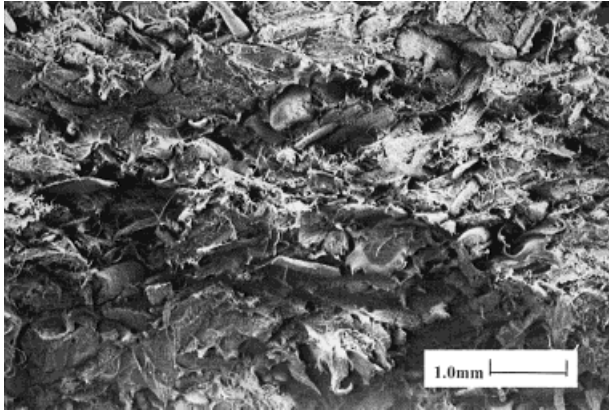


Figure 24 SEM micrograph of EFB-HDPE composites treated with 3-APM silane coupling agent.

ture process, as crack propagation takes place, several types of energy-absorbing mechanisms, which are either matrix-related (such as crazing and shear yielding) and/or fiber-related (such as pull-out, debonding, fiber fracture) will contribute to the toughness of the composites. In addition, the dispersion of the fibers will allow better interaction between the fibers and the matrix. This will obviously increase the efficiency of stress transfer from the matrix to the fibers. Thus, enhancement in the stiffness and strength of the short fiber composites could also be realized.

In the case of the composites, the existence of fiber bundles in the final molded composite product is believed to give a significant contribution to the mode of failure of the composites. The schematic representation shown in Figure 27 illustrates the failure process proposed based on the qualitative evidences from the SEM studies. It is

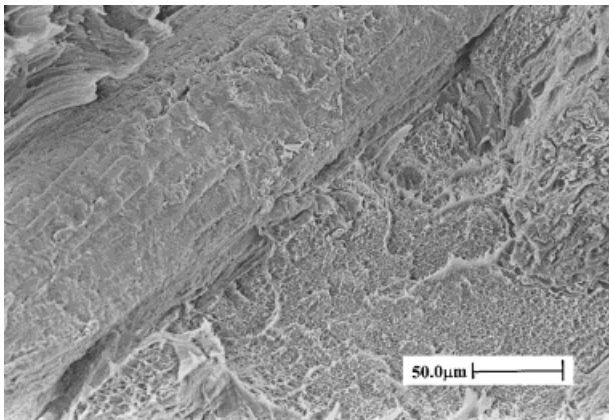


Figure 25 SEM micrograph of EFB-HDPE composites treated with PPEAA.

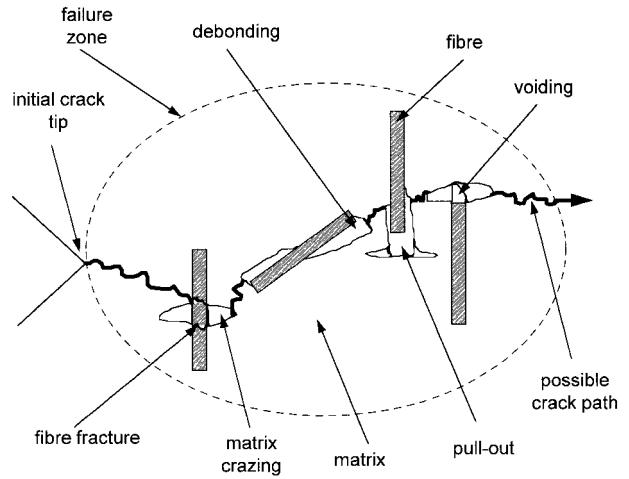


Figure 26 A schematic representation of the mode of failure in short fiber-reinforced thermoplastics.

quite obvious that with the particle size in the range of 270–500 μm pull-out is most likely to be the dominating mechanisms operating in the composites. However, due to the nature of the lignocellulosic filler, which tends to exist in the form of bundles, the strong inclination for fiber bundle pull-out to take place is inevitable. Thus, their energy-absorbing capabilities are not going to be as efficient as that of individual fiber pull-out; and, consequently, the toughness enhancement is limited. In addition, the presence of fiber bundles will also reduced the efficiency of stress transfer from the matrix to the holocelleulose fibers. This will obviously lead to poor stiffness and strength of the EFB-HDPE composites.

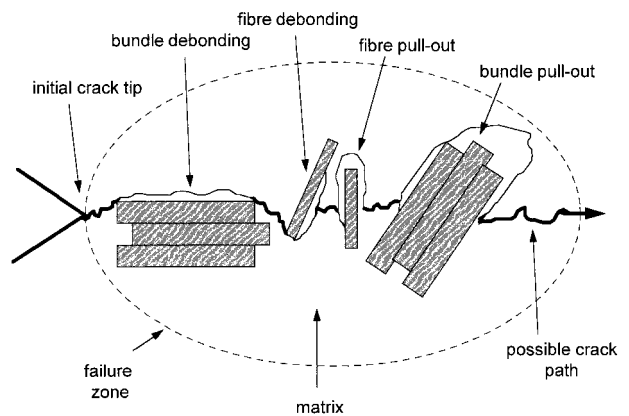


Figure 27 A schematic representation of the mode of failure for EFB-HDPE composites.

CONCLUSION

The main objective of this investigation is to gage the possibility of utilizing the low-cost EFB as alternative lignocellulosic filler materials in HDPE. The conclusions based on this study are as follows.

1. The incorporation of the EFB into HDPE matrix has resulted in the moderate improvement in the tensile modulus of the composites. However, this is at the expense of reduction in tensile strength, elongation at break, and impact strength. The poor performance in the ultimate properties has been attributed to the poor filler–matrix interaction or compatibility and also to the nature of EFB fillers, which has a strong tendency to exist in the form of bundles.
2. In the quest to find an optimum chemical treatment method for the EFB–HDPE composites, 2 types of coupling agent, 3-APM and 3-APE, and 2 types of compatibilizer, PPAA and PPEAA, were applied. In general, 3-APM and PPEAA has to been proven to be more effective in improving the filler–matrix interaction. In the case of PPEAA, the presence of ethylene groups is believed to be responsible in promoting better compatibility between EFB and HDPE. However, the efficiency of either 3-APM or PPEAA in enhancing the filler–matrix interaction is hindered by the presence of lignin surrounding the holocellulose fibers. Thus, only slight improvement was observed in the mechanical performance of the treated composites.
3. The nature of EFB, which has a strong tendency to exist in the form of bundles, even after chemical treatment with either 3-APM or PPEAA, has a strong influence on the efficiency of stress transfer and mode of failure of the composites. The relatively poor stress transfer from the matrix to the filler has resulted in a moderate improvement in the stiffness of the composites. Fiber bundle pull-out is reckoned to be the main energy-absorbing mechanism operating in the EFB–HDPE composites.

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REFERENCES

1. M. Xanthos, *Plast., Rubber Process. Appl.*, **3**, 223 (1983).
2. C. Klason, J. Kubat, and H. E. Stromvall, *Int. J. Poly. Mater.*, **10**, 159 (1984).
3. H. Dalvag, C. Klason, and H. E. Stromvall, *Int. J. Poly. Mater.*, **11**, 9 (1985).
4. C. A. Cruz-Ramos, in *Mechanical Properties of Reinforced Thermoplastics*, D. W. Clegg and A. A. Collyer, Eds., Elsevier, London, 1986.
5. R. G. Raj, B. V. Kokta, and C. Danault, *Int. J. Poly. Mater.*, **12**, 239 (1989).
6. R. G. Raj, B. V. Kokta, F. Dembele, and B. Sanschagrain, *J. Appl. Polym. Sci.*, **38**, 1987 (1989).
7. R. G. Raj, V. Kokta, G. Groleau, and C. Daneault, *Plast., Rubber Process. Appl.*, **11**, 215 (1989).
8. L. Yam, B. K. Gogoi, C. C. Lai, and S. E. Selke, *Polym. Eng. Sci.*, **30**, 693 (1990).
9. G. E. Myers, I. S. Chahyadi, C. A. Coberly, and D. S. Ermer, *Int. J. Poly. Mater.*, **15**, 21 (1991).
10. G. E. Myers, I. S. Chahyadi, C. Gonzales, C. A. Coberly, and D. S. Ermer, *Int. J. Poly. Mater.*, **15**, 171 (1991).
11. J. M. Felix and C. Gatenholm, *J. Appl. Polym. Sci.*, **42**, 609 (1991).
12. J. X. Rietveld and M. J. Simon, *Int. J. Poly. Mater.*, **18**, 213 (1992).
13. K. Joseph, S. Thomas, and C. Pavithran, *Polymer*, **37**, 5139 (1996).
14. M. J. Zaini, M. Y. A. Fuad, Z. Ismail, M. S. Mzansor, and J. Mustafah, *Polym. Int.* **40**, 51 (1996).
15. R. N. Kumar, H. D. Rozman, A. Abusamah, and T. H. Chin, *National Seminar on Utilisation of Palm Tree and Other Palms*, Forest Research Institute of Malaysia (FRIM), 1994.
16. Y. Kobayashi, H. Kamishima, I. Akamatsu, A. H. H. Hassan, M. Husin, K. Hassan, and M. N. M. Yusoff, *Workshop Proc. Palm Oil Res. Inst. Malaysia*, **11**, 7 (1987).
17. H. D. Rozman, H. Ismail, R. M. Jaffri, A. Aminullah, and Z. A. Mohd Ishak, *Int. J. Poly. Mater.*, to appear.
18. H. D. Rozman, H. Ismail, R. M. Jaffri, A. Aminullah, and Z. A. Mohd Ishak, *Polym. Plast. Tech. Eng.*, to appear.
19. M. Y. Ahmad Fuad, R. Shukor, Z. A. Mohd Ishak, and A. K. Mohd Omar, *Plast. Rubber Comp. Appl.*, **21**, 225 (1994).
20. D. M. Bigg, *Polym. Comp.* **8**, 115 (1987).
21. A. M. Riley, C. D. Paynter, P. M. McGenity, and J. M. Adams, *Plast., Rubber Process. Appl.*, **14**, 85 (1990).
22. J. Karger-Kocsis, in *Application of Fracture Mechanics to Composite Materials*, K. Friedrich, Ed., Elsevier, Amsterdam, 1989.
23. Z. A. Mohd Ishak and J. P. Berry, *Polym. Eng. Sci.*, **33**, 1483 (1993).
24. T. Czigany, Z. A. Mohd Ishak, and J. Karger-Kocsis, *Appl. Comp. Mater.*, **2**, 313 (1995).
25. M. J. Folkes, *Short Fibre Reinforced Thermoplastics*, Research Studies Press (John Wiley), Chichester, UK, 1982.