The Effect of Compounding Techniques on the Mechanical Properties of Oil Palm Empty Fruit Bunch–Polypropylene Composites

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ABSTRACT: Oil palm empty fruit bunch–polypropylene (EFB–PP) composites were produced by employing 2 types of compounding techniques, that is, an internal mixer and a single-screw extruder. The mechanical and water absorption properties of both types of composites were investigated. Overall, for both types of composites, the incorporation of the EFB into PP matrix has resulted in the improvement in the tensile modulus. However, the tensile strength, elongation at break, and impact strength decreased with increasing filler loading. Poor filler–matrix interaction or compatibility and, also, the size irregularity of the EFB are believed to be responsible for the poor ultimate performance. Composites produced by an internal mixer (IM) have displayed higher tensile strength, tensile modulus, and impact strength than with those produced by extrusion (EX). The better performance has been attributed to the effectiveness of the IM, which produces better compounding and improves the wetting of the filler surface. Incorporation of compatibilizer and coupling agent, that is, Epolene wax (E-43) and 3-Aminopropyl triethoxysilane (3-APE), respectively, have produced composites with improved tensile strength for both EX and IM composites. In addition, both types of treatment have resulted in an increase in tensile modulus of EX composites and impact strength of IM composites. Water absorption tests have revealed that the presence of coupling agents and compatibilizers have affected the amount of water absorbed, especially for the 3-APE-treated EFB–PP composites. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 70: 2647–2655, 1998

Key words: oil palm empty fruit bunch; lignocellulose; composites; compounding

INTRODUCTION

The utilization of lignocellulosic material, especially wood, as a reinforcing component in polymer composites has received increased attention, particularly for price-driven, high-volume applications.[1–10] This development has been brought about because reinforcement by lignocellulosic fillers offers several advantages over their inorganic counterparts, that is, lower density, greater deformability, less abrasiveness to expensive molds and mixing equipments, and, of course, lower cost. As better understanding of the wood–plastic interaction has been gained, the move to find substitutes for wood has been stimulated, especially by the abundance of similar resources (lignocellulosic material). With the increasing pressures on the forest industries, coupled with the scarcity of natural resources, the need for the efficient use of the resources is vital, especially in the wood composite industries. Thus, this would mean better conversion; more cost-effective utilization, which is environmentally friendly; new technologies; and even the need to look for a substitute for wood.

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As mentioned earlier, lignocellulosics-derived fillers possess several advantages compared to inorganic fillers. More importantly, lignocellulosic-based fillers are derived from a renewable resource and relatively available in abundance, and the potential has not been really tapped.

There has been a growing interest on the utilization of lignocellulosic, apart from wood. These materials have been subjected to various investigations recently, either in replacing existing wood species in making conventional panel products[11] or producing plastics composites.[12–16] The increasing trend in using these nonwood-based materials has been induced by the growing demand for lightweight, high-performance materials coupled with diminishing natural fiber resources (wood in particular) and escalating costs of raw materials and energy.

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. The potential utilization of fillers that were derived from oil palm industries, that is, empty fruit bunch (EFB) and oil palm frond (OPF) for the production of high-density polyethylene (HDPE) composites have been reported by several workers.[13,17,18] The investigation has been focused on the effect of filler loadings, filler particle size distribution, and the effect of coupling agents 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.[13,17] Incorporation of coupling agents and compatibilizers in the EFB composites has also been reported to impart some improvement in properties.[18] Several studies on other lignocellulosic-filled thermoplastic composites have indicated that the mixing procedures could play an important role in determining the properties of composites.[1,19] Thus, it is the objective of the present work to investigate the effect of compounding techniques on the mechanical properties of the composites. In this study, emphasis has been given to EFB instead of OPF for 2 main reasons. From a microstructure–property relationships point of view, both EFB–HDPE and OPF–HDPE composites displayed similar trends, and their overall mechanical properties are quite comparable.[13,17] Secondly, EFB is preferable in terms of availability and cost. EFB is readily available at a typical token price of USD (US Dollar) 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.

EXPERIMENTAL

Materials

Oil palm EFB (comprised of a bunch of fibers in which the palm fruit are embedded and consisting of about 65% holocellulose and 25% lignin) was supplied by Sabutek Sdn. Bhd., Teluk Intan, Perak. The EFB used in this study was ground to the mesh size of 35–60 (270–500 μm). The polypropylene (PP) used was purchased from Polypropylene Malaysia Sdn. Bhd., Malaysia, with a melt flow index and a density of 12.0 g/10 min and 0.903 g/cm^3, respectively. Epolene E-43 and poly(propylene–acrylic acid) (PPAA) were used as compatibilizers and 3-Aminopropyltriethoxysilane (3-APE) was used as a coupling agent. Epolene E-43 (Eastman Chemical Products), a low-molecular-weight ($\bar{M}_n$ = 3900, $\bar{M}_w$ = 9100) maleated PP, has a density of 0.934 g/mL and acid number of 47 was kindly supplied by Suka Chemicals (M) Sdn. Bhd., Selangor, Malaysia. PPAA and 3-APE were supplied by Polysciences Incorporation, USA.

Filler Preparation

EFB filler was delivered in the form of long strands of fibers. The fibers were ground into small particles. An Endecotts sieve was used to separate the particles into different sizes. The filler size used in this study was of mesh 35–60, that is, 270–500 μm.

Filler Treatment

Applications of the coupling agent (3-APE) and compatibilizers (PPAA and E-43) to the PP–EFB composites differ from one another due to the nature of the chemical themselves. 3-APE was delivered in liquid form; and, prior to application, the coupling agent was diluted in ethanol to make 20% solution. The amount of the coupling agent used in this study was about 1% by weight of EFB filler. EFB filler was charged into a bench-top tumbler mixer, and the 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 E-43 and PPAA compatibilizers were used as delivered and added directly to the EFB–PP mix (3% of the weight of PP). The amount of compatibilizer used was earlier found to be suitable to obtain optimum properties of the wood flour-filled PP composites.[20]

Compounding and Processing

Compounding of the materials were carried out using the following 2 methods: an internal mixer and a single-screw extruder. With the internal mixer, the compounding of untreated and treated EFB–PP was carried out using a Haake Rheocord System consisting of a Haake Rheodrive 5000 (drive unit) and a Haake Rheomix 600 with a roller blade (mixer). The mixing was carried out at 180°C with a rotor speed of 25 rpm. With the single-screw extruder (Betol Extruder Model 116), the compounding was carried out at a screw speed of 20 rpm; extruder temperatures were set at 160°C (zone 1), 170°C (zones 2 and 3) and 180°C (die). The compounds were extruded through a single 3-mm rod die and pelletized. The loading of the EFB for both methods of compounding were varied from 0 to 60% by weight of the filler.

The extrudates were hot-pressed in a mold of internal dimensions of 18.5 × 13.5 × 0.5 cm under a pressure of 4 MPa. Hot press procedures involved preheating at 100°C, followed by heating at 180°C and subsequent cooling under pressure. The total molding time was 15 min.

Testing

The sheet produced was cut into the following 2 types of test samples: 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 × 1.5 × 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 ± 2°C and 55 ± 5% RH for approximately 72 h before being tested.

Water Absorption Test

EFB–PP samples were immersed in distilled water at 30°C. The water absorption were determined by weighing the specimens at regular intervals. A Mettler balance type AJ150 was used, with a precision of 1 mg. The water content at any time t, M_t, was calculated by

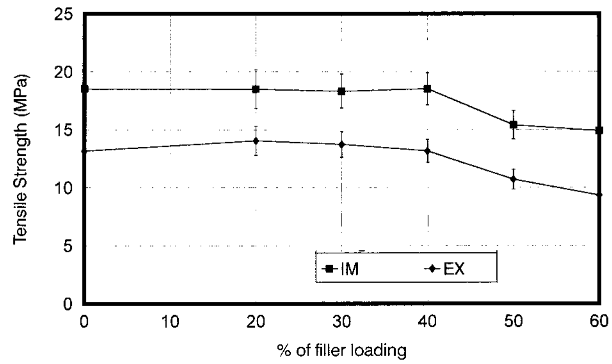


Figure 1 The effect of filler loading on the tensile strength for IM and EX composites.

$$M_t = \frac{W_w - W_d}{W_d}$$

where W_d and W_w are original dry weight and weight after exposure, respectively.

Morphological Study

The fracture surface of the composites (40% EFB, treated with 3-APE) from the tensile test were investigated with a Leica Cambridge S-360 scanning electron microscope. The objective is to get some information regarding fiber dispersion and bonding quality between fiber and matrix and to detect the presence of microdefects, if any. The fracture ends of the specimens were mounted on an aluminum stub and sputter-coated with a thin layer of gold to avoid electrostatic charging during examination.

RESULTS AND DISCUSSION

Effects of Compounding Method

In the following discussion, the codes IM and EX composites will be used to denote the EFB–PP composites that have been compounded using the internal mixer and extrusion techniques, respectively. Figure 1 shows the effect of filler loading on the tensile strength of EFB–PP composites. It appears that both types of composite show a decreasing trend as the filler loading increases. This is in agreement with the trend observed in other lignocellulosic-filled composites.[21,22] Unlike fibers

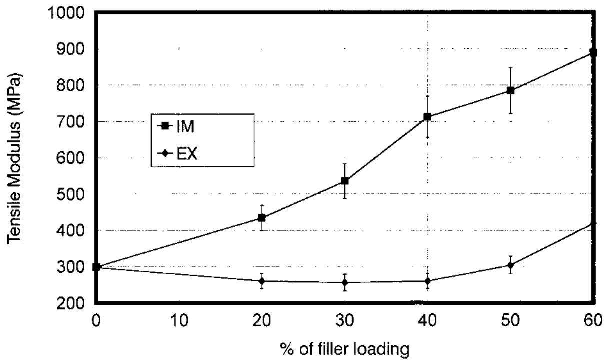


Figure 2 The effect of filler loading on the tensile modulus for IM and EX composites.

that have a uniform cross section and a relatively high aspect ratio (that is, the length to diameter ratio, l/d), for irregular-shaped fillers, such as EFB, their capability to support stress transmitted from the thermoplastic matrix is rather poor. Thus, the strength enhancement in the filled composite is, in general, much lower than that of fiber-reinforced systems. The results from the IM are rather poor. Thus, the strength enhancement in the filled composite is, in general, much lower than that of fiber-reinforced systems. The results reveal that IM composites yield a higher strength than the EX composites. This indicates that the mode of mixing plays an important role in determining the tensile strength of a composite. As the degree of compounding and distribution of fillers in the polymer matrix determine the extent of the formation of interfacial region between these 2 components, it is believed that a better transfer of stress occurred in the IM composites as opposed to the EX composites.

The variation of tensile modulus with respect to filler loading is evaluated, as shown in Figure 2. The tensile modulus for both IM and EX composites increases with an increasing filler loading. This behavior is consistent with the earlier study on PP–wood flour composites.[22] The results suggest that, irrespective of compounding techniques, EFB filler is able to impart greater stiffness to the PP composites.

It is interesting to note that IM composites display a markedly higher modulus than the EX composites. For composites with 60% filler loading, for instance, the modulus for IM composites reach as high as approximately 900 MPa as compared to about 400 MPa for EX composites. Thus, it can be inferred that the inherent stiffness of the EFB fillers can be greatly exploited by using the internal mixer. This may be contributed by the greater ability of the internal mixer to distribute the filler evenly throughout the polymer matrix as compared to the extruder.

It is common to observe that the improvement in the tensile modulus is at the expense of the elongation at break (EB).[22] The EB for both composites decreases as the filler loading is increased (Fig. 3). This may be attributed to the reduction in deformability of a rigid interface between filler and the matrix component, which is also reflected in the increase of stiffness, as shown earlier in the tensile modulus result. Similar observations have been reported by several workers for other lignocellulosic composites.[4–8]

It should be noted that the degree of reduction in EB for EX composites is substantially higher as compared to IM composites. It can also be seen that IM composites display lower EB than the EX composites. A shown by tensile modulus results (Fig. 3), IM composites are stiffer than the EX. Thus, this obviously influences the EB properties of the composites. As better filler distribution is achieved in the internal mixer, more interfacial regions are expected to be formed between the filler and the polymer matrix. Thus, this would render the region stiffer and subsequently reduce the deformability of the composite.

It can be seen that neat PP, which is processed in an internal mixer (as the control sample), displays a higher tensile strength than the ones with the extruder. This may be attributed to the better shearing action of the former, which produces better homogenization and molecular orientation. The results also indicate that this process has detrimental effect on the ductility of the sample as reflected in the EB results.

The effects of filler loading and the method of mixing on the impact strength are shown in Figure 4. This observation is quite expected for filled

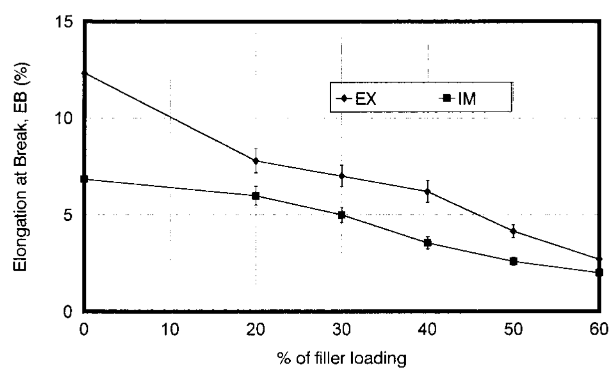


Figure 3 Elongation at break as a function of filler loading for IM and EX composites.

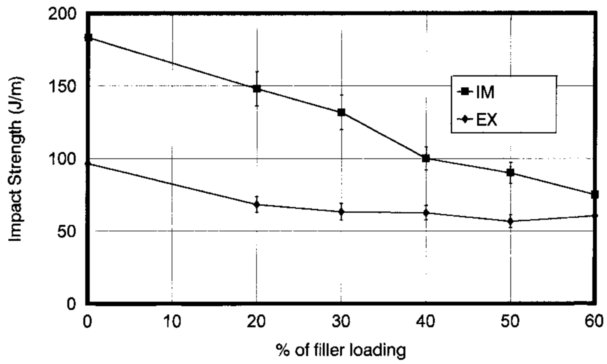


Figure 4 Impact strength versus filler loading for IM and EX composites.

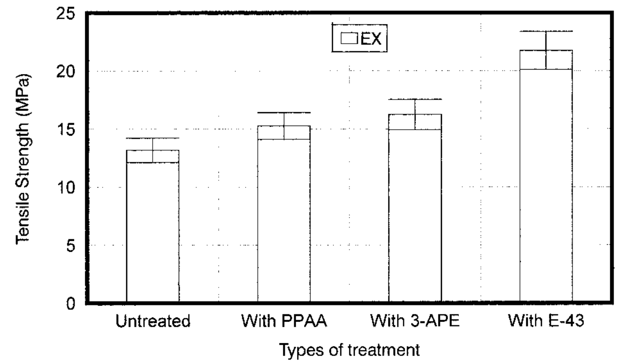


Figure 5 The effect of various treatments on the tensile strength for EX composites.

composites and has been commonly observed.[2,6,23] The results clearly indicate that the presence of EFB has reduced the energy absorbing capabilities of the composites. A similar trend has also been reported by Mohd. Ishak et al.[18] in the case of EFB-filled HDPE. As mentioned earlier, the filler system used in this study consists of irregular-shaped filler with a low aspect ratio. Thus, this influences the capability of the composite to absorb the energy as well as the capability to support stress transferred from the polymer matrix. IM composites exhibit a higher impact strength than the EX composites. Thus, it suggests that composites made by mixing in internal mixer is able to produce higher strength than those prepared through extrusion. This may be attributed by the mixing characteristics of the former. The mixing process in an internal mixer is run by 2 counter-rotating rotors, which give high shear rate, and the mixture is enclosed and confined in a mixing chamber. The process is more effective in improving the distribution of filler in the thermoplastic matrix. Consequently, the wetting of the filler surface is enhanced significantly. Composites with higher degree of homogeneity in terms of wettability and filler dispersion can be expected to possess superior mechanical properties.

Effects of Filler Treatment

Figures 5 and 6 depict the effect of coupling agents on the tensile strength of EX and IM composites, respectively. PPAA fails to impart a significant improvement on both IM and EX composites. As the geometry factor is suspected to play a major role in the performance of a composite as mentioned earlier, these results may indicate that the factor may outweigh the interaction introduced by PPAA. A similar trend has also been reported by Mohd. Ishak et al. in the case of EFB-filled HDPE.[18] However, as can be seen from both figures, there are some improvements in the tensile strength of 3-APE-treated fillers for both IM and EX composites. This may be contributed by the interaction brought about by (1) the silanol groups and the hydroxyl groups of lignocellulosic surface, through hydrogen bondings, and (2) the remaining chain of silane and PP through a van der Waals type of interaction. Thus, the results indicate that both types of bonding are sufficient to make a bridge between the lignocellulosic filler and polymer. This will consequently facilitate the stress transfer between both components. Tensile strength of the composites are further improved with E-43, especially for EX composites. This observation is in agreement with a recent investigation conducted by Myers et al.[20] on the influence of maleated PP on the mechanical properties of wood flour-filled PP. The greater reinforcement by the wood flour in the presence of the E-43 has been attributed to the better dispersion of the

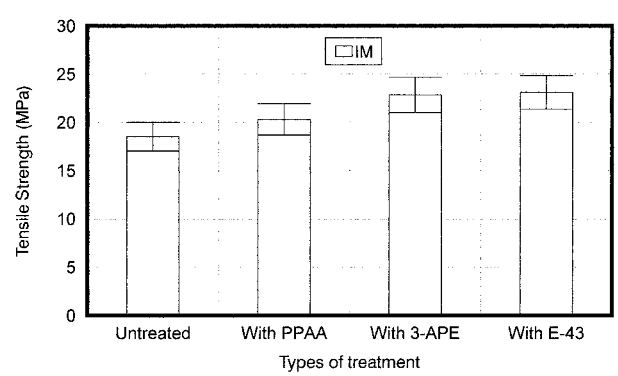


Figure 6 The effect of various treatments on the tensile strength for IM composites.

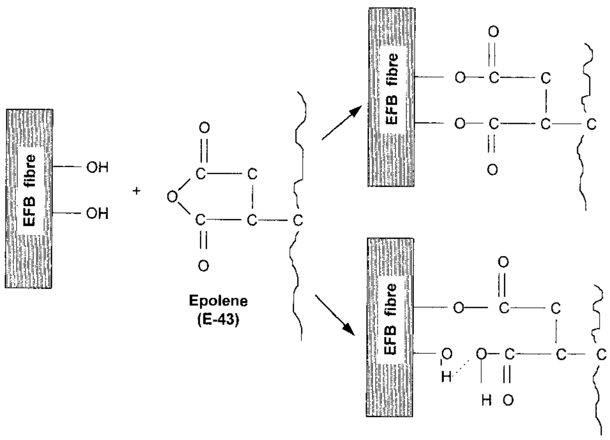


Figure 7 Possible reactions as EFB fibers are surface-treated with E-43.

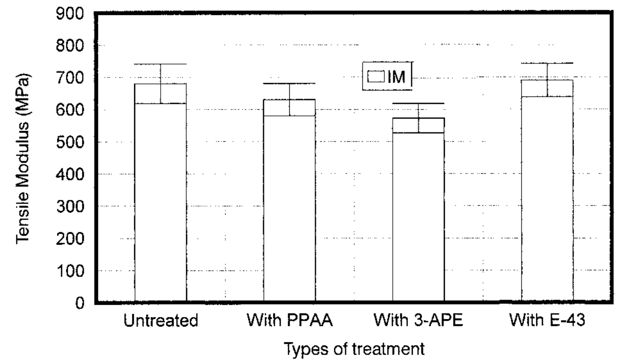


Figure 9 The effect of various treatments on the tensile modulus for IM composites.

wood flour in the PP and better bonding between the constituents materials. Similarly, in the present study, the E-43 compatibilizer could be expected to serve as a bridge that couples the 2 distinct phases, that is, EFB and PP. Figure 7 represents the possible chemical reaction taking place between the functional groups in EFB and E-43. It is believed that a good filler–matrix interactions is derived from the formation of hydrogen bonding between the anhydride groups of E-43 and the hydroxyl groups at the surfaces of EFB fillers. Since E-43 is a derivative of PP, it should be very compatible with PP. In fact, Kishi and coworkers[24] demonstrated that xylene extraction of the polymer from such systems leaves a filler residue of higher weight than that remaining from a comparable system without E-43, and the higher weight residue exhibits infrared peaks expected for an anhydride.

Treatment of EFB fillers by either compatibilizers, that is, PPAA and E-43, and coupling agent, 3-APE, have, to a certain extent, improved the tensile modulus of EX composites (Fig. 8). However, there are no significant improvements shown by IM composites (Fig. 9). The higher tensile modulus of the later composites are in agreement with the earlier trend observed in the case of tensile strength. This again highlights the fact that under the present compounding condition, the internal mixer is capable of enhancing dispersion and wettability of EFB fillers. The resultant increase in the efficiency of stress transfer will obviously lead to composites with a higher tensile modulus. As a reciprocal of tensile modulus, the results in Figures 10 and 11 show that the EB for IM composites is relatively lower than EX composites. However, in general, filler treatment does not produce much influence on the EB properties of both composites.

Incorporation of either coupling agent or compatibilizers does not show significant changes in the impact strength of EX composites (Fig. 12). However, as shown in Figure 13, it is obvious that the impact strength of IM composites has im-

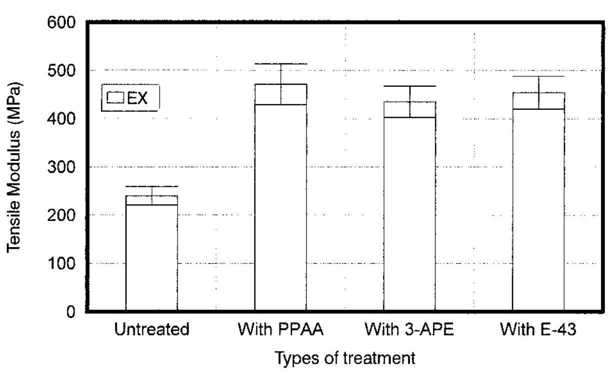


Figure 8 The effect of various treatments on the tensile modulus for EX composites.

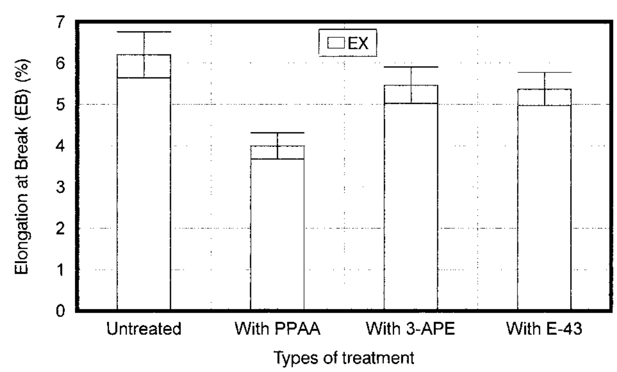


Figure 10 Comparative effect of various treatments on the elongation at break for EX composites.

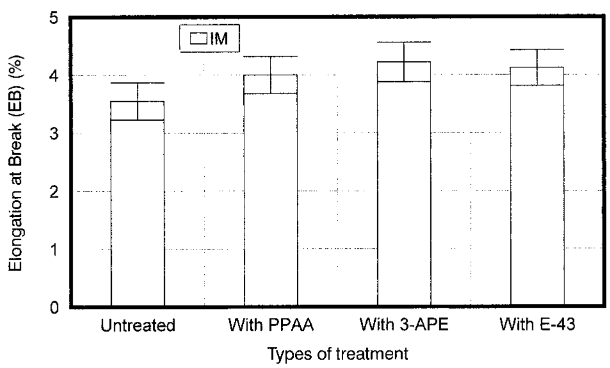


Figure 11 Comparative effect of various treatments on the elongation at break for IM composites.

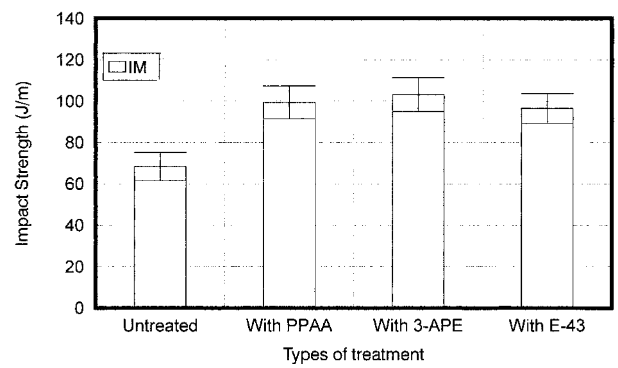


Figure 13 Comparative effect of various treatments on the impact strength for IM composites.

proved as a result of chemical treatment of EFB fillers.

With a higher degree of shear and localized mixing in the internal mixer than the extruder, it is believed that better distribution of lignocellulosic filler in the polymer matrix has occurred in the former. This is further supported by the morphological study of EFB–PP composite (treated with 3-APE) using scanning electron microscopy (SEM). From Figure 14, it can be seen that the EFB fillers are better distributed in the IM composites [Fig. 14(a)] as compared to that of EX composites [Fig. 14(b)]. It is also observed that the sizes of the fillers in the IM composites are relatively smaller than the ones mixed in the extruder. This provide a good indication of the efficiency of internal mixer in breaking the fiber bundles. It is a well-known fact that lignocellulosic filler has a strong tendency to agglomerate among themselves and, thus, exist in the form of fiber bundles.[18,25] In the present context, the smaller particle size and, hence, the larger surface area could provide more interfacial region between lignocellulosic and matrix components. Hence, this type of compounding technique would increase the homogeneity and accessibility of the coupling agents towards providing better bridging between the lignocellulosic and polymer matrix. This is further supported by the SEM micrographs shown in Figure 15(a) and (b), in which

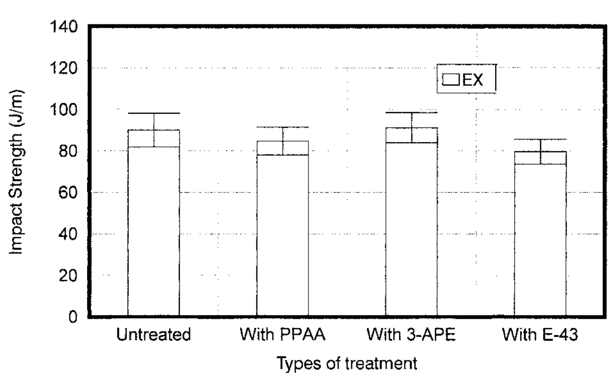
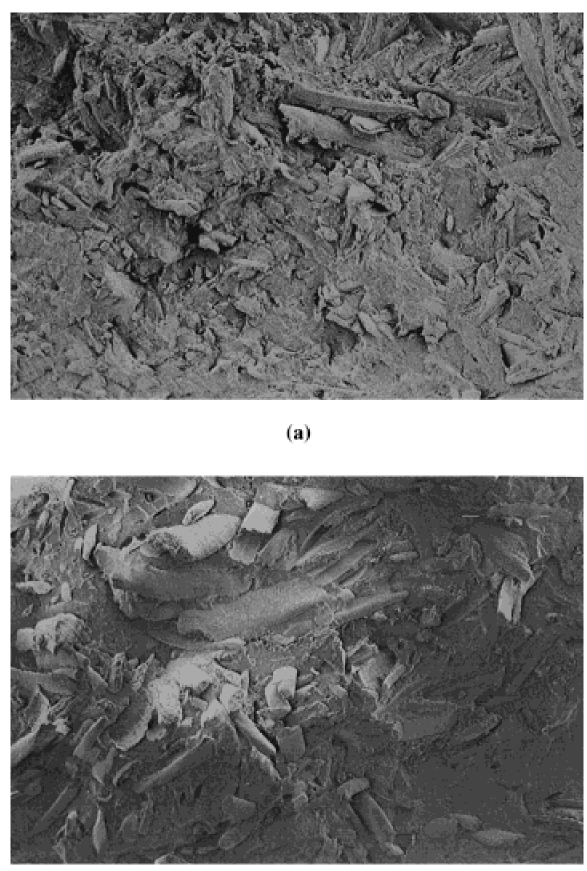


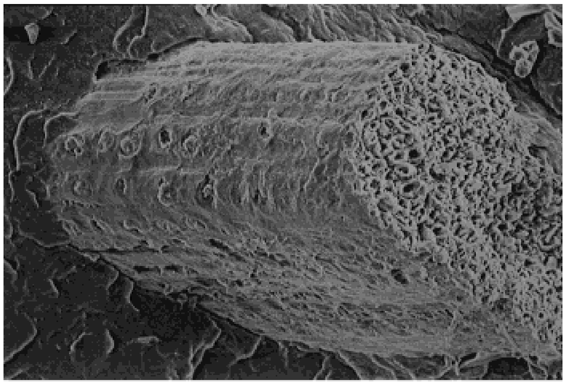
Figure 12 Comparative effect of various treatments on the impact strength for EX composites.



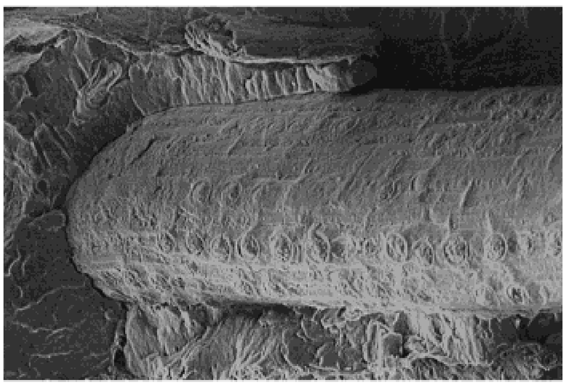
(a)

(b)

Figure 14 SEM micrographs for (a) IM and (b) EX composites with 40% filler loading, treated with 3-APE (magnification ×20).



(a)



(b)

Figure 15 SEM micrographs for (a) IM and (b) EX composites with 40% filler loading, treated with 3-APE, indicating EFB–PP interfacial region (magnification ×500).

better interaction between lignocellulose surface and the polymer matrix has been observed. As better bridging at the interface is in place, better transfer of stress is expected. This phenomena is reflected well in the tensile strength and impact results.

Figure 16 shows that the water absorption of untreated EX and IM composites (with 40 wt % filler loading). As expected, the amounts of water absorbed by both composites are comparable. In addition, both exhibit a similar pattern of water absorption; that is, initial sharp uptake is followed by gradual increase until equilibrium water content was achieved at about 10%. Previously, Klason et al.[2] have demonstrated that in the case of other lignocellulosic thermoplastic composites, the water absorption is proportional to filler loading, as the sorption by the thermoplastic matrix, such as PE and PP, could be neglected.

The effect of different types of chemical treatment on the water absorption of EFB–PP composites are shown in Figure 17. It can be seen that incorporation of coupling agent, 3-APE, have, to a certain extent, reduced the amount of water uptake by the composites. The slightly better water resistance of the EFB–PP composites treated with 3-APE may be attributed to the ability of the chemical to form a protective layer at the interfacial zone, which consequently prevents direct diffusion of water molecules into the lignocellulosic filler. The rather poor water resistance of the PPAA treated EFB–PP composites is not unsuspected. The acrylic acid component in PPAA is known to have a strong affinity towards water molecules. Thus, it tends to facilitate the diffusion of water molecules into the composites. Based on these observations, it could be anticipated that the presence of water will give rise to an adverse effect on the mechanical properties of EFB–PP composites. However, the improvement in the water resistance of EFB–PP composites, which arises as a result of the chemical treatment of the filler surfaces using a coupling agent, can be expected to provide better retention of the mechan-

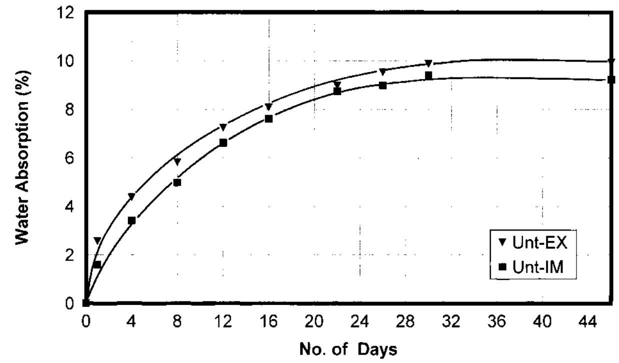


Figure 16 Water absorption profile for untreated IM and EX composites.

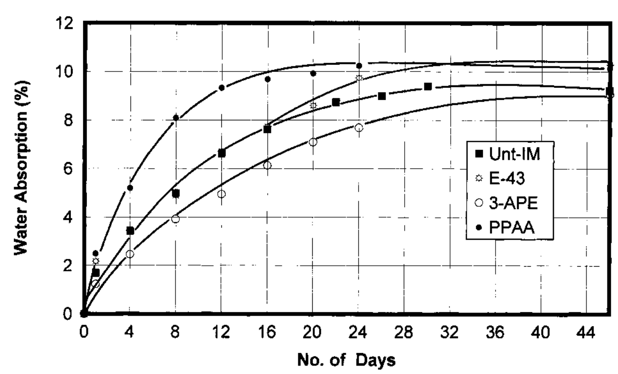


Figure 17 The effect of various treatments on the water absorption profile of IM composites.

ical properties. This subject will be dealt in greater detail in the forthcoming study.

CONCLUSION

The main objective of this investigation is to study the effect of mixing procedures on the mechanical properties of EFB–PP composites. The conclusions made from this study are as follows.

1. The incorporation of the EFB into PP matrix has resulted in the improvement in the tensile modulus. However, this improvement is obtained at the expense of the 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, the size irregularity of the EFB.
2. Composites produced by an internal mixer have displayed higher tensile strength, tensile modulus, and impact strength than with those produced by extrusion. This has been attributed to the better mixing procedures of the former, which involves high shear rate mixing by 2 counter-rotating rotors in an enclosed chamber, in which the frictional energy that evolved assists in heating up the mixtures. Thus, this process is believed to produce better filler dispersion and improve the wetting of the filler surface.
3. Incorporation of E-43 and 3-APE have produced composites with improved tensile and impact properties for both EX and IM composites.
4. Incorporation of 3-APE has also reduced the water absorption of EFB–PP composites; albeit, better water resistance was observed in the latter.

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