Effects of Rice Husk (RH) Particle Size, Glass Fiber (GF) Length, RH/GF Ratio, and Addition of Coupling Agent on the Mechanical and Physical Properties of Polypropylene-RH-GF Hybrid Composites

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ABSTRACT: Polypropylene (PP) hybrid composites based on rice husk (RH) with different particle size and glass fiber (GF) with different length were prepared. The composites were subjected to mechanical and water immersion tests. From the results obtained, it was found that the mechanical properties of the hybrid composites were strongly dependent on the size of RH particle and length of GF. It could be further enhanced with the presence of coupling agent. In this study, two types of coupling agents, i.e., Epolene E-43 (E-43) and 3-(trimethoxysilyl)-propylmethacrylate (TPM), were employed. In general, E-43 imparted significant improvement in the mechanical properties. From the water immersion results, it was found that the hydrophobicity of the composites was increased with the presence of coupling agent.

Key words: polypropylene; hybrid composite; lignocellulose; rice husk; glass; fiber

INTRODUCTION

Since a better understanding of the interaction between lignocellulosic material and plastic has been gained, the move to find substitutes for wood has been stimulated and hastened by the abundance of lignocellulosic materials available from agricultural crops as well as wood industries. The incorporation of lignocellulosic material, especially nonwood fiber as a reinforcing component in thermoplastic composites has received considerable attention recently, particularly for price driven and high-volume applications.[1–4] This development has been attributed to several advantages offered by lignocellulosic material over their inorganic counterparts, such as lower density, greater deformability, less abrasive to equipment, and lower cost per unit volume. On the other hand, the use of high-density inorganic fibers, such as glass fiber (GF) in thermoplastic composites is extensive due to the fact that inorganic fibers offer wide-ranging excellent properties, particularly in ultimate strength of the composites. Nevertheless, their incorporation may not be favorable in terms of cost effectiveness on a volumetric basis. Hence, it would be possible to benefit both inherent characteristics of lignocellulosic materials and glass fibers by combining them in a composite to compensate each other.

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In producing a good lignocellulosic-thermoplastic composite, with regards to mechanical and physical properties, the main obstacle to be solved is the compatibility between reinforcement material and polymer matrix. The compatibility and interfacial properties between them are expected to be poor because of the hydrophilic nature of lignocellulosic materials (contributed by hydroxyl groups in cellulose, lignin, and hemicellulose) and hydrophobic of thermoplastic. Many attempts have been carried out to improve the properties of the composites such as utilization of chemical reagents to enhance the interfacial properties between the constituent materials and addition of GF as a counterpart with lignocellulosic material in the hybrid composite system.

From previous studies,[5,6] coir and oil palm empty fruit bunch had been used as filler together with GF in the preparation of PP hybrid composites. The results showed that the stiffness of the PP hybrid composites increased as the filler content was increased. However, the tensile and flexural strength of the PP hybrid composites decreased as the filler was incorporated, which could be attributed to the incompatibility between fillers and PP matrix. This could be improved with the addition of coupling agent.[6] Thwe and Liao[7,8] had prepared PP hybrid composites from bamboo and glass fiber. They reported that the tensile strength and tensile modulus of the PP hybrid composites could be enhanced by the replacement of bamboo fiber with glass fiber. This could be further improved with the addition of compatibilizer such as maleated-PP. Vasoya et al.[9] had prepared bamboo-glass-bisphenol-C-formaldehyde-acrylate composites. They found that the tensile and flexural strength of the composites decreased and were attributed to the incorporation of more flexible and weaker bamboo fiber. Joseph et al.[10] had also prepared hybrid composites using banana fiber and glass fiber. From the results reported, banana fiber showed higher interfacial shear strength than GF in phenol formaldehyde composites.

In this study, rice husk (RH), one of the lignocellulosic materials which is of great relevance to Malaysia, and GF were used as reinforcement materials in the preparation of hybrid composites based on PP. The objective of this study is to investigate the properties of hybrid composites based on polypropylene (PP) prepared with different RH/GF ratio, GF length, and RH particle size. In addition, two types of coupling agent were also used, which were Epolene E-43 (E43) and 3-(trimethoxysilyl)-propylmethacrylate (TPM) to investigate the coupling effect on the properties of hybrid composites.

EXPERIMENTAL

Materials

RH in powder form was obtained from Bernas Dominals, Seberang Perai Utara, Penang, Malaysia. The GF in chopped-strand form was supplied by Fibre Glass Pilkington (India) with average length of 3.2 mm and 12 μm in diameter. The PP was purchased from Polypropylene Malaysia Sdn. Bhd., Malaysia with melt flow index of 12.0 g/min. Epolene E-43 (maleated-PP with M_w of 9100) with an acid number of 45[11] and 3-(trimethoxysilyl)-propylmethacrylate were supplied by Suka Chemical (M) Sdn. Bhd and Fluka Chemika, respectively.

Filler preparation

RH in powder form was separated to different particle size by an Endecotts sieve. The RH of mesh 60-35 (250–500 μm), 270-100 (53–150 μm) and 400 (<38 μm) were used in this study.

Filler treatment

RH with mesh 100-270 and GF with the length of 3.2 mm were used in this study. Three levels of coupling agent were employed as follows: 1, 3, and 5%, based on the filler content (weight basis). Epolene E-43 was used as delivered and hand-mixed with the mixture of PP and fillers (RH and GF) in dry form before compounding. As TPM was in liquid form, it was diluted in ethanol to make a 20% solution. The solution was added slowly into a round-bottom flask, which consists of fillers (RH and GF). The mixture was continuously mixed for 30 min. The treated fillers were then dried at 100°C to allow complete evaporation of ethanol.

Compounding and processing

The overall fiber content (by weight) in the composite was 60%, whereas the proportion of the RH and GF varied. Compounding of the materials were conducted by Haake twin-screw extruder (counter-rotating). The mixing temperature ranged from 165°C (feeding zone) to 180°C (die zone) with screw speed of 30 rpm. The mixture (RH, GF, and PP) was then extruded through a single rod die and palletized using a Haake electronic rotating cutter. The pellets were placed into a mold with a dimension of 170 mm × 170 mm × 3 mm (length × width × thickness). The mixture was preheated at 180°C for 10 min, followed by hot-pressing at the same temperature for 10 min at a pressure of 8 kgf/cm^2. Then, the sample was cooled for 5 min under the same pressure.

Mechanical test

Three types of mechanical test were carried out, i.e., tensile, flexural, and impact tests. Tensile test was conducted with a sample dimension of 150 mm × 19 mm × 3 mm (length × width × thickness) according to ASTM D638 using Instron Testing Machine model 5582, with a cross head speed of 5 mm/min. As for flexural test (three-point bending), it was carried out according to ASTM D790 using the same instrument with a crosshead speed of 2 mm/min. The sheet was cut into a dimension of 150 mm × 15 mm × 3 mm (length × width × thickness). The Izod impact tests were carried out on unnotched samples with dimension 65 mm × 12 mm × 3mm, using an Impact Pendulum Tester (Zwick) model 5101 according to ASTM D252. The properties were reported based on eight measurements for each composite system for each test.

Immersion test

Immersion test was conducted according to ASTM D570. The sample with a dimension of 30 mm × 15 mm × 3 mm (length × width × thickness) was immersed in distilled water at 30°C after being oven-dried at 105°C to a constant weight. The samples were periodically taken out of the water, surface-dried with absorbent paper before the weight and thickness of the sample were measured. The ultimate water absorption and thickness swelling were calculated by the following equations:

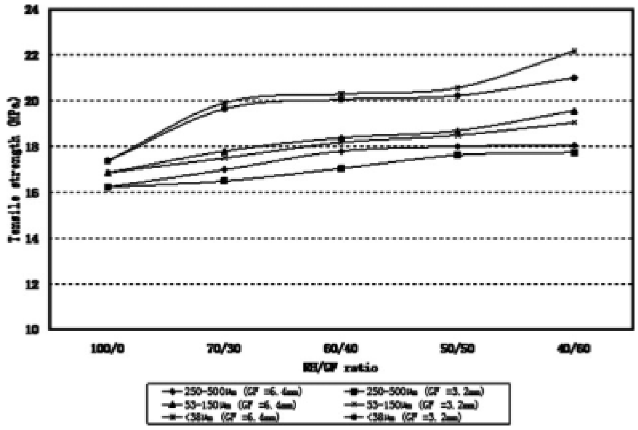

Figure 1 The effect of RH particle size, GF length, and RH/GF ratio on tensile strength of hybrid composites.

$$\text{Water absorption} = \frac{W_w - W_o}{W_o} \times 100$$

where W_w and W_o are the ultimate weight of the sample after a soaking period and initial oven-dried weight of the sample, respectively.

$$\text{Thickness swelling} = \frac{T_w - T_o}{T_o} \times 100$$

where T_w and T_o are the ultimate thickness swelling after a soaking period of immersion and original thickness, respectively.

Fractography study

The fracture surfaces of the composites from the tensile test were investigated with a Leica Cambridge S-360 scanning electron microscope. The objective was to get some information regarding bonding quality between fiber 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 thinlayer of gold to avoid electrostatic charging during examination.

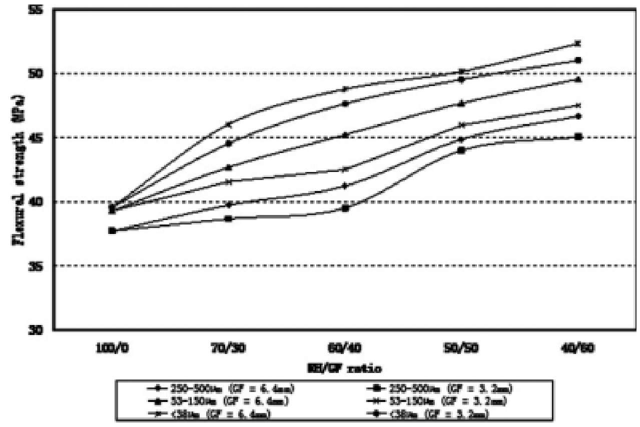

Figure 3 The effect of RH particle size, GF length, and RH/GF ratio on flexural strength of hybrid composites.

RESULTS AND DISCUSSION

Mechanical properties and physical properties

Figure 1 depicts the tensile strength of the hybrid composites. Generally, it appears that all types of composites show an increasing trend as the percentage of GF is increased, independent of the GF length and RH particle size. This reflects that the incorporation of GF has increased the tensile strength of the hybrid composite. The result also shows that composites prepared from longer GF length exhibit higher tensile strength when compared with those with shorter ones. This phenomenon is expected due to the higher aspect ratio of the longer GF. It should be noted that composites prepared from RH with smaller particle size display higher tensile strength when compared with the others, which is believed to be attributed to a better and uniform dispersion of RH in PP matrix, and thus results in a greater

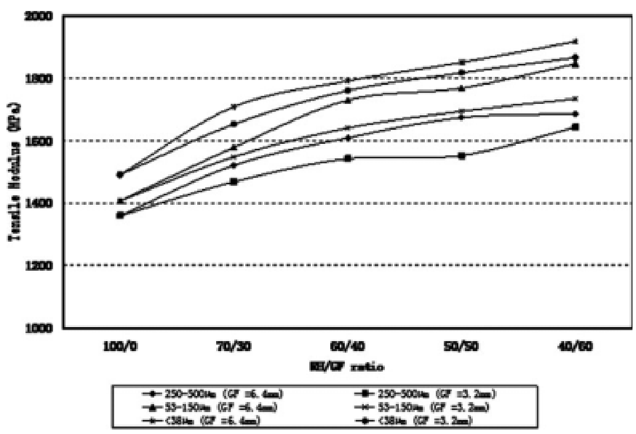

Figure 2 The effect of RH particle size, GF length, and RH/GF ratio on tensile modulus of hybrid composites.

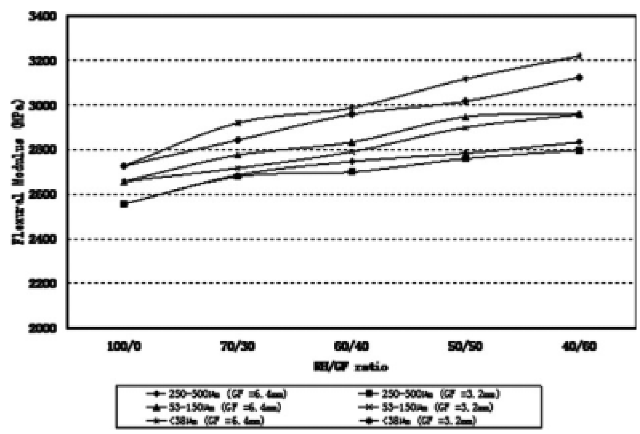

Figure 4 The effect of RH particle size, GF length, and RH/GF ratio on flexural modulus of hybrid composites.

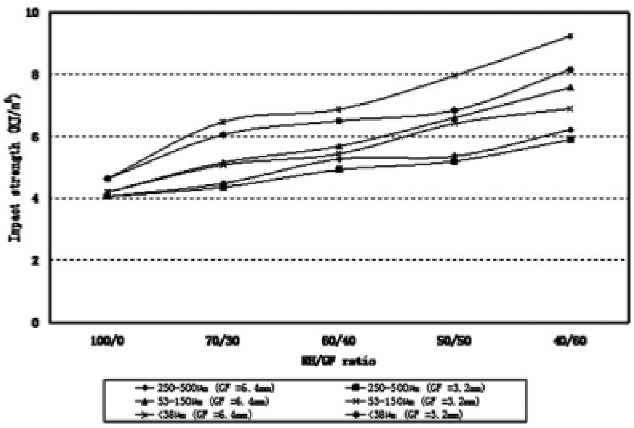

Figure 5 The effect of RH particle size, GF length, and RH/GF ratio on impact strength of hybrid composites.

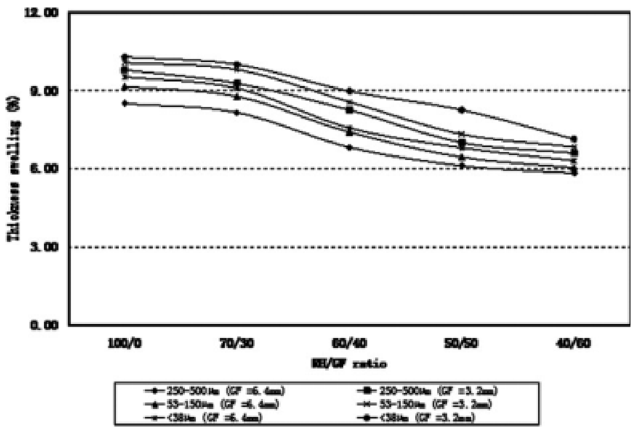

Figure 7 The effect of RH particle size, GF length, and RH/GF ratio on thickness swelling of hybrid composites.

interaction between RH and PP matrix. This observation has been reported in previous studies.[12,13]

Figure 2 shows the effect of RH/GF ratio on the tensile modulus of the hybrid composites prepared at different RH particle size and GF length. As can be seen in Figure 2, the introduction of GF increases the tensile modulus of the hybrid composites. This could be attributed to the inherent stiffness of GF that has increased the stiffness of the composites. As for the RH particle size, the homogeneity of finer RH in the hybrid composites system and a greater interaction with PP matrix has resulted in an increase in the tensile modulus of the composites. This observation is in line with previous study.[12]

The flexural strength of the hybrid composites prepared from different RH particle size and GF length at different RH/GF ratio is depicted in Figure 3. A similar trend as shown in the tensile strength is observed. The smaller RH particle size, longer GF, and higher GF content of a hybrid composite result in higher flexural strength. This may be due to a uniform dispersion of RH particle in hybrid composite system and higher aspect ratio of GF. The inherent stiffness as well the aspect ratio of GF has accounted for the increase in the stiffness of the composites (Fig. 4) since GF has higher stiffness when compared with RH.

Figure 5 shows the impact strength of the composites. Generally, the impact strength increases as the particle size of RH is decreased. This is attributed to a better homogeneity of RH dispersion in PP matrix when smaller particle size is used. In addition, the impact strength of the hybrid composites is found to be enhanced with longer GF. It is also noticed that the efficiency of energy absorption increases as the GF content is increased. As mentioned earlier, this is due to the higher aspect ratio of the longer GF.

Water immersion test shows the behavior of the hybrid composites when immersed in water, studied with respect to the percentage of water absorbed (Fig. 6) and the degree of thickness swelling (Fig. 7). The results show that the percentage of water absorbed decreases as the RH content is decreased. This is expected because RH is a lignocellulosic material that consists predominantly of cellulose, lignin, and hemicellulose. These materials possess polar hydroxyl groups, which may readily absorb water

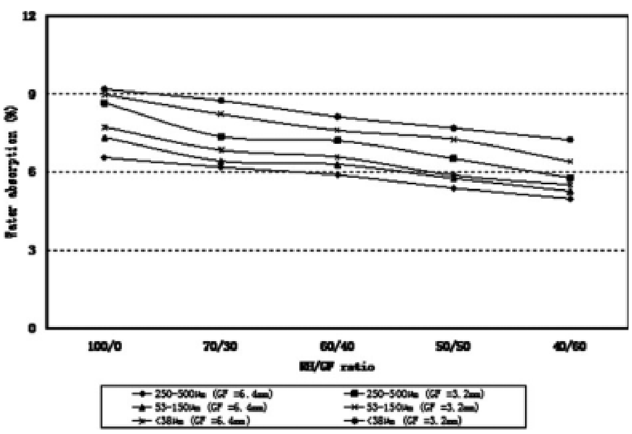

Figure 6 The effect of RH particle size, GF length, and RH/GF ratio on water absorption of hybrid composites.

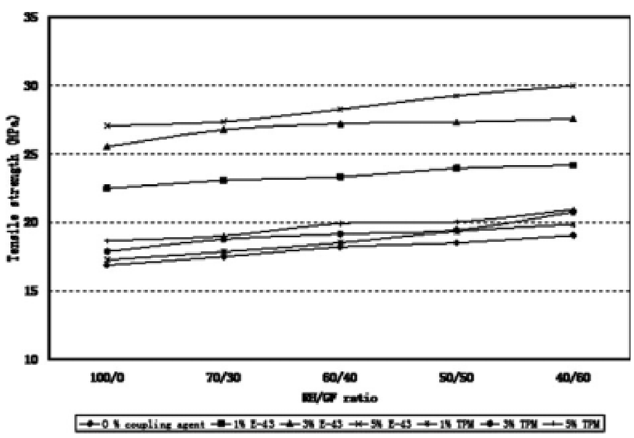

Figure 8 The effect of coupling agents on tensile strength of hybrid composites.

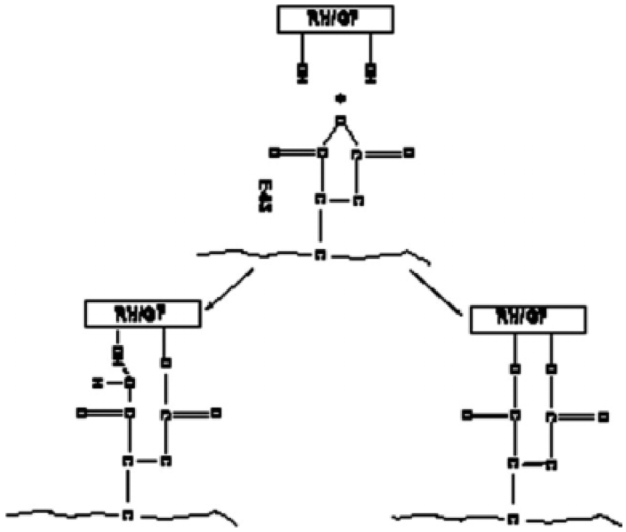

Figure 9 Interaction between RH/GF and E-43.

into its cell wall through the formation of hydrogen bonding. This will subsequently result in the swelling of the cell wall. With respect to the RH particle size, smaller RH particle provides higher total surface area. Thus, more surface area is exposed for water absorption when compared with the larger particle size of RH. For composites with longer GF, the tendency to absorb water is reduced, compared with those prepared from the shorter ones. This could be attributed to the longer GF that gives better distribution and the interaction with the RH in the matrix. This subsequently leaves fewer hydroxyl groups of RH exposed to water.[14]

Effects of coupling agents on the mechanical properties and physical properties of the hybrid composites

The effect of coupling agent on the tensile strength of hybrid composite is shown in Figure 8. In general, the strength of the hybrid composites is improved with the incorporation of coupling agent. Between the two coupling agents employed, E-43 shows greater enhancement of the mechanical properties when compared with TPM. This phenomenon may be attributed to the nature of E-43, which is a derivative of PP (maleated-PP). Hence, the compatibility between E-43 and PP is anticipated to be higher than the compatibility between TPM and PP. E-43 is also believed to serve as a bridge with higher efficiency when compared with TPM to couple two distinct phases, RH/GF and PP through the formation of ester bond, a product from the reaction of hydroxyl groups from the fillers and anhydride groups from E-43 as shown in Figure 9. In addition, the amount of coupling agent incorporated into the hybrid composites system also plays a significant role in determining the strength of the composites prepared. It appears that the tensile strength is enhanced as the percentage of the coupling agent in the hybrid composites system is increased. This phenomenon indicates that more interfacial interaction

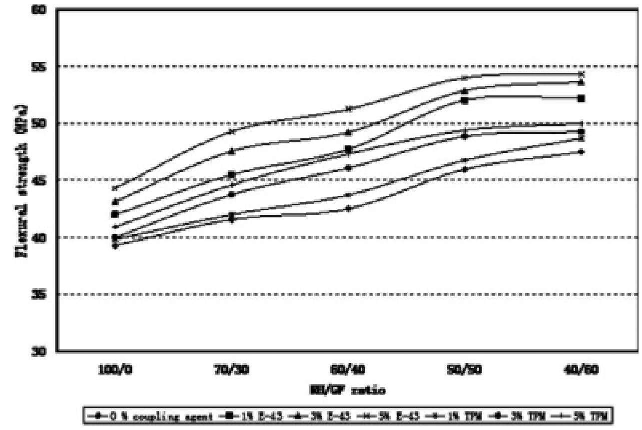

Figure 11 The effect of coupling agents on flexural strength of hybrid composites.

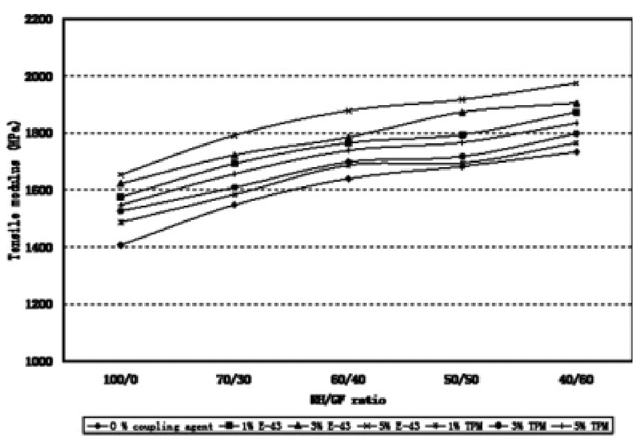

Figure 10 The effect of coupling agents on tensile modulus of hybrid composites.

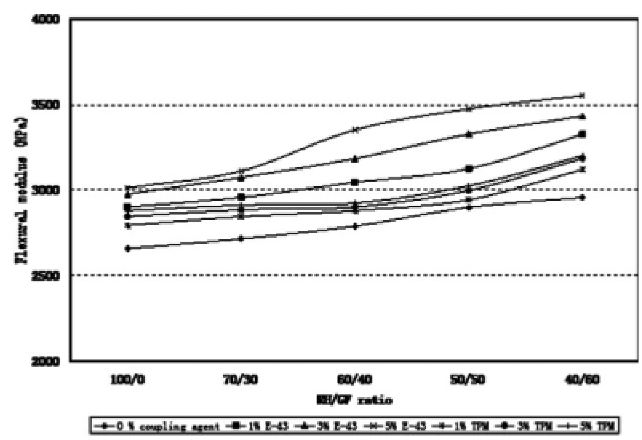

Figure 12 The effect of coupling agents on flexural modulus of hybrid composites.

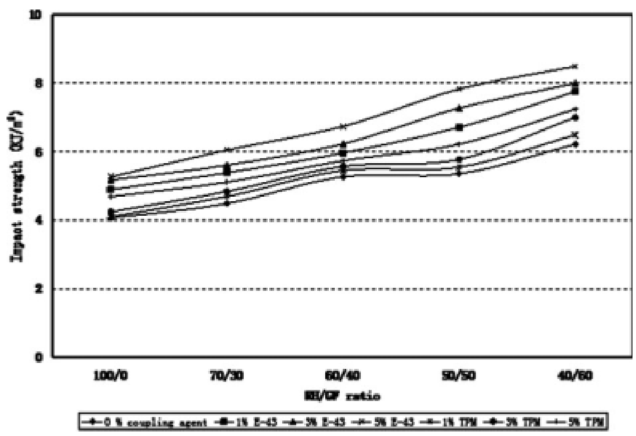

Figure 13 The effect of coupling agents on impact strength of hybrid composites.

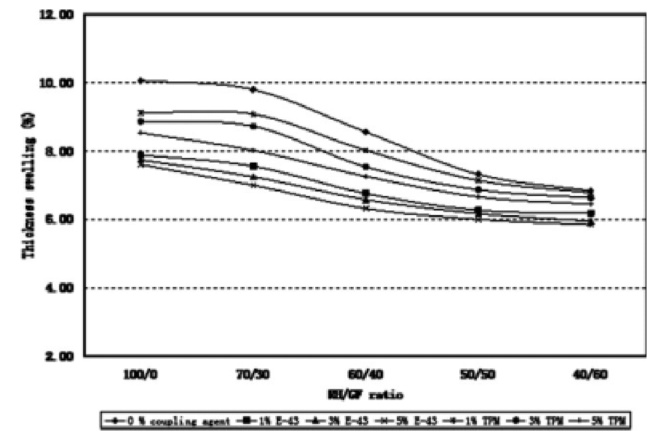

Figure 15 The effect of coupling agent addition on thickness swelling of hybrid composites.

has taken place as the percentage of coupling agent is increased, which would enhance the stress transfer mechanism between fillers and PP matrix.

The tensile modulus of the hybrid composites is found to be influenced by the presence of coupling agent. Results illustrated in Figure 10 show that both coupling agents and GF are able to improve the stiffness of the composites. This indicates that E-43 is able to impart greater stiffness to the hybrid composites than TPM. Again, it is believed that E-43 has improved the interfacial properties between fillers and PP matrix via ester linkages. The stiffness of the hybrid composites is further enhanced with progressive increases in GF content.

Figure 11 depicts the effect of RH/GF ratio on the flexural strength of hybrid composites with the addition of different types of coupling agents. Identical observation as shown in tensile strength is evident, whereby flexural strength increases with increasing coupling agent loading. Hybrid composites with higher amount of GF show higher flexural strength with the presence of coupling agent. Comparatively, E-43 shows a better enhancement in flexural strength than TPM. As for flexural modulus (Fig. 12), the stiffness of the hybrid composites is significantly improved by E-43 and the amount of GF incorporated.

The effect of coupling agent on the impact strength of the hybrid composites varied depending on the type of coupling agent used. As shown in Figure 13, it is obvious that composites prepared from E-43 show better improvement in impact strength than those composites with TPM. It again indicates that E-43 promotes better linkage between fillers and PP matrix. This result is supported by the immersion test results shown in Figures 14 and 15. The water absorption and thickness swelling of the hybrid composites decrease as the amount of coupling agent added is increased. Thus, composites with E-43 exhibit higher hydrophobicity than those without coupling agent or added with TPM. This may due to (i) the ability of composites to absorb water has been reduced as a result of the interaction

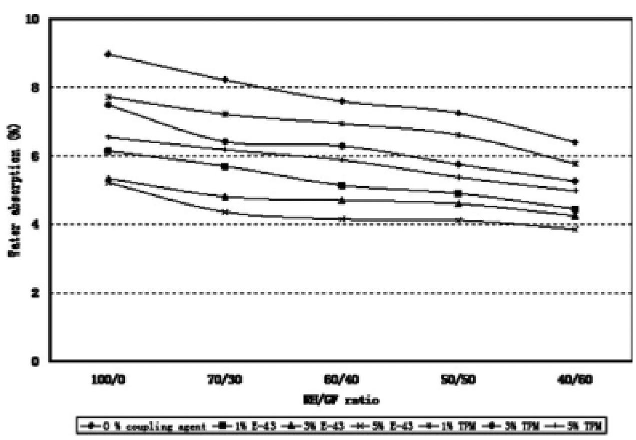

Figure 14 The effect of coupling agents on water absorption of hybrid composites.

Figure 16 Fracture surface of composite prepared from untreated RH and GF.

Figure 17 Fracture surface of composite prepared from TPM treated RH and GF.

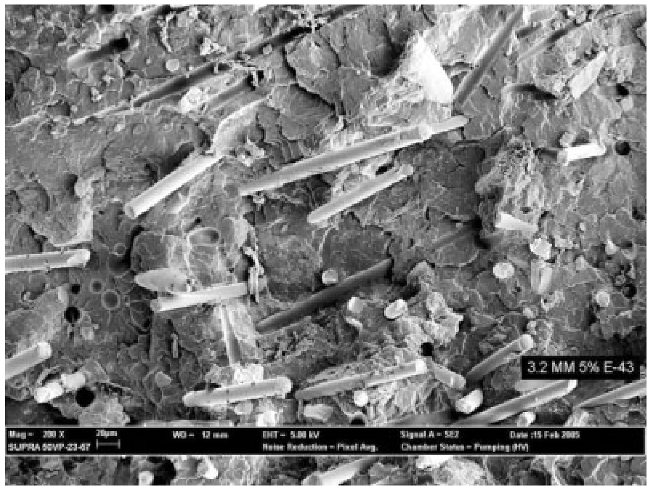

Figure 18 Fracture surface of composite prepared from E-43 treated RH and GF.

between E-43 and hydroxyl groups from fillers, and (ii) the hydrophobicity imparted by the PP chain of E-43. It is also observed that the ability of hybrid composite to absorb water is reduced with the reduction of RH content.

As for morphology study, there are fiber pull out phenomenon observed as indicated in Figure 16. After RH and GF treated with TPM, less fiber pull out phenomenon could be seen (Fig. 17) when compared with the treated one (Fig. 16). However, when RF and GF treated with E-43, there are interactions between matrix, and reinforcement could be observed as indicated in Figure 18. Hence, this explains why RH and GF treated with E-43 could produce composites with higher strength when compared with those prepared from untreated RH and GF, and TPM treated RH and GF.

CONCLUSIONS

The mechanical and physical properties of PP hybrid composites, based on RH and GF were strongly influenced by the particle size of RH and the length of GF. Smaller particle size of RH was found to be better dispersed in the hybrid composite system and although, longer GF with a higher aspect ratio gave a better reinforcement effect to the composites. In addition, the RH/GF ratio also plays an important role in determining the properties of the hybrid composites prepared. However, the hydrophilicity of the RH has contributed to a greater extent to water absorption and thickness swelling. This can be overcome by adding coupling agent into the hybrid composites system. Overall, composites prepared with coupling agent showed higher mechanical properties and lower water absorption, especially those with E-43. This indicates that E-43 has a profound effect on the stress transfer efficiency by introducing a linkage between fillers and PP matrix.

References

1. Felix, J. M.; Gatenholm, P. J Appl Polym Sci 1991, 42, 609.
2. Joseph, K.; Thomas, S.; Pavithran, C. Polymer 1996, 37, 5139.
3. Bledzki, A. K.; Reihmane, S.; Gassan, J. J Appl Polym Sci 1996, 59, 1329.
4. Gassan, J.; Bledzki, A. K. Compos A 1997, 28, 1001.
5. Rozman, H. D.; Tay, G. S.; Kumar, R. N.; Abubakar, A.; Ismail, H.; Mohd.Ishak., Z. A. Polym Plast Technol Eng 1999, 38, 997.
6. Rozman, H. D.; Tay, G. S.; Kumar, R. N.; Abusamah, A.; Ismail, H. Eur Polym J 2001, 37, 1283.
7. Thwe, M. M.; Liao, K. Compos A 2002, 33, 43.
8. Thwe, M. M.; Liao, K. Compos Sci Technol 2003, 63, 375.
9. Vasoya, P. J.; Mehta, N. M.; Parsania, P. H. Polym Plast Technol Eng 2007, 46, 621.
10. Joseph, S.; Sreekala, M. S.; Oommen, Z.; Koshy, P.; Thomas, S. Compos Sci Technol 2002, 62, 1857.
11. Yang, H. S.; Kim, H. J.; Park, H. J.; Lee, B. J.; Hwang, T. S. Compos Struct 2007, 77, 45.
12. Rozman, H. D.; Kon, B. K.; Abusamah, A.; Kumar, R. N.; Mohd Ishak, Z. A. J Appl Polym Sci 1998, 69, 1993.
13. Rozman, H. D.; Ismail, H.; Jaffri, R. M.; Aminullah, A.; Mohd Ishak, Z. A. Int J Polym Mater 1998, 39, 161.
14. Rozman, H. D.; Hazlan, A.; Abubakar, A. Polym Plast Technol Eng 2004, 43, 1129.