

Tensile Properties of Kenaf/Unsaturated Polyester Composites Filled with a Montmorillonite Filler

H. D. Rozman,¹ L. Musa,² A. A. Azniwati,¹ A. R. Rozyanty¹

¹School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia

²School of Material Engineering, Universiti Malaysia Perlis, Perlis, Malaysia

Received 6 May 2009; accepted 27 December 2009

DOI 10.1002/app.32096

Published online 9 September 2010 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: Kenaf/unsaturated polyester composites filled with montmorillonite (MMT) filler were produced. Overall, the study showed that, for samples with kenaf filler only, the strength properties decreased as the kenaf filler loading was increased from 40 to 60%. The increase in the kenaf filler loading reduced the amount of matrix material. This subsequently lowered the ability of the sample to absorb energy or distribute stress efficiently. However, with MMT, the tensile properties improved because of the high aspect ratio and surface area of the MMT. The study

of the effect of kenaf filler size on the tensile properties showed that the samples with the smallest size (74 μm) displayed the lowest tensile properties compared to the larger ones. This was attributed to the agglomeration of the kenaf fillers. The addition of MMT resulted in an overall increase in the tensile strength of the composites compared to those without MMT. © 2010 Wiley Periodicals, Inc. *J Appl Polym Sci* 119: 2549–2553, 2011

Key words: composites; mechanical properties; polyesters

INTRODUCTION

Recently, the use of lignocellulosic materials to make composites has received considerable attention. This has been attributed to several advantages offered by lignocellulosic fillers, such as a lower density, greater deformability, lower abrasiveness to equipment, biodegradability, and lower cost. However, to produce a polymer composite with acceptable mechanical and physical properties, the main obstacle to be resolved is the compatibility between the lignocellulosic material and the polymer matrix. Generally, there are two types of interaction at the interfacial region, that is, covalent and hydrogen bonding. In thermoplastic–wood composites, covalent bonding at the interfacial region can be created with the incorporation of a coupling agent.

Kenaf has gained research interest as a lignocellulosic filler in polymer composites. One of its advantages is the absence of silica content, which is critical in the reduction of abrasiveness to processing equipment.¹ The density of kenaf (0.1–0.2 g/cm^3)² is much lower than that of glass fiber (2.55 g/cm^3),³ carbon fiber (1.77 g/cm^3),³ oil palm empty fruit bunch (0.7–1.55 g/cm^3),³ and hemp (1.48 g/cm^3).⁴ This low density gives the kenaf core an advantage to be used in

applications, such as automotive interior paneling, to reduce fuel consumption.⁵

The market price for lignocellulosic filler is much cheaper than conventional fillers, such as Kevlar, glass fibers, and carbon fibers.⁶ However, the incorporation of a lignocellulosic filler has been shown to adversely affect the strength of composites.^{7–11} This situation may outweigh the importance of economy posed by lignocellulosic composites. From various studies in nanocomposites, nanofiller addition into a polymer matrix increases the strength and the modulus of the composites.^{12–16} With their high surface area and aspect ratio, nanofillers are expected to balance the adverse effect of the irregularities of the lignocellulosic fillers. Thus, in this study, we focused on the effect of a nanofiller on the tensile properties of kenaf/unsaturated polyester composites (USPs). The nanoclay was a montmorillonite (MMT), a layered aluminosilicate that comes from the smectite group. It has individual platelets that measure on the order of the micrometer scale diameter; this gives them an aspect ratio of about 1000 : 1, and the surface area of this clay can approach up to 760 m^2/g .¹⁷ Although it naturally forms stacks of platelike structure, it can still exhibit a high aspect ratio.

EXPERIMENTAL

Materials

Unsaturated polyester resin (P9728) was purchased from Euro Chemo-Pharma Sdn. Bhd. Pte. Ltd.,

Correspondence to: H. D. Rozman (rozman@usm.my).

(Pulau Pinang, Malaysia). The resin was an isophthalic type with thixotropic properties and an acid number of 26. *tert*-Butyl perbenzoate (Fluka 77200, Germany) was purchased from Sigma-Aldrich (Malaysia). Zinc stearate was purchased from Liangtraco Sdn. Bhd. Pte. Ltd., (Pulau Pinang, Malaysia). Kenaf core was supplied by the Forest Research Institute of Malaysia. The MMT filler (PGV grade) was purchased from Nanocor, Inc., (USA) with a purity of 98%.

Composite production

All materials were weighted accordingly to obtain the required formulation. Various formulations were prepared involving three levels of kenaf filler loadings (40, 50, and 60%), three ranges of kenaf filler size (the kenaf fiber was sieved according to mesh-number range; these were 35–100, 100–200, and 200–400; the filler size of each range was 500–250, 150–75, and 75–38 μm , respectively), and three levels of MMT (1, 3, and 5%). All compositions were based on the volume of the mold to maintain the density of the composite at 1 g/cm^3 (the total amount of the compound in grams was based on the volume of the mold). The unsaturated polyester resin was mixed with *tert*-butyl perbenzoate [1 part per hundred resin (phr)] in a mixing container. The mixing was carried out with a high-torque mixer. Then, zinc stearate (zinc stearate acted as a lubricant, and the 6 phr of zinc stearate was constant in all formulations) was added followed by the gradual addition of MMT to ensure homogeneous mixing. Last, kenaf filler was added. Overall, the mixing was carried out for 30 min. The mixture was then poured into a mold and placed in a hot press. The curing was carried out at 135°C with a pressure of 400–500 kg/cm^2 for 5 min, followed by cooling for 25 min (under pressure).

Tensile test

Tensile testing was conducted according to ASTM D 3039 with an Instron testing machine model 5580 (USA). Samples were cut into dimensions of 100 \times 12 \times 5 mm^3 (Length \times Width \times Thickness). The test was conducted at a crosshead speed of 2 mm/min with a gauge length of 60 mm . A minimum of 10 samples were tested in each case.

Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX) analysis

Samples were gold-coated to ± 20 nm thickness with a Sputter Polaron model SC 515 (UK) before their morphology was examined with a Leo Supra 50 VP equipped with Oxford INCA 400 (Germany and United Kingdom, respectively) scanning electron microscope. The same samples for SEM analysis were

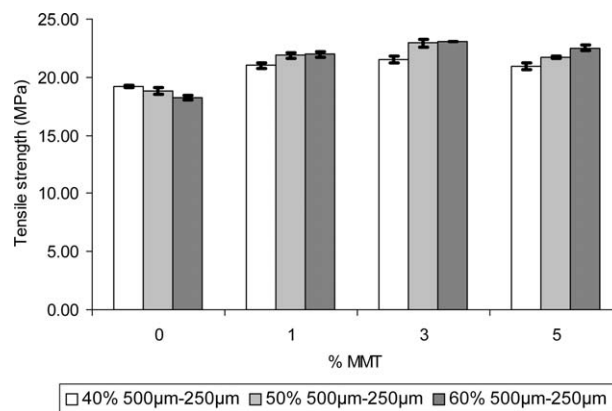


Figure 1 Effect of the kenaf filler and MMT loading on the tensile strength.

used in EDX analysis to qualitatively analyze the elemental presence in the samples. The samples were mounted on an aluminum stub with double-sided tape and then gold-coated with a Polaron SEM coating unit to prevent electrical charging during the examination.

RESULTS AND DISCUSSION

Figure 1 shows the tensile strength results for USPs filled with 40, 50, and 60% kenaf filler with a size of 500 μm together with 0, 1, 3, and 5% MMT. For composites without MMT, the tensile strength decreased as the kenaf filler loading was increased from 40 to 60%. We believe that some interaction occurred between the OH groups of the kenaf filler and the oxygen atom from the C=O groups of USP. However, it seemed that this interaction was not sufficient to exact an increase in the tensile strength. Thus, this may have indicated the direct effect of the depletion of the USP matrix as the result of kenaf filler addition because the weight of the sample was kept constant to maintain the same density. This, hence, reduced the continuous region of the matrix. Furthermore, the depletion of the USP matrix reduced the interaction and interfacial bonding quality between the kenaf filler and the matrix. This affected the efficiency of stress distribution from the matrix to the fillers. This result was in agreement with previous study.¹⁸ Kenaf with irregular shapes and sizes was unable to provide good stress distribution from the matrix. This was further magnified with the increase in the loading. The argument was supported by previous studies.^{19,20} With MMT, the tensile strength increased as the kenaf filler loading was increased. This indicated that the incorporation of MMT outweighed the effect of matrix depletion as the kenaf filler loading was increased. This may have been due to the high aspect ratio and surface area of the MMT. These two attributes enhanced the distribution of stress in the composites.^{21,22}

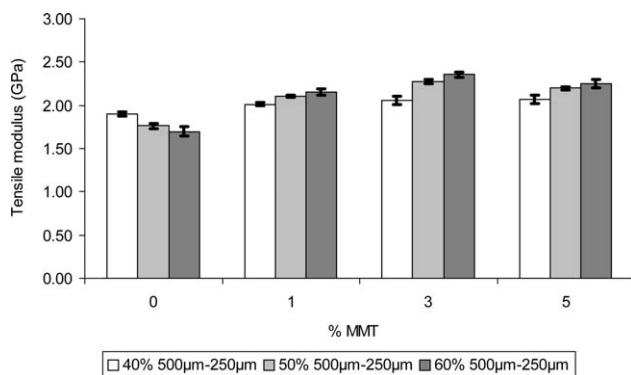


Figure 2 Effect of the kenaf filler and MMT loading on the tensile modulus.

The tensile modulus results, shown in Figure 2, follow the same trend as the tensile strength results. For composites without MMT, the modulus decreased as the kenaf filler loading was increased from 40 to 60%. The matrix, which is a continuous phase, is the main contributor to the modulus of a composite. As mentioned in the tensile strength results discussion, the addition of kenaf filler resulted in the reduction of this continuous phase and its homogeneity, hence, the reduction in modulus. However, with MMT, the tensile modulus increased as the kenaf filler loading was increased. It seemed that the effect of MMT addition overshadowed the effect of the reduction of the matrix and inhomogeneity in the matrix. We believe that it was contributed by the high aspect ratio and high surface area of the MMT, with its high surface area, which meant more OH groups on its surface accessible to interact with oxygen atoms from the C=O of maleic residues of the USP. This phenomenon was clear in the SEM micrograph (shown later in Fig. 9). This may have restricted the mobility of the matrix, which subsequently resulted in an increase in stiffness.

Figure 3 shows the elongation at break (EB) results. In general, the incorporation of MMT and kenaf filler into the matrix resulted in the reduction

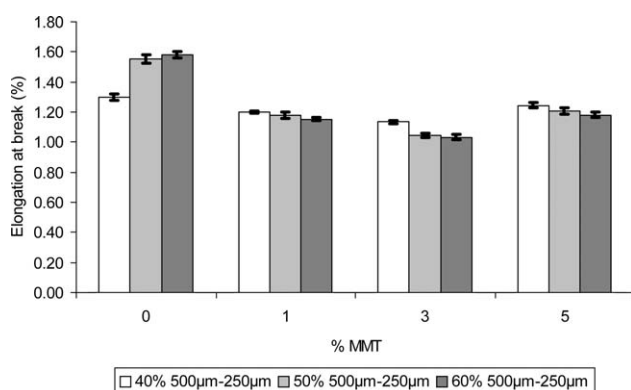


Figure 3 Effect of the kenaf filler and MMT loading on EB.

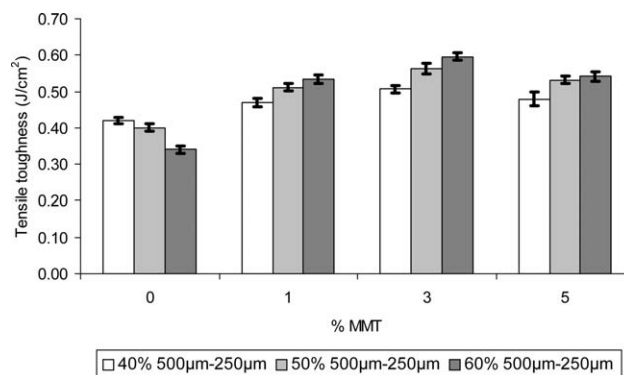


Figure 4 Effect of the kenaf filler and MMT loading on the tensile toughness.

in EB for all of the samples. This was in agreement with the results of the modulus shown in Figure 2.

The tensile toughness results (Fig. 4) show that, for composites without MMT, the toughness decreased as the kenaf filler loading was increased from 40 to 60%. This indicated that as the matrix component was reduced (with the addition of kenaf filler); the ability of the sample to absorb energy was subsequently reduced. This showed that the contribution of the continuous phase of the matrix to absorb energy was superior to any interaction and interfacial bonding between the kenaf filler and the matrix. However, with MMT, the tensile toughness increased as the kenaf filler loading increased. It was obvious that the incorporation of MMT increased the energy absorption of the sample, albeit the matrix component decreased as the kenaf filler loading was increased. This may have been due to the high aspect ratio and surface area of the MMT. The former, with more OH groups on its surface to interact with oxygen atoms from the C=O of maleic residues of the USP, produced more bridging between the components. This helped in absorbing the energy being distributed from the matrix to the fillers.

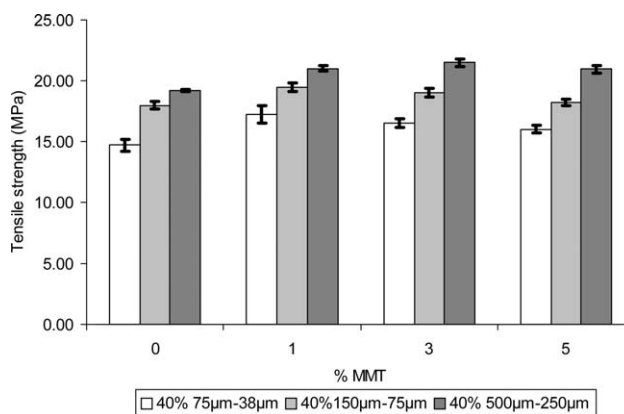


Figure 5 Effect of the kenaf filler size and MMT loading on the tensile strength.

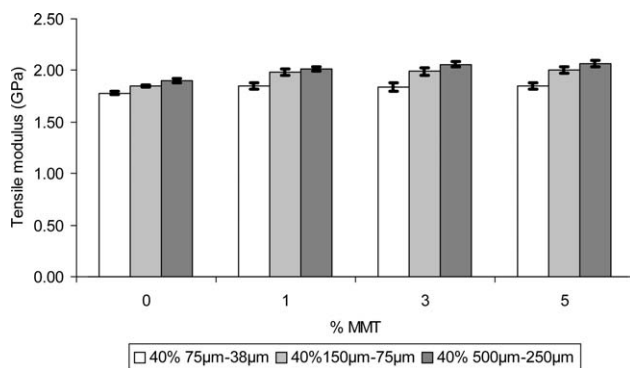


Figure 6 Effect of the kenaf filler size and MMT loading on the tensile modulus.

Figure 5 shows the tensile strength results for the USPs filled with 40% kenaf filler with sizes of 74, 150, and 500 μm , together with 0, 1, 3, and 5% MMT. For those without MMT, the samples with the smallest size in this study (74 μm) showed the lowest tensile strength as compared to the larger ones. By looking at the overall results, we believed this to be due to agglomeration of the kenaf fillers. This is understandable because, being a lignocellulosic materials, kenaf had OH groups contributed by cellulose, hemicellulose, and lignin. These groups were able to form hydrogen bonds with each other, which, hence, resulted in the agglomeration of the particles. These agglomerations created stress points in the matrix and made the stress transfer from the matrix less efficient. For those with bigger size fillers, with fewer OH groups, the ability to form agglomerations was lower than those with smaller ones. Thus, less stress points were expected in the composites with larger size particles. This may explain why the tensile strength of the samples with larger size particles was higher than those with the smaller ones. The addition of 1% MMT resulted in an overall increase in the tensile strength of the composite as compared to the composites without MMT. Those with 3 and 5% had results comparable to those with 1%. Thus, it was clear that the incorpora-

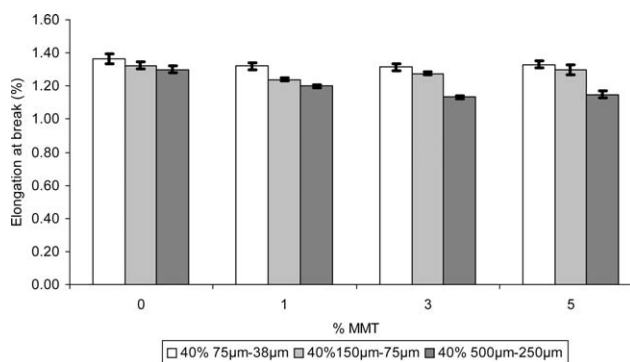


Figure 7 Effect of the kenaf filler size and MMT loading on EB.

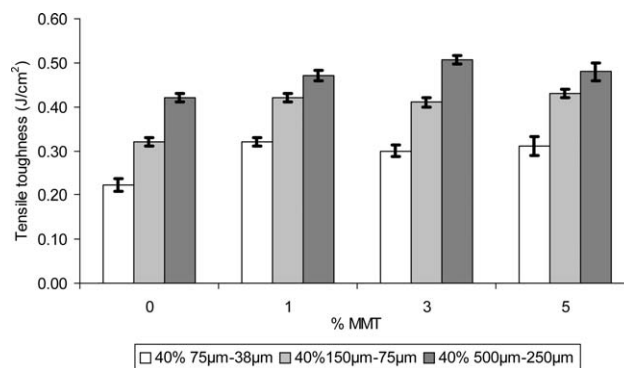


Figure 8 Effect of the kenaf filler size and MMT loading on the tensile toughness.

tion of more MMT resulted in better stress distribution in the sample.

The tensile modulus results (Fig. 6) show the same trend as the tensile strength results (Fig. 5). The incorporation of MMT increased the modulus of the composites. The modulus increased as the size of the kenaf filler increased. This indicated the contribution of the inherent modulus of the kenaf filler in the composite.

Figure 7 shows the EB results. In general, the incorporation of MMT and kenaf filler into the matrix resulted in the reduction of EB for all samples. This was in agreement with the results of the modulus shown in Figure 6.

The tensile toughness results (Fig. 8) show that, for all samples, the toughness increased with increasing filler size. All samples with MMT displayed higher toughnesses than those without. This indicated that the incorporation of MMT increased the energy absorption of the sample. This may have been because of the high aspect ratio and surface area of the MMT. The MMT, with more OH groups on its surface to interact with oxygen atoms from the C=O of maleic residues of the USP, produced more bridging between the components. This helped to absorb the energy being distributed from the matrix to the fillers.

TABLE I
EDX Data for the MMT-UNT Particles Shown in Figure 9

Element	wt %
C	13.62
O	42.17
Na	0.71
Mg	1.99
Al	8.61
Si	28.14
K	2.19
Fe	2.57
Total	100.00

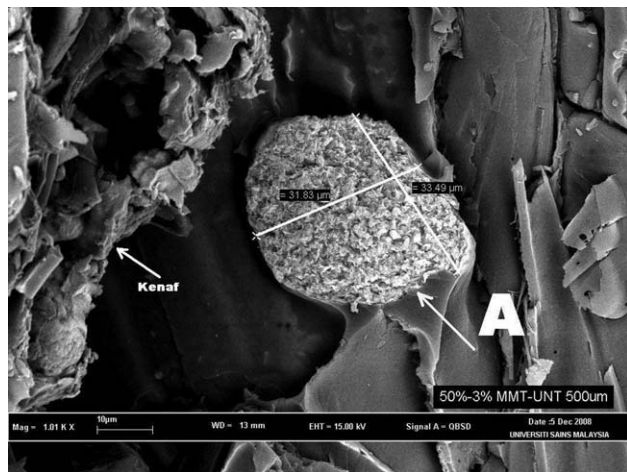


Figure 9 SEM micrograph of the kenaf/USP with 50% filler (kenaf filler size = 500–250 μm) and 3% MMT (magnification = 1010 \times).

Figure 9 shows the micrograph of the sample with a 50% filler loading (kenaf filler size = 500 μm) with 3% MMT. It was obvious that the MMT filler, although in an agglomerated form, was coated with the matrix material (arrow A). A good example of compatibility is shown by the arrow A, which we believe to be due to the interaction of OH groups on the MMT surface with oxygen atoms from the C=O of maleic residues of the USP. The proof that the filler was MMT was shown in the EDX analysis (Table I).

CONCLUSIONS

In general, this study showed that, for samples with kenaf filler only, the strength properties decreased as the kenaf filler loading was increased from 40 to 60%. This resulted in the reduction of the amount of matrix material. This subsequently lowered the ability of the sample to absorb energy or distribute stress efficiently. However, with MMT, the tensile properties improved because of the high aspect ratio

and surface area of the MMT. The study of the effect of kenaf filler size on the tensile properties showed that the samples with the smallest size, 74 μm , showed the lowest tensile properties compared to those with the larger ones. This was attributed to agglomeration of the kenaf fillers. The addition of MMT resulted in an overall increase in the tensile strength of the composite as compared to those without MMT.

References

- Okudaira, Y. Seminar on Kenaf: A New Source of Growth; Le Meridien: Kuala Lumpur, Malaysia, 2005.
- Fu, S. Y.; Lauke, B.; Mader, E.; Yue, C. Y.; Hu, X. *Compos A* 2000, 31, 1117.
- Sreekala, M. S.; Thomas, S. *Compos Sci Technol* 2003, 63, 861.
- Wambua, P.; Ivens, J.; Verpoest, I. *Compos Sci Technol* 2003, 63, 1259.
- Shibata, S.; Cao, Y.; Fukumoto, I. *Polym Test* 2006, 25, 142.
- Bolton, A. J. *Mater Technol* 1994, 9, 12.
- Balasuriya, P. W.; Ye, L.; Mai, Y. W. *Compos A* 2001, 32, 619.
- Doan, T.-T.-L.; Gao, S.-L.; Mader, E. *Compos Sci Technol* 2006, 66, 952.
- Felix, J. M.; Gatenholm, P. *J Appl Polym Sci* 1991, 42, 609.
- Rozman, H. D.; Tan, K. W.; Kumar, R. N.; Abubakar, A.; Mohd Ishak, Z. A.; Ismail, H. *Eur Polym J* 2000, 36, 1483.
- Yang, H. S.; Kim, H. J.; Son, J.; Park, H. J.; Lee, B. J.; Hwang, T. K. *Compos Struct* 2004, 63, 305.
- Forness, T. D.; Yoon, P. J.; Keskkula, H.; Paul, D. R. *Polymer* 2001, 42, 9929.
- Kotek, J.; Kelnar, I.; Studenovskiy, M.; Baldrian, J. *Polymer* 2005, 46, 4876.
- Lee, S. R.; Park, H. M.; Lim, H. L.; Kang, T.; Li, X.; Cho, W. J.; Ha, C. S. *Polymer* 2002, 43, 2495.
- Liu, X.; Wu, Q. *Polymer* 2001, 42, 10013.
- Wang, Z.; Lan, T.; Pinnavaia, T. J. *Chem Mater* 1996, 8, 2200.
- Lebaron, P. C.; Wang, Z.; Pinnavaia, T. J. *J Appl Clay Sci* 1999, 15, 11.
- Rozman, H. D.; Musa, L.; Abubakar, A. *Polym Plast Technol Eng* 2005, 44, 489.
- Devi, L. U.; Bhagawan, S. S.; Thomas, S. *J Appl Polym Sci* 1997, 64, 1739.
- Rozman, H. D.; Musa, L.; Abubakar, A. *J Appl Polym Sci* 2005, 97, 1237.
- Zhang, M.; Singh, R. P. *Mater Lett* 2004, 58, 408.
- Kaynak, C.; Tasan, C. C. *Eur Polym J* 2006, 42, 1908.