A Comparison Study of Lignocellulosic–Thermoplastic Composites Prepared from Different Compounding Techniques

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ABSTRACT: A comparison study was carried out to determine the effect of different types of compounding technique, i.e., internal mixer, twin screw extruder, and high speed mixer in the preparation of kenaf-polypropylene composite. The effect of percentage kenaf loading and particle size of kenaf (core) on the flexural properties of the composite was investigated. From the results, the incorporation of kenaf, regardless of particle size had resulted in the reduction of flexural strength of the composite. However, flexural modulus of the composites increased as the percentage of kenaf loading was increased due to the increasing of the stiffness contributed by kenaf fiber. Composites produced from internal mixer had displayed higher flexural properties as compared to those prepared from high speed mixer and twin screw extruder. It was believed that this phenomenon was attributed to the effectiveness of internal mixer with a better compounding mechanism which improved the wetting and distribution on kenaf within the polypropylene matrix. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 124: 4547–4553, 2012

Key words: compounding technique; kenaf; lignocellulosic–thermoplastic composite

INTRODUCTION

Synthetic fibers, such as glass, carbon, and aramid fibers, have been utilized as composite materials in various industries. Among which, glass fiber is widely used in the preparation of polymer composite for automotive and construction industry due to its lower cost and better mechanical properties. However, the use of glass fiber has several drawbacks such as nonbiodegradability, poor recycling properties, health risk if inhaled, high density, and high energy consumption in the preparation of its products.[1] Owing to the issue of sustainability and concern of the environment, which has been high on the worldwide agenda lately, the trend is slowly changing towards the use of lignocellulosic materials as a reinforcing agent in most polymer composites, which are known as green composites.[2–13] According to Clemons, the perspective of plastic industries has been changed, attributed to the success of wood plastic composites product. This development is also due to greater understanding of wood, equipment development and opportunities to enter new markets.[14] In addition, the need for efficient use of resources has become vital with the pressures on the forest industries coupled with the scarcity of natural resources. Thus, many attempts have been made to utilize lignocellulose material (nonwood) in various industries and lignocellulose-based polymer composite is one of them.

In Malaysia, kenaf is a new type of agriculture crop which can produce fibers with excellent strength and has great potential to be used as a raw material for nonwoven material.[8] In addition, kenaf has been identified by the Government and the National Kenaf, Tobacco Board as a strategic crop recently. Kenaf has long been recognized as a potential raw material for various types of value-added product. It is believed that these lignocellulosic materials could be utilized in automotive and building market. But, research and technology in this area is still in its infancy. Hence, more research and developments need to be carried out.

Furthermore, Zampolani et al.[15] compared different types of natural fiber–polypropylene composites in terms of flexural properties. Comparisons were made with 40% natural fiber of flax, kenaf, hemp, coir, and sisal. From the result, flax/PP composites showed higher flexural strength followed by kenaf/PP, hemp/PP, coir/PP, and sisal/PP composite. It was shown that, kenaf fibers have a potential to be used in the preparation of lignocellulose–thermoplastic composite.

Homogenous mixture between matrix and filler/fiber must be achieved in the preparation of a good

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lignocellulosic–thermoplastic composite, in terms of mechanical properties. Without this, stress concentration point will be easily created in the composite system, which subsequently initiates the crack propagation and reduces the mechanical properties of composite.[6–10] Hence, the mixing technique of thermoplastic–lignocellulosic composite preparation is vital in ensuring that a homogenous system. According to Clemons (2002), extrusion is the most common technique for wood thermoplastic composite production with different types of extruder and processing strategy. The approaches taken by various manufactures can be from single step to produce a final product directly, or by use of a multistep method for compounding and profiling on multiple extruders or processing equipment. From the previous study,[13] single screw extruder and internal mixer were used to compare the efficiency of mixing technique to produce thermoplastic–lignocellulosic composite. The composite prepared by internal mixer showed better mechanical properties than the one prepared by single screw extruder. This may be attributed to the high shearing mixing of internal mixer which produces better compounding and subsequently improves the wetting of the filler surface. It is believed that poor filler–matrix interaction or compatibility is responsible for the poor ultimate performance. Hence, it can be said that compounding technique plays a significant role in manufacturing lignocellulosic–thermoplastic composite.

In this study, a comparison study was carried out to determine the effectiveness of different compounding techniques, i.e. twin screw extruder, internal mixer and high speed mixer to produce lignocellulosic–thermoplastic composite based on kenaf (core). In addition, the effect of kenaf (core) particle size and kenaf loading on mechanical properties of composites prepared from different technique was also investigated.

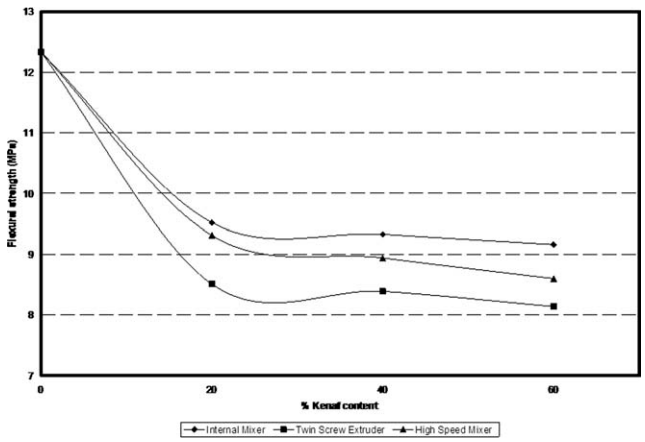

Figure 1 The effect of kenaf loading (mesh number of 35–60) and mixing technique on flexural strength of kenaf-PP composite.

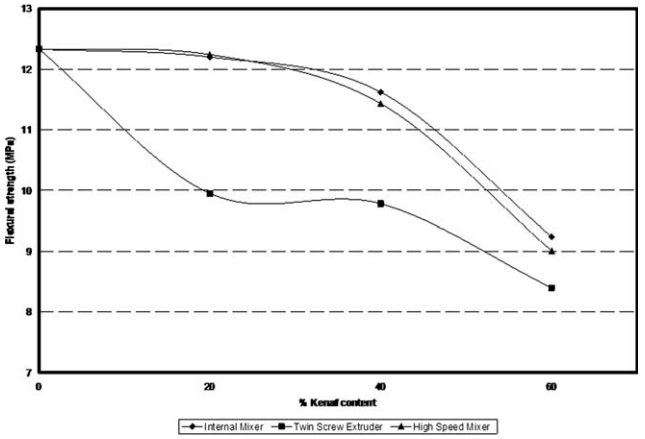

Figure 2 The effect of kenaf loading (mesh number of 60–80) and mixing technique on flexural strength of kenaf-PP composite.

EXPERIMENTAL

Materials

Polypropylene (PP) impact copolymer Titanpro SM240 was purchased from Titan Petchem (M) Sdn. Bhd to be used as matrix. Its density was 0.9 g/cm^3 and melt flow index was 25 g/10 min at 230°C. Kenaf core was provided by National Kenaf, Tobacco Board, Malaysia.

Preparation of kenaf (core) particle

Kenaf core were ground by Lab Wood Chipper Model WRB90LB/4P and dried in an oven at 105°C ± 5 for 24 h prior to sieving process. Particles were sieved with Retsch Test Sieve Model 5667 to three ranges of sizes with mesh number of 35–60, 60–80, and 80–270, respectively.

Compounding techniques

Lignocellulosic thermoplastic composites were prepared using different compounding techniques, i.e.,

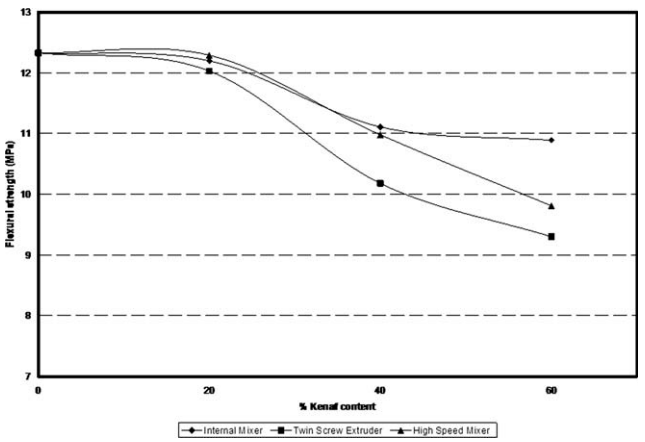

Figure 3 The effect of kenaf loading (mesh number of 80–270) and mixing technique on flexural strength of kenaf-PP composite.

TABLE I
Average Aspect Ratio of kenaf (core) with Different Particle Size.

Mesh no.	Average aspect ratio
35–60	2.77 (±0.17)
60–80	3.08 (±0.21)
80–270	3.52 (±0.15)

internal mixer, twin screw extruder and high speed mixer. The percentage of kenaf incorporated into was varied from 20, 40, and 60%.

Internal mixer

Internal mixer, Haake Rheomix 3000 was used to mix kenaf core particles and PP at 180°C. The mixing was performed in the following order. Polypropylene was first gradually added into the mixing chamber with a rotor speed of 30 rpm. kenaf core particle was added when PP was in a complete molten form. The rotor speed was increased to 40 rpm and the mixture was further mixed for ~ 10 min. The premix compound was then taken out from the mixer and kept for further process.

Twin screw extruder

PP and kenaf were dry-mixed before being fed into twin screw extruder. The mixing was carried out at 180°C from the feeding zone to the die zone, at a rotor speed at 35 rpm. The mixture was then extruded and pelletized.

High speed mixer

PP and kenaf was fed into high speed mixer chamber (volume of mixing chamber = 5000 cm^3) with a rotor speed of 4000 rpm for approximately 20 min to ensure kenaf particle were evenly mixed with the molten PP. The resulting mixture was taken out from the mixing chamber and kept for further process.

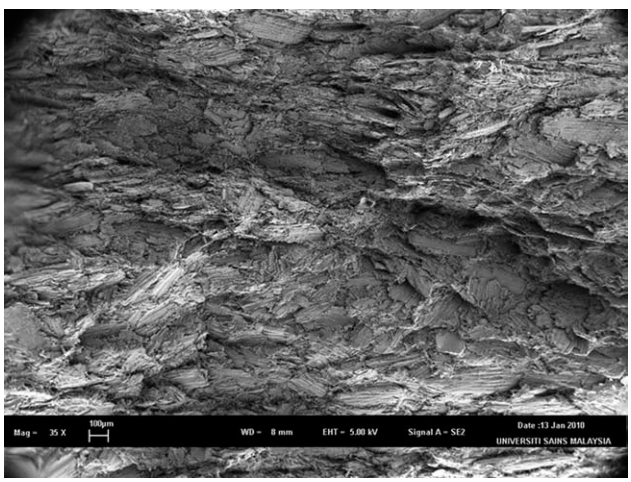

Figure 4 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using IM.

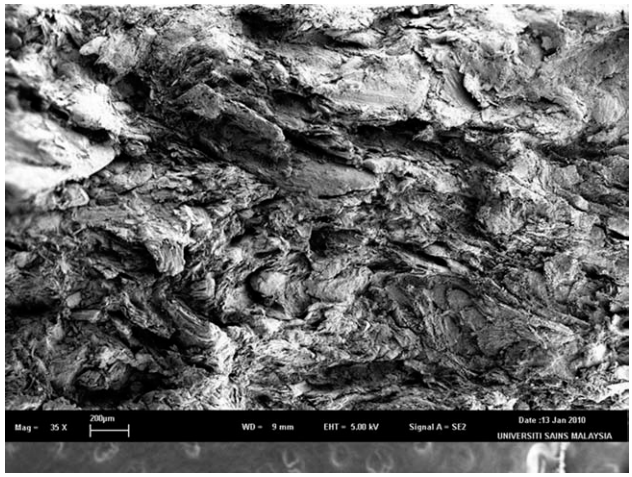

Figure 5 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using HSM.

Composite preparation

The mixture was hot-pressed using Carver Laboratory Press, Model-m 250 ton capacity, and the mold used was made from alloy with dimension of 180 mm × 180 mm × 3 mm (length × width × thickness). Pressing process was divided into three stages; preheating, hot pressing, and cold pressing. Preheating was carried out for 10 min before the sample was further subjected to 180°C with a pressure of 8 kgf/ cm^2 for 15 min. Then, the samples were cold pressed under the same pressure for ~ 15 min.

Flexural test

Flexural test (three point bending) was conducted according to ASTM D790 using Instron Machine,

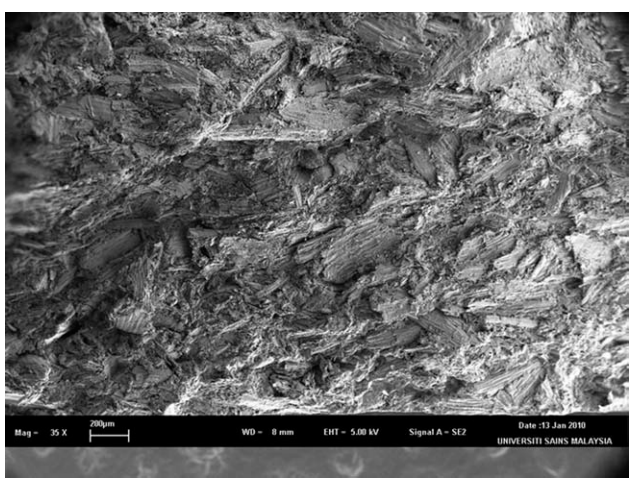

Figure 6 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using TSE.

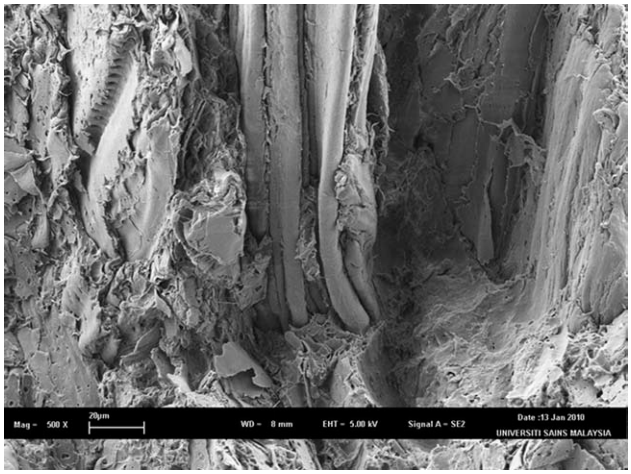

Figure 7 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using IM.

Model 5582. Flexural test involve flexural strength, flexural modulus, and flexural toughness. Samples were cut into dimension of 150 mm × 15 mm × 3 mm (length × width × thickness). Five replicates were tested. The crosshead speed was 2 mm/min and the span length was 48 mm.

Scanning electron microscopy study

The morphology study was carried out using a field-emission scanning electron microscope (FE-SEM) Leo Supra 50VP. The fractures obtained from the tensile test were used for SEM study and dried in an oven at 105°C ± 5 for 24 h. The specimens were mounted on an aluminium stub and sputter coated with a thin layer of gold to avoid electrostatic charging during examination.

Melt flow index

Melt Flow Index (MFI) analysis was conducted using Gotech GT-7100-MIB Melt Flow Indexer with manual Miworks software based on ASTM D1238 standard. Parameter involved were analysis temperature of 230°C, barrel preheating time of 240 s, die length of 8 mm, die diameter of 2.096 mm, load 2.16 kg, and time interval of extrudate cutting was 5 s. Five replicates were tested for each sample.

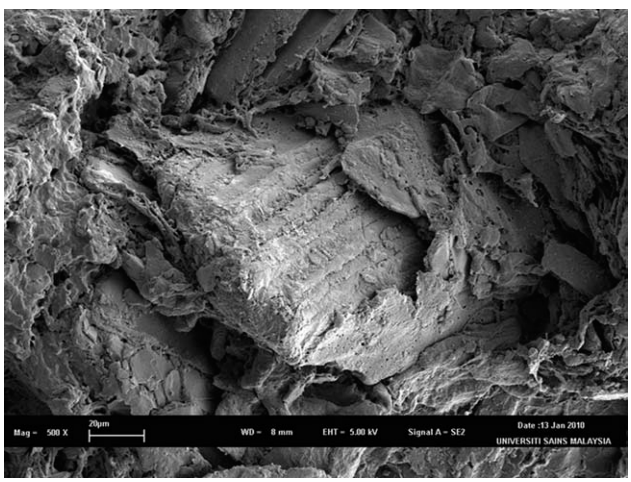

Figure 8 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using HSM.

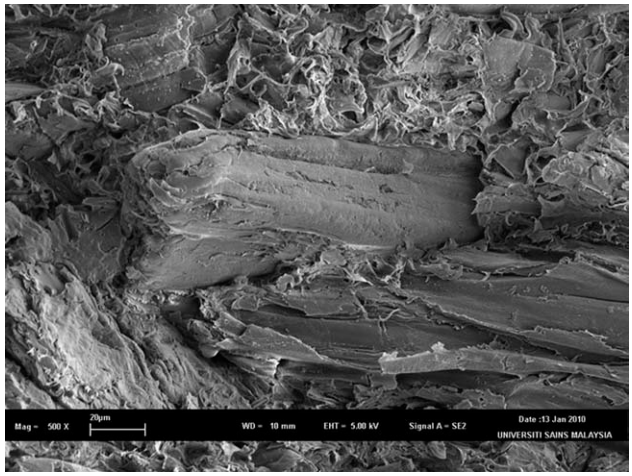

Figure 9 SEM micrograph of kenaf-PP composite with filler mesh number of 80–270 using TSE.

RESULTS AND DISCUSSION

The performance of kenaf-PP composites in flexural strength are presented in Figures 1–3. In general; flexural strength of the composite decreases as the kenaf loading is increased, regardless the kenaf particle size and mixing technique. These results indicate a common phenomenon where the strength of lignocellulosic–thermoplastic composites decreases as the content of lignocellulosic material is increased.[5–7] This is expected due to the hydrophilic nature of kenaf that could not form a good interaction with hydrophobic thermoplastic matrix. This weak interaction facilitates the formation of stress

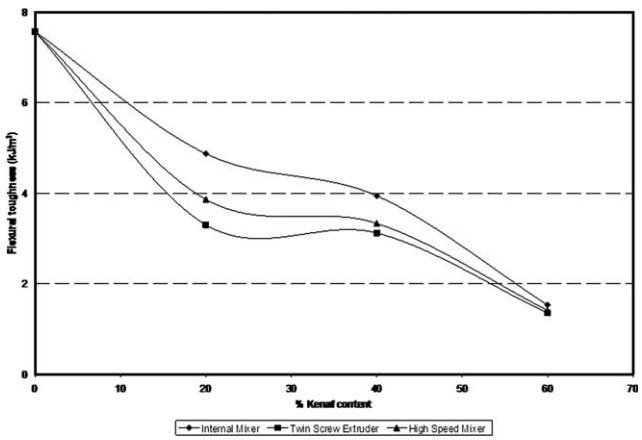

Figure 10 The effect of kenaf loading (mesh number of 35–60) and mixing technique on flexural toughness of kenaf-PP composite.

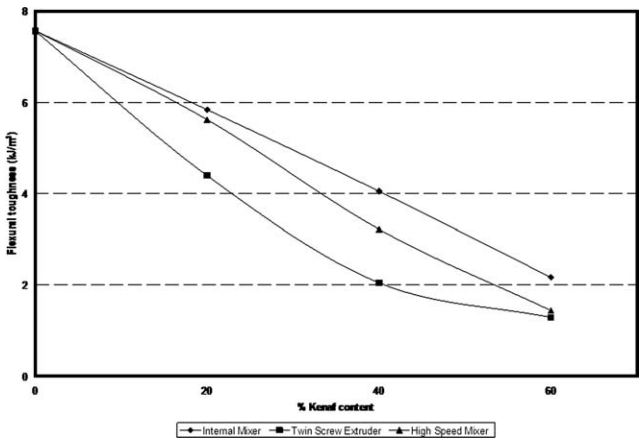

Figure 11 The effect of kenaf loading (mesh number of 60–80) and mixing technique on flexural toughness of kenaf-PP composite.

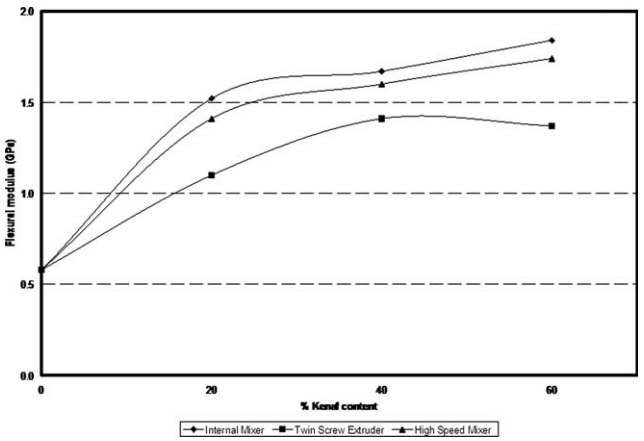

Figure 13 The effect of kenaf loading (mesh number of 35–60) and mixing technique on flexural modulus of kenaf-PP composite.

concentration points and subsequently reduces the stress transfer efficiency from the matrix to the filler. The efficiency is further reduced with the irregular shaped and low aspect ratio of the filler.[13] As for the effect of kenaf particle size on the flexural strength of the composites, generally, it can be seen that composites produced from small kenaf particle size exhibit higher flexural strength. This observation is in agreement with previous study[16] where smaller particle size has a greater surface area which could have higher filler–matrix interaction. In addition, the homogeneity of the kenaf-PP composites is improved when kenaf with smaller particle size is employed. Furthermore, the aspect ratio of kenaf is greater for smallest particle size as compared with bigger particle size as depicted in Table 1. This phenomenon has enhanced the properties of the composites prepared.

The efficiency of three mixing technique is also shown in Figures 1–3. It is clearly seen that composites prepared from internal mixer (IM) possess higher strength, followed by high speed mixer (HSM) and twin screw extruder (TSE). It is known that the mixing technique plays a crucial factor in determining mechanical strength as well as physical properties of a lignocellulosic–thermoplastic composite. Mixing technique influences the homogeneity of kenaf filler in thermoplastic polymer. It is known that good dispersion and wetting of the filler in thermoplastic matrix gives rise to better interfacial adhesion. Figures 4–6 depict SEM micrographs at low magnification (35 X) for composite prepared from 60% kenaf filler with mesh number of 80–270 using different mixing techniques. It seem like all mixing techniques could produce a composite with satisfactory distribution of kenaf filler. By comparing the wettability of kenaf filler in PP matrix, at higher magnification (500 X) as shown in Figures 7–9, it can be seen that the kenaf filler wetting in PP matrix in composite produced using IM is better than those produced by HSM and TSE. Thus, it is expected that

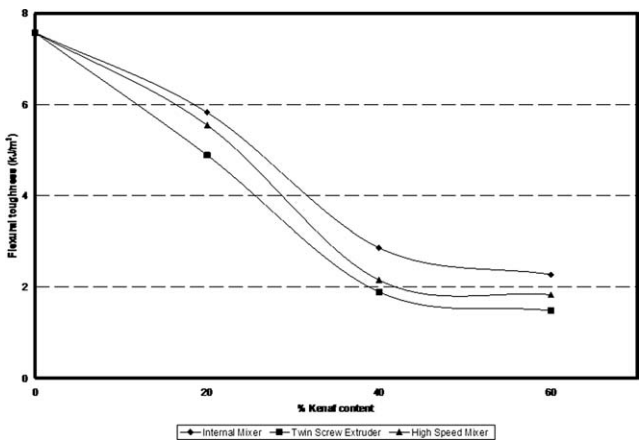

Figure 12 The effect of kenaf loading (mesh number of 80–270) and mixing technique on flexural toughness of kenaf-PP composite.

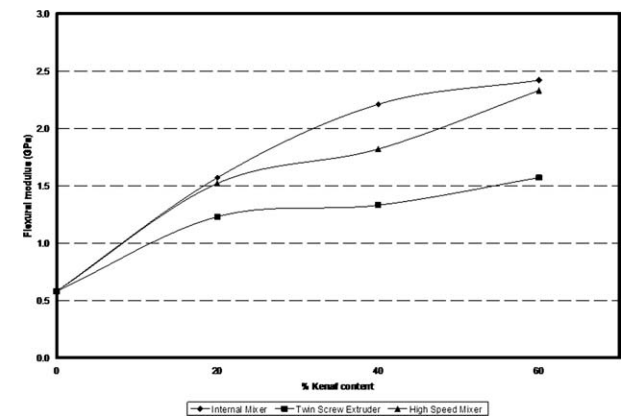

Figure 14 The effect of kenaf loading (mesh number of 60–80) and mixing technique on flexural modulus of kenaf-PP composite.

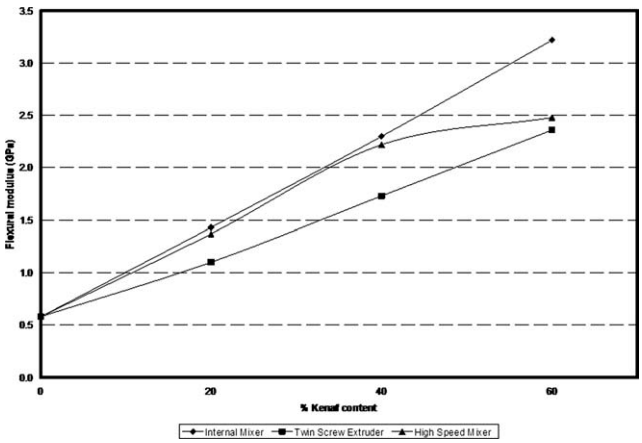

Figure 15 The effect of kenaf loading (mesh number of 80–270) and mixing technique on flexural modulus of kenaf-PP composite.

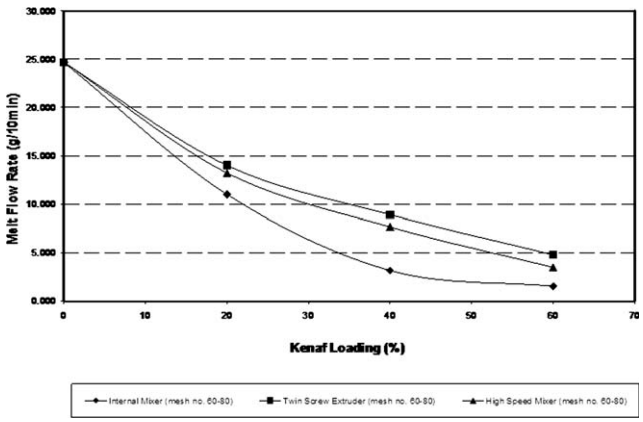

Figure 17 The effect of kenaf loading (mesh number of 60–80) and mixing technique on MFI of kenaf-PP composite.

composites prepared using IM has a better stress transfer mechanism as compared to those using HSM and TSE. It is clearly seen that the filler is tightly embedded in the matrix for IM samples as compared with those with HSM and TSE. This indicates that the high shear mixing by two counter-rotating rotors in an enclosed mixing chamber has resulted in a better interaction between filler and matrix. It is interesting to see the degree of wetting of the filler and the matrix for those with HSM and TSE are quite comparable, though the mixing mechanisms of both techniques are different.

Figures 10–12 represent flexural toughness of the composites prepared from kenaf filler with different mesh number using different mixing techniques. It can be seen that the toughness decreases when more kenaf filler is added. This trend is similar with the observation in flexural strength (Fig. 1). From previous study[13] it is found irregular shape of lignocellulosic filler with low aspect ratio reduces the composite capability to absorb energy of fracture. As toughness represents the energy needed to fail a sample, thus, less energy is needed to bring the sample to failure when more kenaf filler is incorporated. This is related to the poor interaction between kenaf and PP due to the incompatibility between them. As for mixing technique, composite produced using IM exhibits better performance than the others. This may be attributed to a better dispersion and mixing of kenaf filler in the composite.

Figure 13 displays the stiffness of kenaf-PP composite prepared from kenaf filler of 35–60 mesh number using different compounding technique. In contrast with strength and toughness results, flexural modulus of kenaf-PP composites increases as the kenaf loading is increased, irrespective of compounding technique. According to Tay et al. (2010), it is expected because lignocellulosic in general has its inherent stiffness, which is higher than the matrix. This inherent stiffness adds up with the stiffness of the matrix, resulting in higher modulus.[10] This is in agreement with the result shown Figures 14 and 15, where higher kenaf filler content

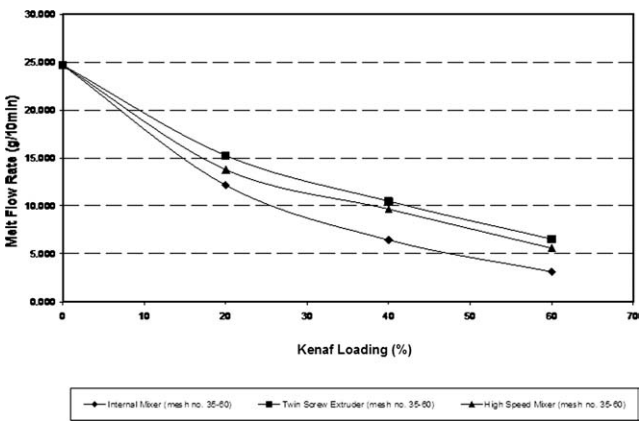

Figure 16 The effect of kenaf loading (mesh number of 35–60) and mixing technique on MFI of kenaf-PP composite.

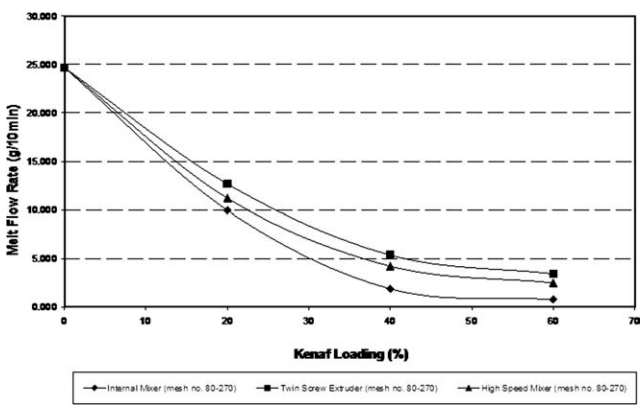

Figure 18 The effect of kenaf loading (mesh number of 80–270) and mixing technique on MFI of kenaf-PP composite.

produces composite with higher stiffness. However, the degree of increment is greater when small kenaf filler is used (by comparing Figs. 13–15). This is attributed to better wetting of kenaf filler in PP matrix.

The compounding technique has a significant effect on the flexural modulus of kenaf-PP composites. It is clearly seen that IM could produce composites with higher flexural modulus. It is known that the mixing process in IM which is carried out by two counter-rotating rotors gives higher shear rate and the compounding process is conducted in an enclosed and confined chamber. From the results, it is known that this mixing mode is more effective than HSM and TSE in enhancing the distribution of kenaf filler in PP matrix. A higher degree of kenaf filler wetting and better filler dispersion in PP matrix has contributed to the stiffness of kenaf-PP composite.

Figure 16 depicts the result of melt flow index (MFI) for kenaf-PP composites prepared from kenaf filler with mesh number of 35–60 using different compounding technique. Generally, all samples show a marked decrease in MFI value as the kenaf content is increased. The degree of reduction is obviously greater when smaller kenaf filler is employed (Figs. 17 and 18). This is due to a greater surface area of smaller size kenaf filler which leads to a greater interfacial interaction in PP matrix and subsequently lower MFI value. The behavior is further magnified MFI value if kenaf filler and PP are compounded in IM. This is again due to an improved in mixing of the kenaf filler in PP matrix.

CONCLUSION

Kenaf-PP composites were prepared using different compounding techniques, i.e., IM, HSM, and TSE. It was found that composites prepared using IM possessed higher flexural strength and stiffness. This phenomenon indicated that IM had higher efficiency to produce composites with better filler dispersion and wettability. This was due to the higher shear rate and an enclosed system of IM mixing chamber which had resulted in an improved homogeneity of the compounding. Smaller kenaf particle size also showed higher flexural properties than those with bigger ones. This was due to a better interaction with PP matrix. However, the incompatibility of hydrophilic surface of kenaf filler and hydrophobic PP had resulted in the reduction of flexural properties when kenaf filler content was increased.

References

1. Wambua, P.; Ivens, J.; Verpoest, I. Compos Sci Technol 2003, 63, 1259.
2. Mohanty, A. K.; Misra, M.; Drzal, L. T. J Polym Environ 2002, 10, 19.
3. Mohd. Ishak, Z. A.; Yow, B. N.; Ng, B. L.; Khalil, H. P. S. A.; Rozman, H. D. J Appl Polym Sci 2001, 81, 742.
4. Stark, N.; Rowlands, R. Wood Fiber Sci 2003, 35, 167.
5. Rozman, H. D.; Tay, G. S.; Kumar, R. N.; Abubakar, A.; Ismail, H.; Mohd. Ishak, Z. A. Polym-Plastic Technol Eng 1999, 38, 997.
6. Rozman, H. D.; Tan, K. W.; Kumar, R. N.; Tay, G. S.; Abubakar, A. Int J Polym Mater 2000, 46, 195.
7. Rozman, H. D.; Tay, G. S.; Kumar, R. N.; Abusamah, A.; Ismail, H. Eur Polym J 2001, 37, 1283.
8. Rozman, H. D.; Tay, G. S.; Kumar, R. N.; Abusamah, A.; Ismail, H.; Mohd. Ishak, Z. A. Polym-Plastic Technol Eng 2001, 40, 103.
9. Rozman, H. D.; Zuliahani, A.; Tay, G. S. J Appl Polym Sci 2010, 115, 3456.
10. Tay, G. S.; Mohd. Zaim, J.; Rozman, H. D. J Appl Polym Sci 2010, 116, 1867.
11. John, M. J.; Anandjiwala, R. D. Compos Part A: Appl Sci Manufact 2009, 40, 442.
12. Rahman, M. R.; Huque, M. M.; Islam, M. N.; Hasan, M. Compos: Part A 2008, 39, 1739.
13. Rozman, H. D.; Peng, G. B.; Mohd. Ishak, Z. A. J Appl Polym Sci 1998, 70, 2647.
14. Clemons, C. Forest Product J 2002, 52, 10.
15. Zampaloni, A.; Pourboghrat, F.; Yankovich, S. A.; Rodgers, B. N.; Moore, J.; Drzal, L. T.; Mohanty, A. K.; Misra, M. Compos Part A: Appl Sci Manufact 2007, 38, 1569.
16. Rozman, H. D.; Ismail, H.; Jafrri, R. M.; Aminullah, A.; Mohd Ishak, Z. A. Int J Polym Mater 1998, 39, 161.