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Micromechanical Property Investigations of Poly(lactic acid)–Kenaf Fiber Biocomposites

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In this research, investigation on the interfacial shear strength of poly(lactic acid)–kenaf fiber biocomposite was investigated using microbond tests. Tensile properties and fracture behaviors of single kenaf fiber are tested via in situ monitoring with acoustic emission (AE). During tensile loading, acoustic signal recorded higher amplitude of above 20 dB up to the maximum force, which corresponds to breakage of single kenaf fiber. Based on microbond tests and AE evaluation, a correlation has been established on failure of kenaf fiber, which is due to debonding of filament and internal structure, cracking of fiber and breakage of fiber.

KEYWORDS poly(lactic acid), kenaf fiber, microbond, acoustic emission, interfacial shear strength

INTRODUCTION

Composite materials based on all-cellulose are getting more and more attention. Focus is now given to environmental aspects: renewable, biodegradable, compostable, and sustainable. Apart from this, the lightness of materials is also the driving force for their usage. Poly(lactic acid) (PLA) is one of the most studied matrix materials because of its promising mechanical properties. Although in the last few decades glass fiber has received considerable attention, natural fibers, such as flax, hemp, sisal, jute, and kenaf, are the

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most attracted fibers in the 21st century. Development of all-cellulose composite based on kenaf fibers has been reported relatively less compared to flax, hemp, sisal, and jute fibers. The hub of different studies lies in the mechanical and thermal characterization, while the important feature that binds the matrix and fiber together leading to interfacial adhesion is reported less in the open literature.

Mechanical performance of composite materials strongly depends on the properties of the fiber and matrix. Excellent properties of each constituent can be efficiently transferred from one phase to another if the interface between fiber and matrix is good. Up-to-date research on micromechanical and interfacial adhesion of natural fiber and biodegradable matrix, however, is hardly found until recently, which has been reported by Le Duigou and coworkers (2010) on flax fiber and PLA matrix.

Le Duigou et al. (2010) summarized that the characterization of interfacial region is very complex and can be performed at different levels that is nanoscopic, microscopic, and macroscopic. On top of that investigation on surface adhesion between single fiber and matrix by means of micromechanical tests can be classified into two groups, namely, direct and indirect methods (Morlin and Czigány 2005). Examples of direct methods include fragmentation (Greenfield et al 2000), Broutman test (Broutman 1969), fiber push-out (Zhou et al. 2001), fiber pullout and microbond test (Day and Cauich Rodrigez 1998; Eichhorn and Young 2004; Zinck et al. 2001). On the other hand, indirect methods include conventional mechanical testing and acoustic emission (AE). There are various parameters, such as the effect of matrix chemistry, fiber surface treatments, and fiber diameter, that can be evaluated during characterization of the interface on the behavior of composite materials (Zinck et al. 2001). Day and Cauich Rodrigez (1998) found that the position of the grips, which support the microdroplet, is important and affects the interfacial shear stress distribution. In addition, they also suggested using two droplets where one of these will be restrained while the other will be displaced. The load–displacement curve will be recorded and the change in stiffness indicates the load at which yielding or debonding occurred. This load can then be used to calculate the interfacial shear strength using shear lag model.

The acoustic emission method offers various advantages compared to other nondestructive testing methods (Anuar et al. 2007; Barre and Benzeggagh 1994; Haselbach and Lauke 2003; Joung-Man Park et al. 2006; Miller and McIntire 1987; Romhany et al. 2003). This method is capable of detecting dynamic processes associated with the degradation of structural integrity. Typically, certain areas within structural system will develop local instabilities before the structure fails. In macrocomposite, for example, AE method has been successfully used to monitor the types of fracture sources and their progress by analyzing AE parameters such as AE energy, AE amplitude, and their frequency emitted from the fiber fracture, matrix cracking, fibrillation, and debonding. Barre and Benzeggagh (1994) reported on glass fiber-reinforced polypropylene that acoustic signal amplitude varies corresponding to damage mode. In general, the AE energy because of fracture of fiber is larger than the cracking of matrix and debonding (Anuar et al. 2007; Romhany et al. 2003). Park et al. (2006) has used AE to describe microfailure of jute and hemp fibers. They reported that microfailure with fibril splitting occurred for jute fiber, whereas final fiber fracture was seen for hemp fiber.

The purpose of this paper is to investigate the applicability of microbond tests to determine surface adhesion of kenaf fiber and PLA matrix by adopted test method developed by Morlin and Czigány (2005). In addition, this paper also dealt with single fiber tests via in situ monitoring using AE to characterize failure behavior of kenaf fiber.

EXPERIMENTAL

Materials

Poly(lactic acid) of injection molding grade (3051D) used for the experiment was obtained from Nature Works Ltd. (China). The kenaf bast fiber was supplied by Kenaf Natural Fiber Industries (Sdn. Bhd., Kelantan, Malaysia) in the form of long fiber and was harvested at the age of 6 months.

Methodologies

Microbond Test

Microbond test is a method that is used to measure interfacial shear strength between fiber and matrix. Microbond tests were conducted using a microbond device and fixed onto the Zwick 005 tensile tester. The device contains two steel blades that can be positioned with micrometers. The role of the steel blades is to support the droplets and hold them during the debonding process as shown in Figure 1.

A microdroplet of PLA is placed onto the kenaf fiber, and the diameter of the microbond (D), length of the microbond (L_0) and diameter of kenaf fibers (d_f) were determined using optical microscope (Olympus BX51) attached to photo-camera (C-5060). Figure 2 shows photomicrograph of kenaf fiber with PLA matrix droplet. Forty specimens were prepared for the microbond tests. If shear stress is constant along the interface, the average values of interfacial shear strength were calculated by using Equation 1. From the microbond tests, the maximum force ($F_{\max}$) was measured during pull out of fiber.

$$\tau = \frac{F_{\max}}{d_f \cdot \Pi \cdot L_0} \tag{1}$$

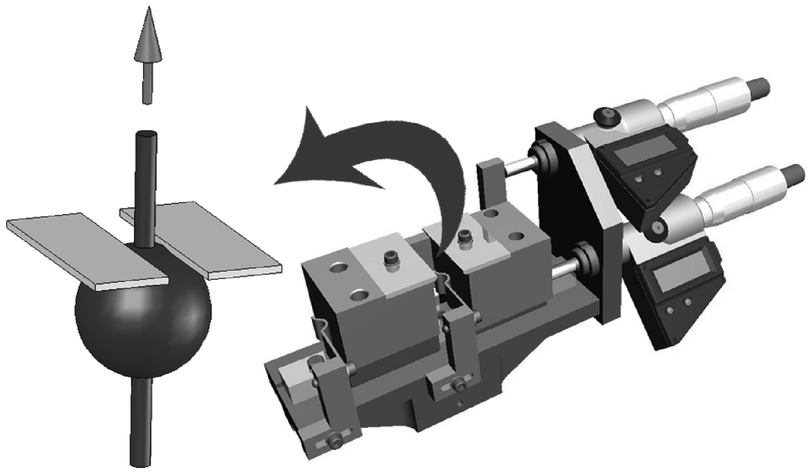

FIGURE 1 Schematic diagram shows microdroplet test arrangement.

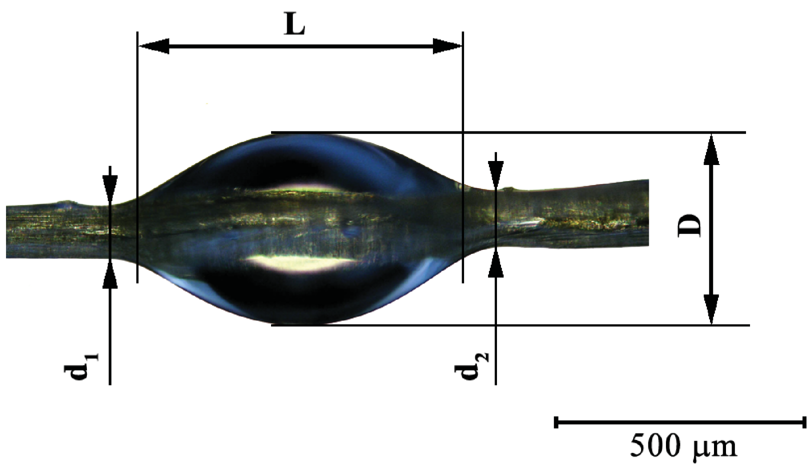

FIGURE 2 Kenaf fibers with PLA matrix droplet.

Single kenaf fiber test

Single fiber test was used to measure the strength and stiffness of the single fiber. This test was carried out using Zwick Z005 tensile tester with 20-N load cell, and strain rate used was 0.5 mm/min. Initial length of kenaf fiber, based on window frame, was 25 mm as shown in Figure 3. Thirty samples were prepared and tested.

Acoustic emission (AE)

In order to understand the fracture behavior of kenaf fiber, AE measuring method was used in situ during loading of single fiber test. The arrangement

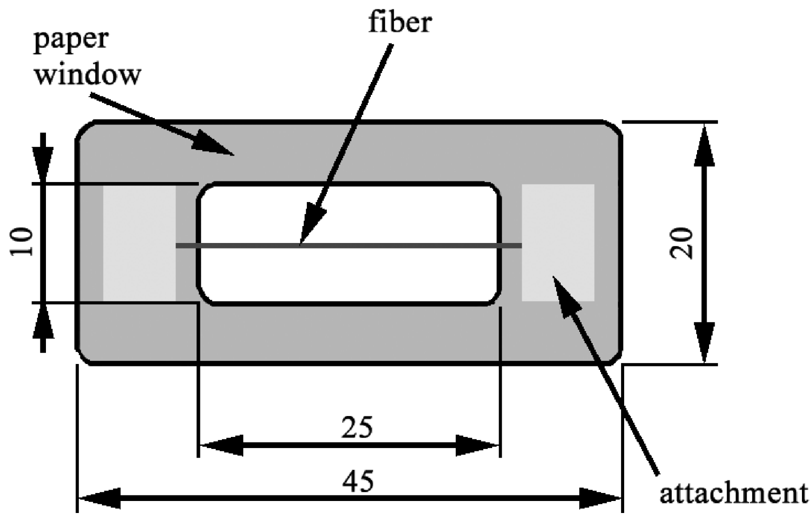

FIGURE 3 Window frame used in the preparation for single fiber tests.

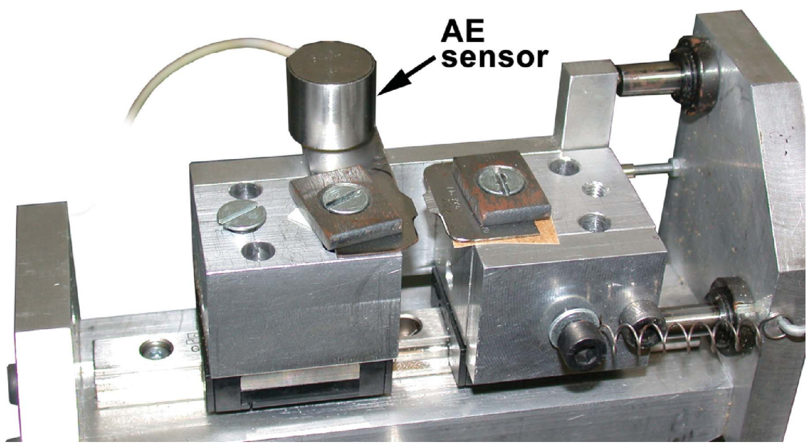

FIGURE 4 Arrangement of the AE microphone.

of the device used is shown in Figure 4. During the tests, the following primary AE signals were measured, calculated, and stored: elapsed time, number of events, peak amplitude, and event width and rise time. Different failure modes were assigned to the different signal levels determined based on the force–displacement curve and the physical parameters of the sound waves.

RESULTS AND DISCUSSION

Microbond Tests

The microbond test was carried out to measure the interfacial adhesion between single kenaf fiber and PLA matrix. Out of 40 specimens obtained, only 10 were successfully measured, and the rest broke down during the measurement. Details of the diameter of kenaf fiber, dimension of micro-droplet, and force needed for pulling out fiber from the matrix are tabulated in Table 1. It is noted that the force recorded during microbond tests varied in a wide range. This could be due to the uneven structure of natural fiber as shown in Figure 7. The average interfacial shear strength is 5.41 ± 2.23 MPa. The data obtained is comparable to PP-flax, PP-hemp, and PP-sisal as reported by Morlin and Czigany (2005). Tensile strength of the PLA matrix is 63.3 MPa. Hence, interfacial adhesion normalized to the tensile strength of the matrix is 8.55%.

The force–displacement curve of single kenaf fiber and PLA matrix is shown in Figure 5. Debonding has occurred at maximum force, F_{max}, followed by drastic failure, which is assigned to breakage of the fiber. At the end of microbond tests, friction of fiber matrix occurred due to uneven and bump surfaces of the kenaf fiber as shown in Figure 7.

Referring to the structure of kenaf fiber, as shown in Figure 7, generally surface of kenaf fiber are naturally rough and the dimensions are inconsistent. The first peak of Figure 6 is due to the debonding of the kenaf fiber and droplet and this is similar to what has been described in Figure 5. It is noted, however, in Figure 6 that there is a second peak present after the debonding. This phenomenon is due to the inconsistency of the diameter of kenaf fiber. After kenaf fiber and PLA droplet has debonded, the droplet starts to move on the fiber. The droplet, however, will set back by the roughness of fiber surface if the diameter of the fiber is bigger. This is the explanation of why the second peak is present, which is not related to the adhesion of kenaf fiber and PLA droplet.

Single Kenaf Fiber Test

Single kenaf fiber tests were carried out to characterize stress and elongation at break of kenaf fiber. Table 2 shows properties of single kenaf fiber. Typical force–displacement curve obtained from single kenaf fiber test is shown in Figure 8. Properties of single kenaf fiber are very useful to predict theoretical values of tensile properties and compare with the true values obtained from experiment. The density of kenaf fiber is lower than glass fiber (about 2.5 g/cm^3 (Bledzki and Gassan 1999)). Density of kenaf fiber measured is 1.13 g/cm^3. Hence, specific tensile strength and modulus are 117.30 MPa/g/cm^3 and 9.89 GPa/g/cm^3, respectively.

TABLE 1 Interfacial Shear Strength According to Equation (1)

	Kenaf fiber				Microdroplet		
	Diameter 1, d_1 (μm)	Diameter 2, d_2 (μm)	Average diameter, d_f (μm)		Diameter, D (μm)	Length, L (μm)	
1	125.10	109.30	117.20	0.0001172	613.00	823.00	0.000823
2	64.10	51.30	57.70	0.0000577	309.00	495.00	0.000495
3	57.80	90.00	73.90	0.0000739	51.80	816.00	0.000816
4	64.10	46.50	55.30	0.0000553	644.00	686.00	0.000686
5	83.40	83.40	83.40	0.0000834	478.00	609.00	0.000609
6	75.40	48.10	61.75	0.0000618	390.00	600.00	0.000600
7	76.90	62.60	69.75	0.0000698	445.00	662.00	0.000662
8	105.80	105.80	105.80	0.0001058	331.00	502.00	0.000502
9	123.50	155.40	139.45	0.0001395	486.00	829.00	0.000829
10	101.40	138.20	119.80	0.0001198	470.00	699.00	0.000699
11	84.90	83.50	84.20	0.0000842	542.00	715.00	0.000715
12	72.10	91.40	81.75	0.0000818	335.00	590.00	0.000590

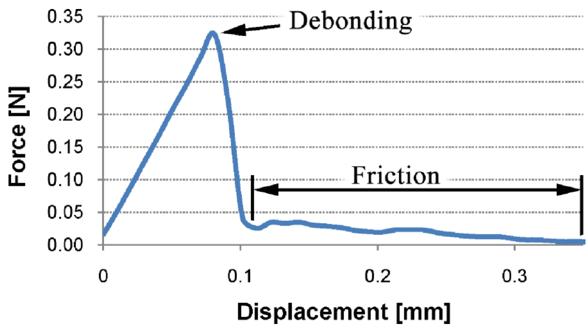

FIGURE 5 Force–displacement curve obtained from microbond tests for single kenaf fiber.

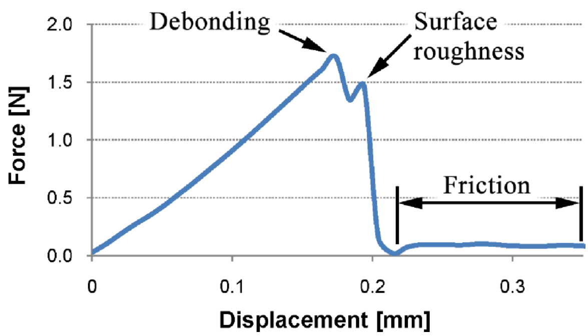

FIGURE 6 Force–displacement curve for uneven surface of kenaf fiber.

Acoustic Emission

Single filament kenaf fiber

In general, from the tensile tests, two types of force–displacement curves have been observed, which are associated with the characteristics of kenaf fiber as shown in Figure 7. The first type of force–displacement curve was observed for 60% of the kenaf fiber tested. Typical force–displacement curve for single kenaf fiber is illustrated in Figure 9. Generally, the count event was greater than 20 dB. The higher signal, which more than 20 dB, refers to the failure of the fiber. In the early part of the failure this can be related to the debonding in the internal structure of the kenaf fiber. The middle range of the event can be associated with the microcracking and debonding of the filament. The higher signal occurred in the range of 90–100 dB and can be assigned to the breakage of the kenaf fiber. Higher amplitude was seen at F_{max} and the range of signals was substantially higher than at the beginning of the test.

FIGURE 7 Photomicrographs of kenaf fiber under optical microscope.

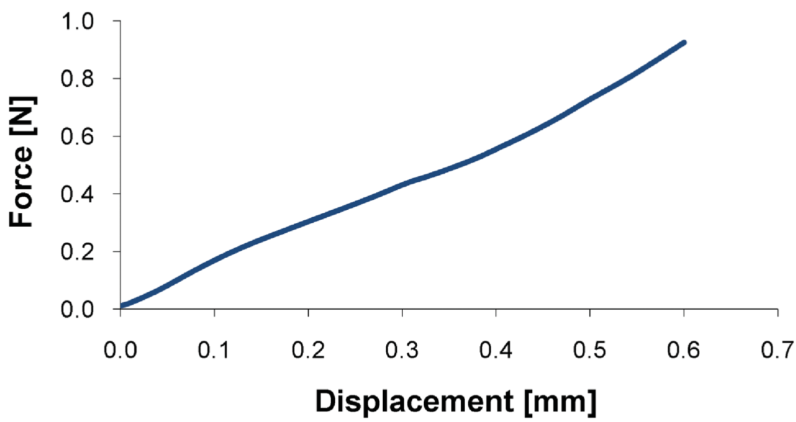

FIGURE 8 Typical tensile curve obtained from single kenaf fiber test.

MULTIFILAMENT KENAF FIBER

Out of 30 samples of kenaf fiber tested, 25% of the samples were classified as multifilament. Generally, the same observation, as seen in microbond tests for multifilament kenaf fiber, can be used to described activities in AE analysis. An example of force–displacement and amplitude–displacement plot of PLA–KF biocomposite for multifilament kenaf fiber with diameter of 86 μm is shown in Figure 10. The plot shows that in the early stage of applied force, lower amplitude signal was emitted. On increasing the elongation, toward middle of the tests, around 0.4 mm, mid-range signal (40–60 dB) were observed. This is caused by the breaks or debonding of smaller fibers from others. There is another group with higher signal of displacement of above 0.5 mm. This is the beginning of final fiber break

TABLE 2 Properties of Single Kenaf Fiber Determined from Single Fiber Test

Force (*N*)	0.72 ± 0.32
Elongation at break (mm)	0.39 ± 0.13
Strain at break (%)	1.56 ± 0.53
Tensile strength (MPa)	132.55 ± 18.80
Tensile modulus (GPa)	11.18 ± 2.44

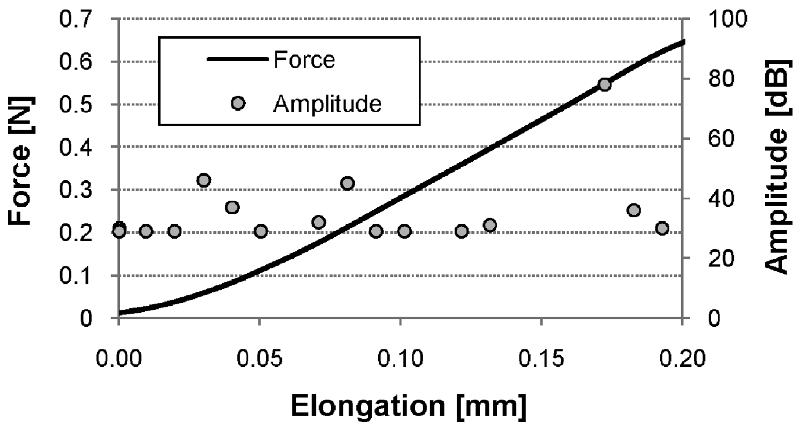

FIGURE 9 Typical force and amplitude versus displacement curve for single kenaf fiber.

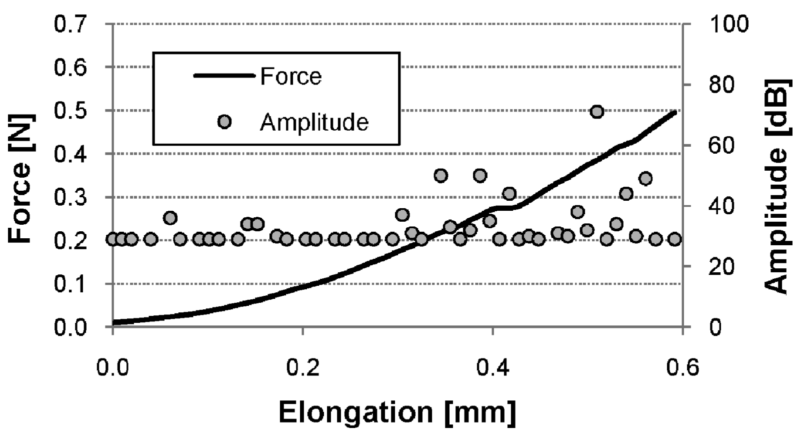

FIGURE 10 Typical force and amplitude versus displacement curve for multifilament kenaf fiber.

with the amplitude of above 90 dB. It is not a simple crack through the cross section because it can be seen that the force is increasing until F_{max}. This is initiation to the final failure. This process has caused some smaller AE events between 40 dB and 60 dB.

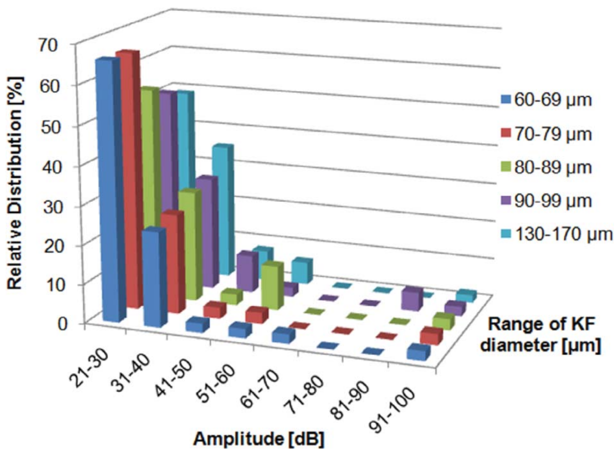

FIGURE 11 Amplitude distributions of events in single kenaf fiber.

The amplitude distribution of kenaf fiber tested is shown in Figure 11. Three single kenaf fibers were left out because fiber failed during the tensile testing. It was observed that the range of the amplitude was between 21 dB and 100 dB (20 dB was the environmental signal threshold level). Figure 11 illustrates the average AE event counts for various kenaf fiber diameters ranging from 60–170 μm. It is noted that on average the bigger the diameter, the lower the amplitude level, which decreases from approximately 70% (for diameter range of 60–80 μm) to virtually 54% (for diameter range of above 90 μm). Accordingly, the failure occurs due to debonding of filament, fiber cracking, and fiber breakage.

CONCLUSION

The micromechanical properties of PLA–KF biocomposite was investigated using microbond tests. Interfacial shear strength of PLA–KF is 5.41 ± 2.23 MPa. The value is comparable as those measured for hemp/PP and flax/Mater-Bi. For better understanding and to ensure accuracy of interfacial adhesion, however, it is suggested that future works shall be carried out on wetting of fiber and matrix via dynamic contact angle measurement. From single fiber tests, tensile strength and tensile modulus of single kenaf fiber are 132.55 ± 18.80 MPa and 11.18 ± 2.44 GPa, respectively. AE signal recorded lower amplitude at the early stage of single fiber test. This was due to debonding of filament structure and cracking of kenaf fiber. Higher amplitude (90–100 dB) assigned to fiber break was detected at the maximum force. The excellent mechanical properties of kenaf fiber shows that there is a great potential to use this fiber in high-performance polymer composites instead of glass fibers, particularly considering the light density of kenaf fiber and biodegradability of this system.

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