

## Effect of Fiber Content on the Properties of Natural Fiber Hybrid Composites

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**Abstract:** Natural fiber reinforced composites prepared by using an automatic hot press moulding machine. Jute/polyethylene composites and jute/kenaf/polyethylene hybrid composites with different fiber content (5, 10, 15 and 20 %) investigated. Physical properties of the hybrid composites were investigated using Fourier transform infrared spectroscopy (FTIR) and water absorption capacity. Universal testing machine used for investigating mechanical properties of this hybrid composite. Thermal properties of the composites were investigated by thermo-gravimetric analysis. Morphological properties of the composites were investigated by scanning electron microscopy. The SEM results showed that the significant difference was found between the single fiber reinforced composite and hybrid composites because of strong bonding between fiber and matrix. TGA and DSC result showed the higher thermal stability due to the higher activation energy of treated fibers/PE with POFA composite. Water Absorption capacity showed the reduced water absorption percentage for treated fibers/PE with PVA composites

**Keywords:** *Natural Fiber; SEM; FTIR; TGA; Tensile Strength*

**Introduction:** Many research works have been conducted and still ongoing as to the possibility of using natural fibers that are wholly degradable. The worldwide availability of natural fibers and other copiously accessible agro-waste is liable for new polymer science and engineering research and to invent for a sustainable technology. Natural fibers have numerous points of interest compared with glass fiber, for instance they are biodegradable and recyclable [1]. Natural fibers such as banana, coir, jute, sisal, kenaf and hemp gained the most attention of materials scientists and researchers for their application in automotive components. Natural fiber composites have lower densities and higher impact energy absorption capacity compared to glass fiber reinforced composites [2]. Natural fibers like kenaf and jute have some advantages over conventional reinforcement materials in terms of cost, density, renew ability; recycle ability, abrasiveness and biodegradability [3]. Hybridization is a system that overcomes the drawbacks of the composite materials reinforced by single natural fiber, such as low durability, poor resistance, and high water absorption of natural fibers with thermoplastic matrix [4]. Hybrid composite contain two or more types of fiber, a balance in cost and performance achieved through proper material design [5]. Recently hybrid composites are well-known as highly economical, high-performance structural materials and increasing application. Fiber content, length of individual fiber, orientation, extent of intermingling of fiber, fiber to matrix

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bonding and arrangement of both the individual fiber affect the mechanical, morphological and thermal properties of a hybrid composite.

Jute is a natural biodegradable renewable lignocellulose material. The compositions of jute fibers include mainly cellulose (58-63%), hemicellulose (20-22%), lignin (13-15%), pectin and trace masses of organic and inorganic pigments [6]. The maximum producers of jute are India, China, Thailand, Germany, Brazil and Bangladesh. Because of the rising ecological awareness, the improvement of biodegradable materials from renewable sources is easily available [7]. The accessibility of huge quantities of fiber with well-defined mechanical properties is a universal constraint for their productive use. Jute fibers are viable, low-density, low-cost, and lightweight [8]. Jute is traditionally used for packaging fabrics, sacking, mats, manufacturing hessian, carpet backing, bags, ropes, twines and tarpaulins etc. [9]. Kenaf is one of the common natural plant fibers which have been extensively used over the past few years. Kenaf has a bast fiber which contains 75% cellulose and 15% lignin and offers the advantages of being biodegradable and environmentally safe [10]. Kenaf fibers have the advantages of being available in large quantities, non-abrasiveness during processing, low density, it exhibits high specific mechanical properties and biodegradability [11].

#### **Materials & Method:**

**Jute and Kenaf fiber:** Raw jute fibers were collected from the Bangladesh Jute Research Institute (BJRI), Dhaka, Bangladesh. The compositions of jute fibers are mainly cellulose (45-71.5 wt%), hemicelluloses (13.6-21 wt%), and lignin (12-26 wt%) small amounts of pectin and trace masses of organic and inorganic pigments [12]. Raw kenaf fibers were collected from Malaysian Agricultural Research and Development Institute (MARDI).

**High Density Polyethylene (HDPE):** High density Polyethylene (HDPE), which used as the matrix material, was manufactured by Siam Polyethylene Co. (Thailand). It had specific gravity of 0.96 g/cc and melting temperatures of 127 °C.

**Composites Preparation:** The middle part of the jute/kenaf fibers were removed and chopped into 3 to 4 mm long. Before fabrication the jute and kenaf fibers were placed in air convection oven for 24 hours at 75°C. HDPE was mixed thoroughly with jute and kenaf fibers respectively. The procedure was repeated to reach the desired weight fraction. Then the mold was placed in automatic hot-press machine under 5 MPa at 190°C for 1 hour.

#### **Characterization**

**Fourier Transforms Infrared (FTIR) Spectroscopy:** The infrared spectra of the all specimens were recorded on a Shimadzu FT-IR 81001 Spectrophotometer. The spectra are described in the results and discussion section. The transmittance range of the scan was 4000 to 500cm<sup>-1</sup>.

**Tensile properties:** Tensile test was conducted according to ASTM D 638-01 using a Shimadzu Universal Testing Machine (MSC-5/500, Shimadzu Company Ltd, Japan) operating at a cross head speed of 5mm/min (Annual book of ASTM standard; 2002). The dimensions of the specimen were 115mm (L)  $\times$  6.5mm (W)  $\times$  3.1mm (T).

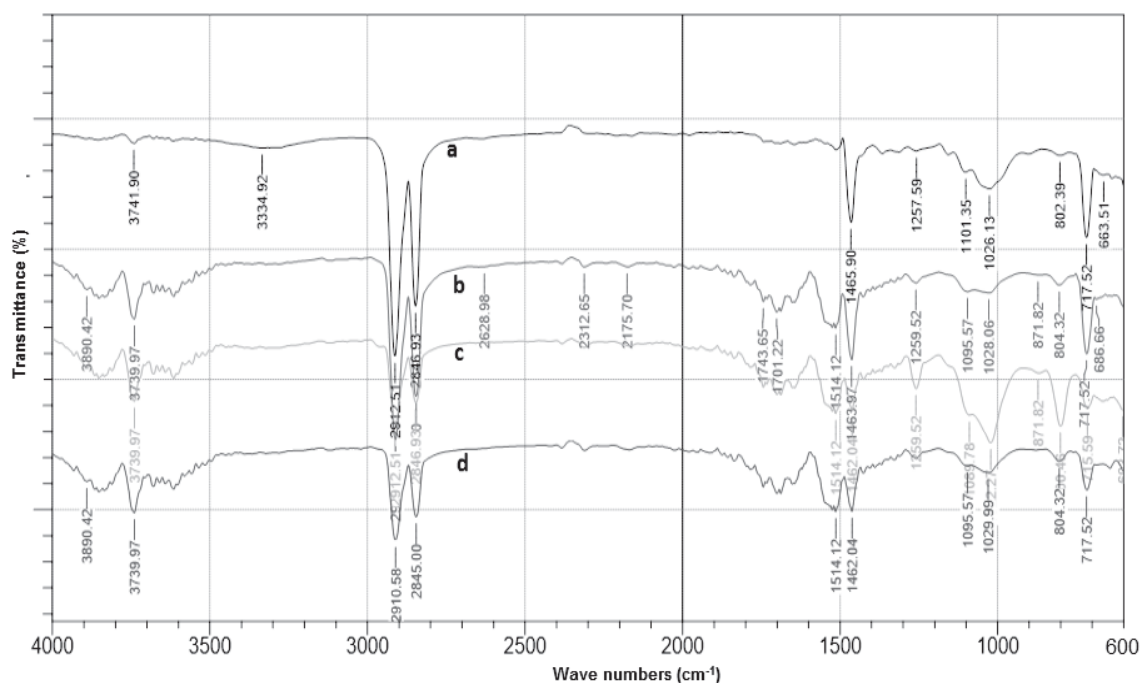
**Scanning electron microscopy (SEM) analysis:** The interfacial bonding between fibers and matrix was examined using a HITACHI (TM 3030) supplied by JEOL Company Ltd, Japan. The samples were sputter-coated with gold and observed under the SEM.

**Water absorption test:** The water absorption test was conducted as per ASTM D570-99. The test specimens were immersed in deionized water at 27°C for different periods (up to 22 days). The weight of the samples was determined initially. After certain time intervals, samples were taken out, wiped and dried, weighted immediately.

**Thermo gravimetric analysis (TGA) and Differential scanning calorimetry (DSC):** TGA and DSC were conducted with a TGA/DSC1 STAR System, Mettler Toledo thermal analyzer per ASTM E1131 and ASTM D3418 respectively. The thermal analysis was done in a nitrogen gas atmosphere with flow rate of 30 mL min<sup>-1</sup> at the heating rate of 10°C min<sup>-1</sup> from 50°C to 800°C.

## Results and Discussion:

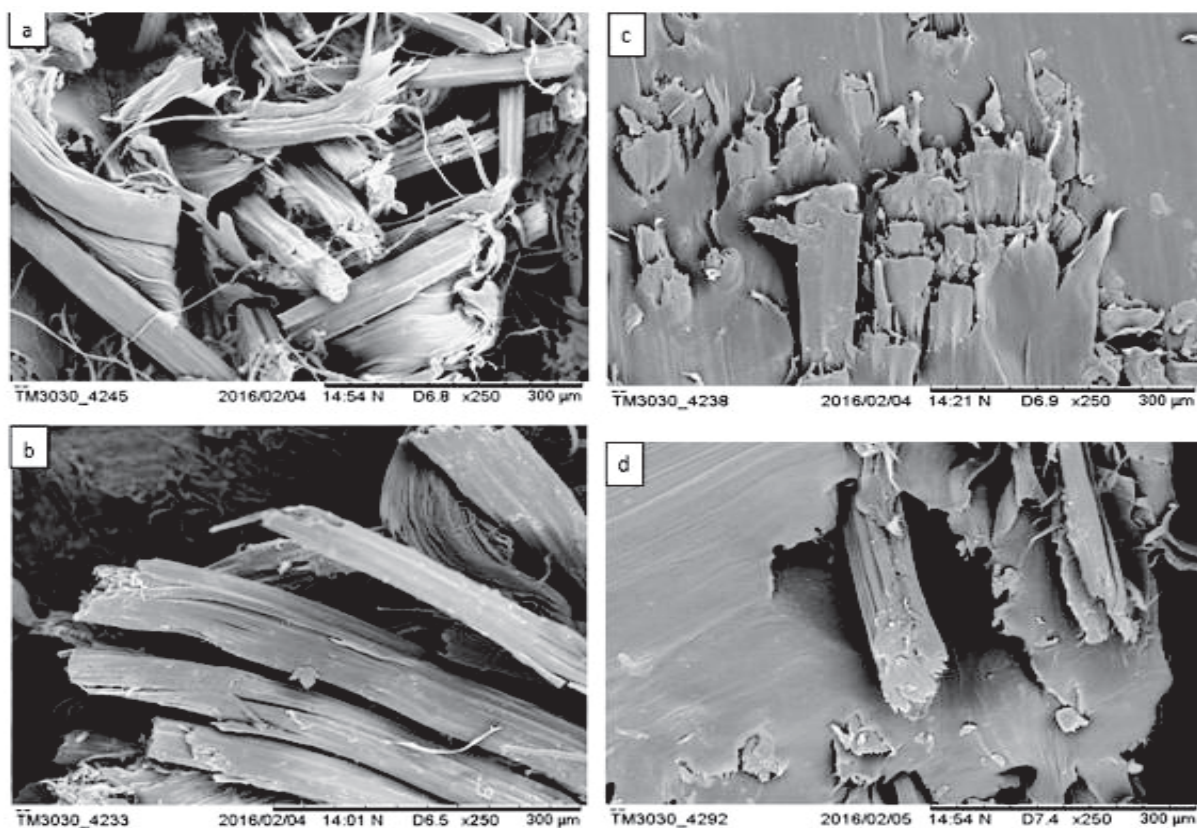
**Fourier transforms infrared (FTIR) spectroscopy:** The FTIR spectra analysis of jute/Polyethylene composite (JPEC) and jute/kenaf fiber/Polyethylene composite (JKPEC) at different fiber loadings (10, 15, and 20%) are shown in Figure 1.



**Fig. 1:** FTIR spectra of a: JPEC, b: JKPEC 10%, c: JKPEC 15%, and JKPEC 20%

An extensive absorption band in the region of  $4000$  to  $3300\text{cm}^{-1}$  due to stretching of H-bonds in O-H groups [13]. The band from  $1500$  to  $1450\text{cm}^{-1}$  is associated with the  $\text{CH}_2$  deformation vibration of an aromatic nature. The peak intensity shown at  $1259\text{cm}^{-1}$  was due to the C-O stretching vibration of the acetyl group in lignin and hemicelluloses [14]. A common absorption band from  $1100$  to  $1020\text{cm}^{-1}$ , specified as C-O deformation for primary alcohol in lignin [15]. The C-H stretching vibration of methyl and methylene groups in cellulose and hemicelluloses was observed from  $2900$  to  $2800\text{cm}^{-1}$ . The IR spectrum of JPEC showed peak intensity at  $3741$  to  $3334\text{cm}^{-1}$ , whereas JKPEC at different fiber (10, 15 and 20%) loadings peak intensities at  $3890$  to  $3739\text{cm}^{-1}$ . An adsorption band from  $1751$  to  $1700\text{cm}^{-1}$  are assigned to the C=O stretching of the carboxyl and acetyl groups in hemicelluloses of the hybrid fiber at different fiber loadings [16].

**Scanning Electron Microscopy (SEM) analysis:** SEM micrographs of the tensile fracture surfaces of the JPEC and JKPEC at different (10, 15 and 20%) fiber loadings are shown in fig 2(a-d). The figures indicate that there was considerable difference in the interfacial interaction between fiber and matrix in the composite system.



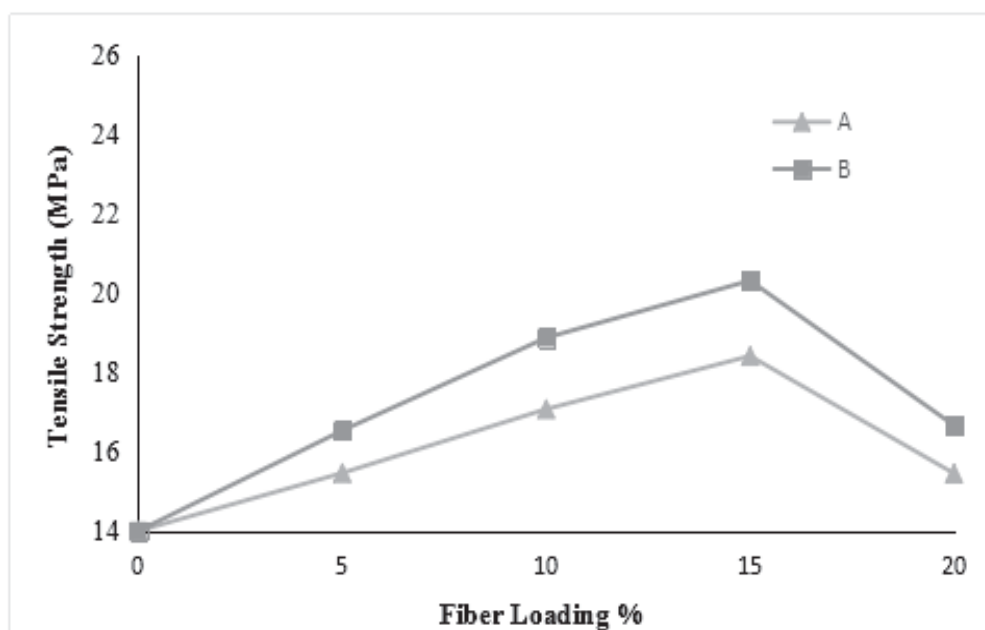
**Fig. 2:** SEM images of the fracture surfaces of a) JPEC 15% b) JKPEC 10%; c) JKPEC 15%; and d) JKPEC 20%



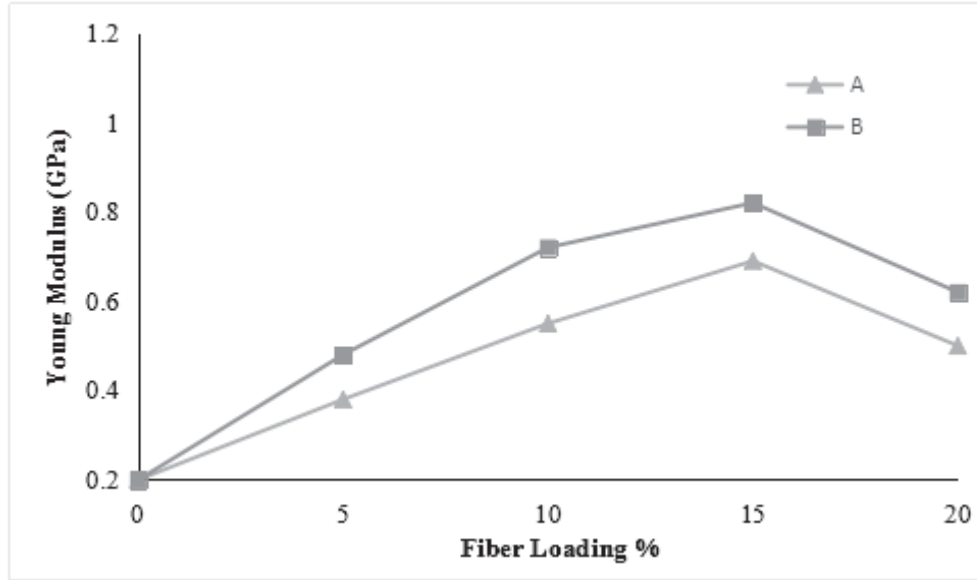
The SEM image of jute polyethylene composite showed the pullout traces of fibers with rough surfaces, micro-voids, and agglomeration at 15% fiber loading (Figure 2a). Poor interfacial bonding between the hydrophilic fiber and the hydrophobic matrix is main reason of this [16]. Figures 2 (b) and (d), showed relatively uneven surfaces and number of pull-out traces of fiber with void spaces as well as agglomeration [17].

Figure 2(c) shows better interfacial bonding between fiber and polymer matrix and less number of pull-out traces of fiber compared to Figures 2 (b) and 2 (d); [18]. At 15% fiber loading, JKPEC showed good compatibility between hydrophilic fiber and hydrophobic matrix. The effect of fiber loading on the morphology properties of composites is explained by the homogeneity of fiber and wettability of polyethylene matrix [19]. Good distribution between fiber and matrix among all the JKPEC composite at 15% loading showed smoother surface.

**Tensile properties:** The tensile strength and Young's modulus of JPEC, and JKPEC are shown in figures 3 and 4. The tensile strength and Young Modulus increase continuously up to 15% fiber loading for JKPEC and then decrease after further addition of fiber. The JKPEC at 15% fiber loading showed greater tensile properties than JPEC at 15% fiber loading. The homogeneity of fiber and wettability of polyethylene matrix has an important factor on mechanical properties of the composite system [20]. The JPEC and JKPEC composites exhibit a decrement after 15% fiber content because of fiber agglomeration [21]. Poor fiber distribution and low load transfer capacity is the main reasons that caused poor tensile properties at lower level of fiber content for both composite. The optimum fiber content varies with the fiber-matrix ratio and fiber-matrix interaction [22]. The hybridization of jute and kenaf fibers with polyethylene matrix increased the tensile strength and Young's modulus [23].

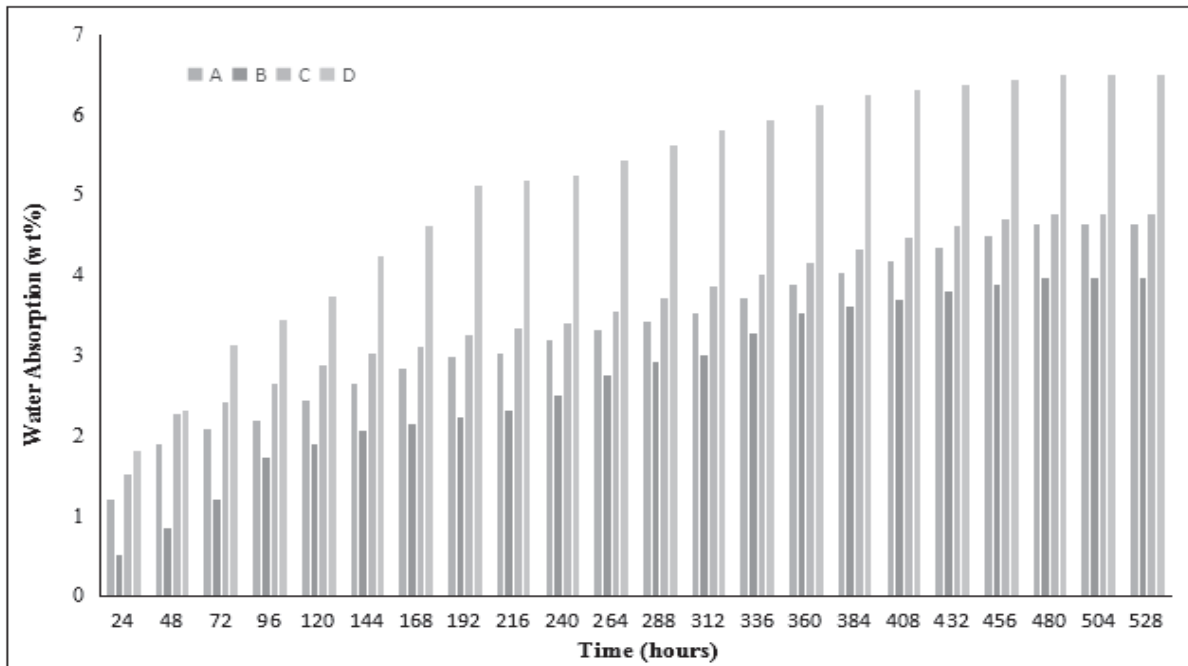


**Fig. 3:** Tensile strengths at different fiber loadings for a) JPEC, and b) JKPEC



**Fig. 4:** Young's Modulus at different fiber loadings for A) JPEC, and B) JKPEC

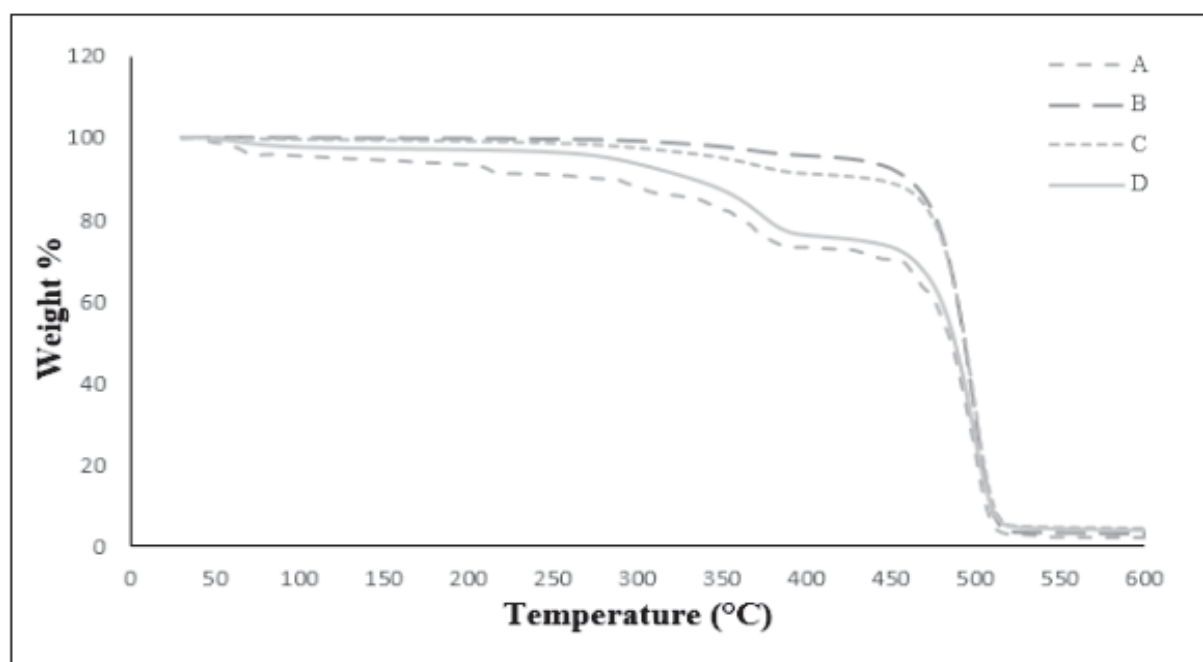
**Water absorption:** Water absorption behaviour of natural fibers depends on free hydroxyl groups present in cellulose and hemicelluloses. Other factors such as porosity, void content, lumen size, and fiber-matrix adhesion also affect water absorption behaviour of the composites [24].



**Fig. 5:** Water absorption behaviour for raw jute polyethylene and jute/kenaf polyethylene composites at different fiber loading where; A: JPEC (15%), B: JKPEC (10%) C: JKPEC (15%) and D: JKPEC (20%)

Figure 5 shows the water absorption percentage for jute polyethylene and jute /kenaf polyethylene composites in a different immersion time. During the experiment the composites absorbed water with different profiles. The composite continues to absorb water up to 528 hours (22 days), and then the values remained constant. After 528 hours (22 days) the lowest amount of water was absorbed by JKPEC (10%) followed by JPEC (15%), JKPEC (15%), and JKPEC (20%). The moisture absorption depends on void content in the composites and any increase in the void content lead to increasing water absorption [25]. In hybrid composite, arrangement of fiber retards the absorption of moisture into the composite, because the voids have been filled up during the formation of composite [26]. JKPE composite at 10% fiber loading showed lower percentages of water absorption. The higher loading of natural fibers is poorly dispersed within the composite system forming air bubble and agglomerations [27], which is the cause of higher water absorption.

**Thermo-gravimetric analysis (TGA):** The thermal properties of composites depend on various factors such as individual properties of the natural fibers that form the composite, the volume fractions of fibers, the micro-structural arrangement, and the interfacial bonding between the fibers and matrix. The thermal stability of JPEC and JKPEC at different fiber loading (10, 15 and 20%) are shown in Figure 6. It was observed that the thermal stability of JKPEC at 15% fiber loading significantly increases compared with JPEC, JKPEC (10%) and JKPEC (20%) respectively.



**Fig. 6:** TGA Curves for raw jute polyethylene and jute/kenaf polyethylene composites at different fiber loading where; A: JPEC (15%), B: JKPEC (15%), C: JKPEC (10%) and D: JKPEC (20%)

There are three stages of weight loss for the composites, the initial weight loss in the range 100–200 °C because of the evaporation of water contained in the natural fiber. The second stage of weight loss occurred at 200–380 °C, which was due to degradation of lignin and hemicellulose. The third stage of weight loss occurred above 380 °C which represent the cellulose degradation [28]. Hybrid composites at lower fiber loading showed improve thermal stability than JPEC [27]. The thermal stability of hybrid composites decrease with the fiber loading due to the lower thermal conductivity of individual fibers. Thus, JKPEC at 20% fiber loading showed weak thermal stability compared to JKPEC at (15%) fiber loading.

**Table 1.** Thermal Characteristics of JPEC (15%), and JKPEC (10, 15 and 20%)

Sample	T <sub>i</sub> (°C) <sup>a</sup>	T <sub>m</sub> (°C) <sup>b</sup>	T <sub>f</sub> (°C) <sup>c</sup>	W <sub>Ti</sub> (%) <sup>d</sup>	W <sub>Tm</sub> (%) <sup>e</sup>	W <sub>Tf</sub> (%) <sup>f</sup>	Activation Energy, E <sub>a</sub> (J/K)
JPEC	282	449	504	95.47	89.78	13.09	198.0039
JKPEC (10%)	312	462	508	95.81	89.95	13.25	204.4410
JKPEC (15%)	306	456	516	96.06	90.36	13.85	220.6951
JKPEC (20%)	298	450	510	95.76	90.19	13.40	211.1543

<sup>a</sup>Temperature corresponding to the beginning of decomposition.

<sup>b</sup>Temperature corresponding to the maximum rate of mass loss.

<sup>c</sup>Temperature corresponding to the end of decomposition.

<sup>d</sup>Mass loss of temperature corresponding to the beginning of decomposition.

<sup>e</sup>Mass loss of temperature corresponding to the maximum rate of mass loss.

<sup>f</sup>Mass loss of temperature corresponding to the end of decomposition.

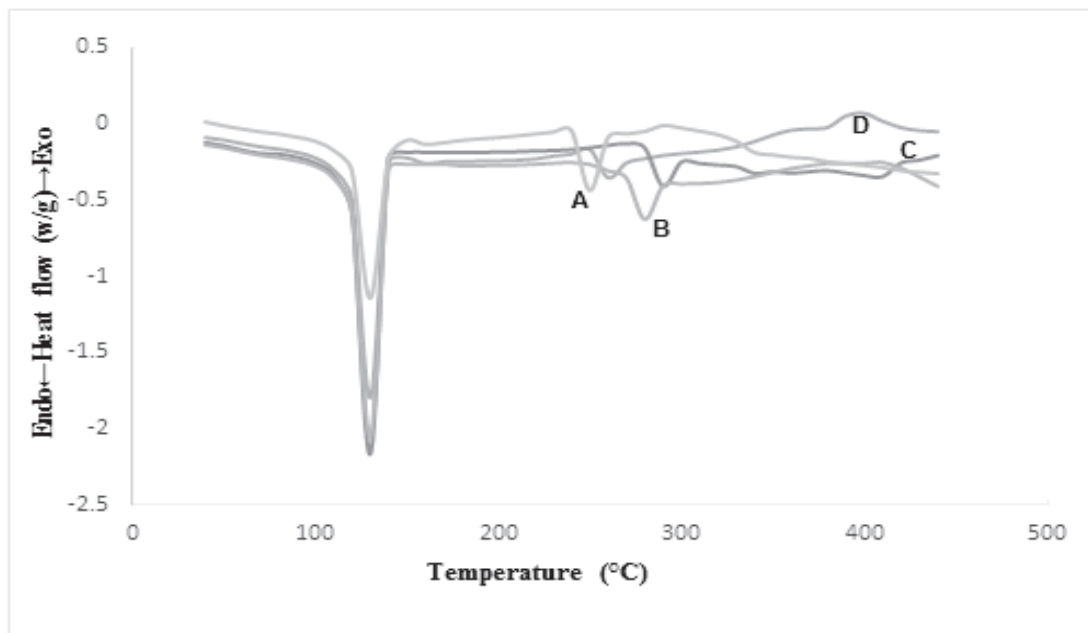
**Differential scanning calorimetric (DSC) Analysis:** Differential scanning calorimetric (DSC) analysis is carried out to determine the thermal energy released or absorbed via chemical reactions of the fiber constituents during heating. This lead to the exothermic and endothermic reactions. Endothermic reactions provide information on sample melting, phase transitions, evaporation, dehydration and pyrolysis. Exothermic reactions provide information on crystallization, oxidation, combustion, decomposition and chemical reactions [29]. The volume fraction of voids presents in the hybrid composite material is a very important factor which affects its thermal conductivity.

Figure 7 shows the DSC curves of the raw jute polyethylene composites JPEC (15%) and raw jute/kenaf fiber reinforced polyethylene (JKPEC) composites at different fiber loading (10, 15 and 20%). Firstly, the broad endothermic peak observed within 125–135 °C corresponds to the evaporation of absorbed water by the fibers. Lastly, the sharp endothermic peak obtained within 245–295 °C indicates the degradation of cellulose, leading to char formation [18]. Decomposition peaks of JKPEC (15%) composites exhibited higher value compare than other composites.

At 15% fiber loading, this randomness causes some fibers to be in contact with other fibers and hence a higher thermal resistance and a lower thermal conductivity for the hybrid composites are



attained [29]. The results obtained in the present study indicated that fiber loading have a great influence on the thermal properties. Heat capacity and thermal stability of the hybrid composites decrease with the fiber loading. This means that hybrid composites containing jute and kenaf fiber with polyethylene require longer time to be heated or cooled than the jute with polyethylene composite. The detailed thermal responses of the JPEC (15%) and JKPEC at different fiber loading (10, 15 and 20%) composites were summarized in Table 2.



**Fig. 7:** DSC Curves for different weight fiber loading where; A: JPEC (15%), B: JKPEC (10%), C: JKPEC (15%), and D: JKPEC (20%)

**Table 2.** Thermal Characteristics of DSC for JPEC (15%) and JKPEC (10, 15 and 20%)

Sample	Stage	DSC Analysis	
		$T_{\max}$ (°C)	Nature of Peak
JPEC (15%)	1 <sup>st</sup>	128.91	Endo
	2 <sup>nd</sup>	252.38	Endo
JKPEC (10%)	1 <sup>st</sup>	131.72	Endo
	2 <sup>nd</sup>	280.49	Endo
JKPEC (15%)	1 <sup>st</sup>	132.88	Endo
	2 <sup>nd</sup>	290.71	Endo
JKPEC (20%)	1 <sup>st</sup>	130.63	Endo
	2 <sup>nd</sup>	260.98	Endo

**Conclusion:** The SEM results showed differences among the composites because of the strong interfacial bonding between the jute/kenaf fiber and polyethylene. Hybrid composites yield better distribution and lesser void spaces showed better surface morphology than JPEC. Good

distribution between fiber and matrix among all the composite JKPEC at 15% fiber loading showed smoother surface and less agglomeration than another hybrid composites. After analyzing the tensile strength and Young's modulus of all the composites, it was observed that hybrid composite showed better properties than jute polyethylene composite (JPEC). The JKPEC at 15% fiber loading showed better tensile properties than another hybrid composite. Hybrid composites with increased fiber loading (20%) are poorly dispersed inside the composite system forming air bubble and agglomerations which reduces the mechanical properties of the composite. Between all the composites, JKPEC (10%) displayed less water absorption. Because of lower fiber loading than other composites JKPEC (10%) showed this characteristic. Higher fiber loading composites showed greater water absorption capacity. After observing the TGA and DSC results, it was clearly seen that hybrid composites exhibited higher thermal decompositions compared to jute polyethylene composite. Among all the composites, JKPE composite at 15% fiber loading displayed better thermal stability than other hybrid composites.

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