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# Comprehensive Characterization of Lignocellulosic Fruit Fibers Reinforced Hybrid Polyester Composites

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**Abstract:** This work is focused to fabricate and characterize hybrid polyester composites containing equal weight percentages of lignocellulosic fruit fibers namely *Cocos nucifera* and *Luffa cylindrica*. Samples with four different compositions (10%, 20%, 30%, and 40%) containing random fiber orientation are fabricated using handlayup technique and their effect on the flexural strength and impact strength is studied and compared with their individual counterparts. Enhanced mechanical properties are obtained when the combined weight percentage of both fibers is 30% by weight. This hybrid composite sample is characterized by Fourier Transform Infrared spectroscopy, X-ray diffraction and Thermogravimetric analysis. Surface morphology of the fractured sample with elemental analysis is studied using scanning electron microscope and Energy Dispersive Analysis of X-rays. FTIR peaks confirm the presence of biopolymers cellulose, hemicellulose and lignin. The thermogram confirms the presence of lignin, an amorphous hydrophobic biopolymer with strong intermolecular, intramolecular hydrogen bond and cross linking of the molecules requiring more energy to breakdown resulting in good thermal stability of the hybrid composites around 200°C. Elemental analysis gives O/C ratio of 0.45 corroborating the lignin presence at the surface.

**Keywords:** Hybrid Composites, Lignocellulosic Fibers, Characterization Studies, Mechanical Property, SEM with EDAX

## 1. Introduction

Abundant availability and accessibility of the natural fibers have stimulated increasing interest among the academicians and researchers to replace the synthetic fibers due to the growing environmental constraints and regulatory requirements such as recyclability, sustainability and renewability. [1-2] Appreciable mechanical properties, environmental friendly characteristics of the natural fibers, in addition to their light weight, non-toxicity, easier processing, easier handling, low cost, low density, high specific strength, and non-abrasiveness have made them potential candidates for reinforcement in the development of the composites. [3-4] The fibers derived from leaves (abaca, banana, pineapple) stem (jute, hemp, kenaf and ramie) fruit (coir, sponge guard and oil palm) seed (cotton), grasses (bamboo), roots (broom) and animal fibers (chicken feather, wool and silk) are used in the development of composites. [5-7] Though usage of natural fibers is accompanied with low strength, poor moisture resistance, dimensional instability and hydrophilicity, the performance of the composites are enhanced by reinforcing two or more fibers in a single matrix resulting in a hybrid composite. [8]

In this present work, hybrid composite samples are prepared using unsaturated polyester resin matrix reinforced by lignocellulosic fruit fibers namely *Cocos nucifera* L. (Coconut tree), a lignocellulosic fiber from Arecaceae family [9, 10] and *Luffa cylindrica* fiber, an annual herbaceous plant from the Cucurbitaceous family. [11, 12] The aim of this work is to study the effect of hybridization on the mechanical properties of the hybrid composites along with the characterization of the

samples that offer optimum mechanical properties using Fourier Transform Infrared spectroscopy, X-ray diffraction, Thermogravimetric analysis in addition to their morphological study and elemental analysis.

# 2. Materials and Methods

### *2.1. Extraction*

In this study, raw fibers of ripened *Cocos nucifera* and *Luffa cylindrica* fruits that are collected in the farms near Madurai, India are used for the preparation of the samples. The *Cocos nucifera* fibers are mechanically extracted from the coconut husk while the outer core of the *Luffa cylindrica* fruit is cut and separated from the inner core for the sample preparation. Initially, the fibers are washed with water to remove unwanted dirt and impurities present in them and then they are dried under sunlight to remove moisture.

### *2.2. Sample Preparation*

The hybrid composite samples are prepared using chopped *Cocos nucifera* and *Luffa cylindrica* fibers as reinforcement and unsaturated polyester resin as matrix along with methyl ethyl ketone peroxide (MEKP) as catalyst and cobalt napthenate as accelerator respectively. Samples are prepared using a wooden mould in four different weight proportions (10%, 20%, 30% and 40%) of reinforcement such that all the samples contain equal weight percentages of *Cocos nucifera* and *Luffa cylindrica* fibers. [8]

### *2.3. Fourier Transform Infra-red (FTIR) Spectroscopy*

FT-IR analysis of the manually powdered hybrid sample is recorded by SHIMADZU FTIR spectrometer in the spectrum range of 4000-400cm$^{-1}$ with an average of 32 scans and a resolution of 4cm$^{-1}$ at room temperature using the standard potassium bromide (KBr) pressed pellet technique for identifying the functional groups in the sample.

### *2.4. X-Ray Diffraction (XRD)*

X-Ray diffraction (XRD) is carried out to study the crystal structure of the hybrid composite sample. XRD patterns of the manually powdered hybrid sample is recorded with a X'Pert Pro-PANalytical system at room temperature in the 2θ ranging between 5°C and 50°C, with measurements made in continuous mode with a step size of 0.04 using CuKα radiation (λ=1.54060 Å) generated at a voltage of 40 kV and current of 30 mA.

### *2.5. Thermogravimetric Analysis (TGA)*

Shimadzu Thermal Analyzer is used to record the thermogram of the hybrid sample. The manually powdered sample is placed in a platinum crucible and heated using a continuous flow of nitrogen gas from room temperature to 800°C at a constant heating rate of 20°C / min. The furnace is purged using nitrogen by maintaining a constant flow rate of maintained at 30mL/min before analysis.

### *2.6. Mechanical Studies*

The flexural test is performed on the samples of size 125 x 12.5 x 3 mm according to ASTM D790 standards using a three point bending test at a crosshead speed of 2mm/min and the support span length being 50mm. [10] The Impact test is performed on the unnotched samples of size 65 x 15 x 6 mm according to ASTM D256 standards using an Izod impact machine of 25J capacity. The effect of fiber loading on the resin is studied and the weight percentage of fibers that give maximum strength is found. [10, 12] Three samples are tested for each combination and the average value is reported.

### *2.7. Scanning Electron Microscopy (SEM) and Energy Dispersive Analysis of X-Rays (EDAX)*

The morphology of the fractured surfaces of the hybrid samples are examined using a Scanning Electron Microscope (VEGA 3 TESCAN) which is operated at 20 kV. Prior to SEM analysis, the samples are sputter coated with gold to avoid charging. Energy Dispersive Spectrometer (EDS) analyzer (Bruker) is used for the elemental analysis of the composites. Microanalysis is performed at an accelerating voltage of 20.0 kV and at a working distance of 14.4 mm.

# 3. Results and Discussion

### *3.1. FT-IR Analysis*

The FT-IR spectrum of the composite sample in Figure 1 shows intense absorption in the range of 3600–3200 cm$^{-1}$, which is related to characteristic hydroxyl stretching vibration of free and hydrogen bonded –OH group specific to α-cellulose[13] and the bend at 3529cm$^{-1}$ shows the intramolecular hydrogen bond in the phenolic group in lignin.[14] The sharp edged peak at 2964cm$^{-1}$ and 2860cm$^{-1}$ is attributed respectively to the symmetrical and asymmetrical C–H stretching vibrations of alkyls confirming the presence of pectin[10]. The peak at 2096cm$^{-1}$ corresponds to the C=C stretching vibration of alkynes and the sharp intense peak at 1720 cm$^{-1}$ is ascribed to the C=O stretching vibration of acetyl and uronic ester groups related to hemicelluloses and the ester linkage of the carboxylic group corresponding to the ferulic and p-coumaric acids of lignin.[15] The peak at 1604cm$^{-1}$, 1581cm$^{-1}$ and 1502cm$^{-1}$ is attributed to the presence of C=C aromatic symmetrical stretching, aromatic stretching vibration and ring breathing with C=O stretching characteristic of lignin. [10, 12, 16] The peak at 1296cm$^{-1}$ corresponds to C-O and C-C stretching in the crystalline phase of cellulose. The presence of cellulose glycosidic COC symmetrical stretching and stretching of C–OH of hemicellulose is found at 1182cm$^{-1}$ and 1085 cm$^{-1}$ respectively [17]. The peak at 954 cm$^{-1}$ is concomitant with glycosidic C-C stretching and the peak at 896cm$^{-1}$ is associated with the C-O-C stretching at the β-(1-4) glucosidic linkages between sugar units in hemicellulose and cellulose II. The peak at 827cm$^{-1}$ is attributed to the C-OH out of phase bending, while the peak at 557 cm$^{-1}$ is due to the torsional

vibration of pyranose ring present in glucose. [18] Thus the presence of cellulose, hemicellulose and lignin are confirmed through FTIR.

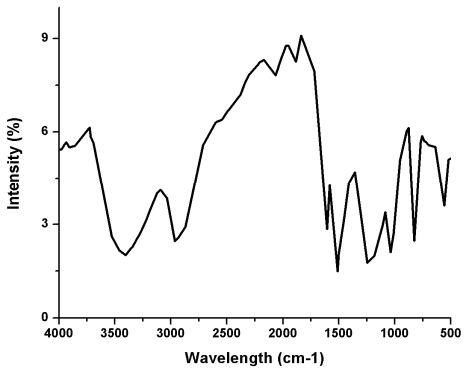


***Figure 1.*** *FT-IR spectra of the hybrid composite.*

### *3.2. XRD Analysis*

The XRD of the hybrid composite sample in Figure 2 does not show any sharp peak, the characteristic of a crystalline material. Hence it can be inferred that the sample lacks systematic and regular arrangement of atoms due to its amorphous nature. The 2θ peaks that correspond to the crystalline nature of native cellulose present in *Cocos nucifera* [10] and *Luffa cylindrica* [12] fibers are at 22.19° and 22.14° respectively as reported in previous researches. Similarly, the amorphous part of the sample showed peak intensities at 16.27° and 18.22° for *Cocos nucifera* [10] and *Luffa cylindrica* [12] fibers respectively. In the present study, there is a shift in 2θ value towards lower diffraction angle and a broad peak (also referred as halo) centered at 18.10° is observed due to the increase in d-spacing which shows that the sample is amorphous. [19] The amorphous nature of the sample is attributed to the presence of hemicellulose, lignin and disordered structure (amorphous content) of cellulose present in the fibers, the polyester resin used for sample preparation and also due to decrease in crystallite size which makes the diffraction peaks to broaden so that they merge into each other resulting in a single broad diffraction peak. This is further confirmed by the energy dispersive x-ray analysis which is reported later in this study.

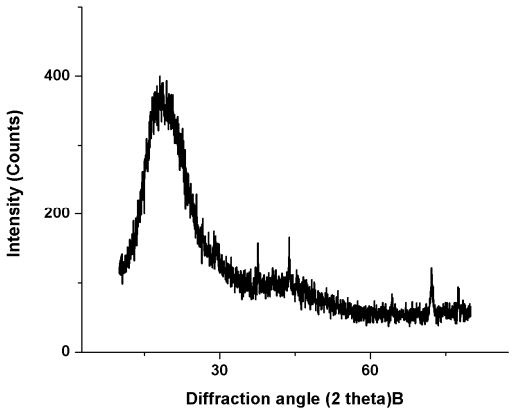


***Figure 2.*** *XRD of the hybrid sample.*

### *3.3. Thermogravimetric Analysis*

The thermal behaviour of the composite is studied by the thermogravimetric analysis which is generally expressed as a function of mass loss with respect to temperature. [20] As shown in Figure 3, in the first stage of decomposition there is a mass loss of about 10% in the temperature ranging between 29°C and 205°C. The mass loss in this stage is mainly attributed to the evaporation of moisture content evinced by the presence of hydroxyl content in the fibers and other volatile matter present in the composite as reported by other researchers. [20, 21] The second stage decomposition takes place in the temperature between 205°C and 457°C with a mass loss of 57.25% and is attributed to the thermo oxidative degradation of hemicellulose that takes place by the degradation of its subunits arabinogalactan and galactoglucomannan at a temperature between 220°C and 315°C followed by the thermal degradation of cellulose in the temperature ranging between 290°C and 360°C by major reactions namely dehydration, thermoxidation, transglycosylation, depolymerization with volatilization of levoglucosans at temperature above 300°C.[22,23] Finally, the third stage degradation takes place in the temperature range 457°-627°C during which lignin degrades by the breakage of its sub units Guaiacylpropane, syringylpropane and p-hydroxyphenylpropane that are held by strong bonds. [24]

The thermogram of *Cocos nucifera* fibers [10] shows stage wise degradation process in the temperature ranges 28°C–148°C, 255°C-306°C, 309°C–431°C while *Luffa cylindrica* fibers[12] exhibits degradation in the temperature ranges 49°C–118°C, 118°C–322°C and 322°C–455°C. In the present study, the degradation stages of the hybrid composites are in the temperature range 29°C-205°C, 205°C-457°C and 457°C-627°C. Comparing this with themogravimetric analysis of the *Cocos nucifera* and *Luffa cylindrica* fibers, higher temperature is necessary for the hybrid composites to undergo degradation in all the three stages which is due to the presence of strong intermolecular and intramolecular hydrogen bond and cross linking of the molecules requiring more energy to breakdown. The initial decomposition of the hybrid composites start at around 205°C with only 10 % loss which clearly shows a good thermal stability of these composites up to 200°C.

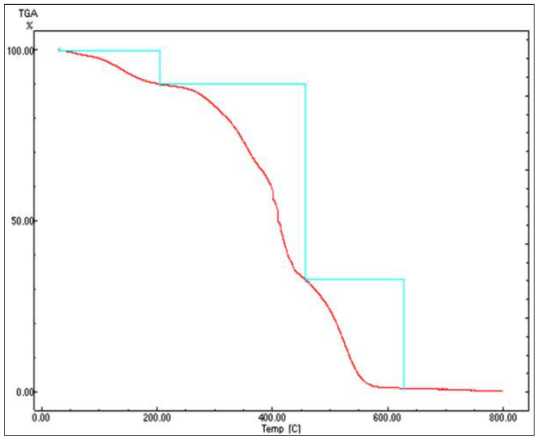


***Figure 3.*** *Thermogram of the hybrid sample.*

### *3.4. Mechanical Studies*

In the present study, the flexural strength of the hybrid composite obtained is 18.2, 20.4, 21.6 and 21 MPa for 10%, 20%, 30%, 40% fiber loading respectively. This shows that the inclusion of fibers in the polymer matrix increases the mechanical properties of the composites. It is due to the effective stress transfer that takes place between the matrix and the fibers. The increase in fiber loading causes the composite to be stiffer and stronger and the maximum flexural strength is obtained for 30% fiber loading beyond which there is a decrease in flexural strength agreeing with the previously published results. [10, 25, 26] The decrease in flexural strength at higher fiber loadings is due to improper wettability and poor dispersion of the fibers in the matrix.

Similarly, the impact strength of the hybrid composites too show increasing trend for increased fiber loading till 30% fiber loading [12, 27] and the values are 6.40kJ/m$^2$, 10.27 kJ/m$^2$, 11.80 kJ/m$^2$ and 11.33 kJ/m$^2$ for 10%, 20%, 30%, 40 % fiber loading respectively. Impact strength of the composite is related to toughness and it is influenced by the interfacial bond strength, matrix and fiber properties. In this study, the increase in impact strength of the hybrid composites is due to the synergistic stress transfer effect of *Cocos nucifera* and *Luffa cylindrica* fibers on the interlocking bond. The high loading bearing capacity of the lignocellulosic fruit fibers are capable of absorbing high impact energy during impact agreeing with previous results. [12] The mechanical properties are shown in Table 1.

***Table 1.*** *Mechanical Properties of the hybrid composite.*

| S. No | Weight proportion of the samples in % | | | Flexural Strength | Impact Strength |
|---|---|---|---|---|---|
| | Polyester Resin | *Cocos nucifera* fibers | *Luffa cylindrica* fibers | MPa | kJ/m$^2$ |
| 1 | 90 | 5 | 5 | 18.2 | 6.4 |
| 2 | 80 | 10 | 10 | 20.4 | 10.27 |
| 3 | 70 | 15 | 15 | 21.6 | 11.8 |
| 4 | 60 | 20 | 20 | 21 | 11.33 |

### *3.5. SEM and EDAX*

SEM analysis of the fractured sample is done to study the morphology and the dispersion of the natural fibers in the fibers. As shown in Figure 4, the fibers are randomly oriented and an improved interfacial adhesion between fiber and matrix is achieved resulting in effective stress transfer and also there is no fiber pullout. Failure of the composite samples takes place during mechanical testing due to the weak interfacial bonding at the fiber matrix interface that could not transfer stress resulting in debonding (shown using arrows in the Figure 4). The presence of many voids (shown in Figure 4) is another reason for the failure of the composite.

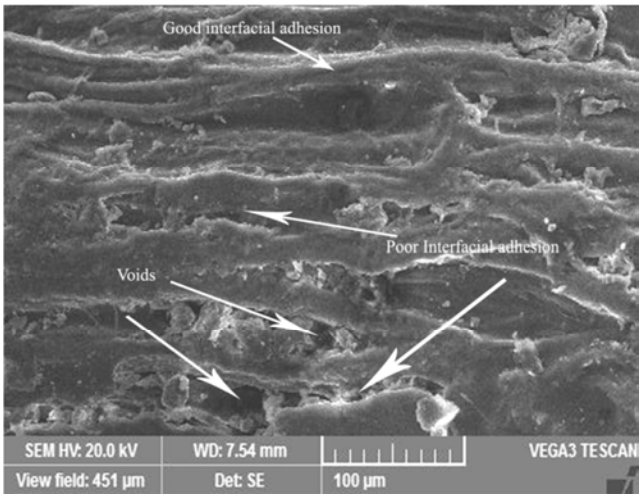


***Figure 4.*** *SEM image of fractured sample.*

The elemental analysis of the fractured sample as shown in Figure 5, using EDAX shows the presence of C (68.12%), O (30.44%) along with minor traces of K (0.78%), Ca (0.36%), and Si (0.3%) agreeing with previous research.[11] The high content of Carbon and Oxygen in the surface is due to the lignocellulosic fibers that essentially consist of biopolymers cellulose, hemicellulose, and lignin and the traces of minerals potassium, Calcium and Silicon are the macronutrients that the plant has obtained from the soil through their roots. An O/C ratio of 0.83 shows the presence of cellulose, hemicellulose and pectin and an O/C ratio of 0.31 to 0.40 shows the presence of lignin as reported in the literature. [29, 30] The O/C ratio of 0.45 confirms the presence of high proportion of lignin at the surface.

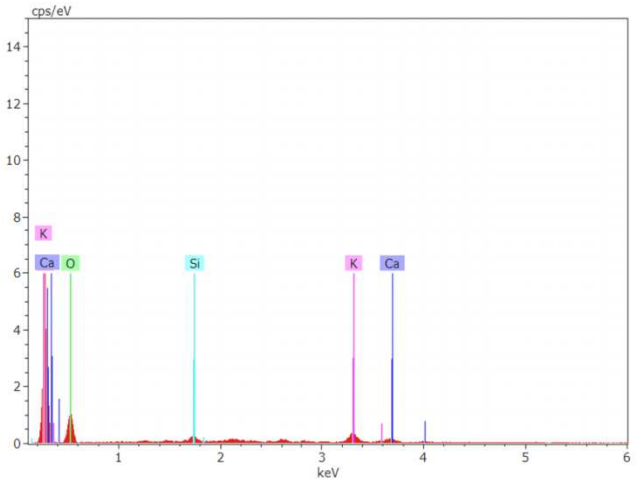


***Figure 5.*** *EDAX of the hybrid sample.*

## 4. Conclusions

In this work, the hybrid samples are prepared using equal weight percentages of the *Cocos nucifera* and *Luffa cylindrica* fibers and it is found that enhanced mechanical

properties are obtained for samples that contain 30% fiber loading. The FTIR study confirms the presence of cellulose, hemicellulose and lignin. The amorphous nature of the sample is ascertained by the broad peak obtained in X-ray diffraction studies. TGA confirms the sample is thermally stable till 200°C on account of the high complex structure of lignin. The O/C ratio too confirms the presence of lignin at the surface of the sample. From the study, it can be concluded that the lignocellulosic fruit fibers can be effectively used as promising reinforcements for making hybrid polymer matrices composites that can suit a variety of applications.

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## References

[1] Wambua, Paul, Jan Ivens, and Ignaas Verpoest. (2003). Natural fibres: can they replace glass in fibre reinforced plastics?. *Compos*. *Sci*. *Tech*. 63 (9): 1259-1264.

[2] Thakur, V. K., M. K. Thakur, and R. K. Gupta. (2014). Review: raw natural fiber–based polymer composites. *Int. J. Polym. Anal. Charact* 19 (3): 256-271.

[3] Jawaid, M., and H. P. S. Abdul Khalil. (2011). Cellulosic /synthetic fiber reinforced polymer hybrid composites: A review. *Carbohydr. Polym.* 86 (1): 1–18.

[4] Ho, Mei-po, Hao Wang, Joong-Hee Lee, Chun-kit Ho, Kin-tak Lau, Jinsong Leng, and David Hui. (2012). Critical factors on manufacturing processes of natural fibre composites. *Compos Part B: Eng* 43 (8): 3549-3562.

[5] Saheb, D. Nabi, and J. P. Jog. "Natural fiber polymer composites: a review." *Adv. Polym. Tech*. 18, no. 4 (1999): 351-363.

[6] Thakur, Vijay Kumar, and Manju Kumari Thakur. (2014) "Processing and characterization of natural cellulose fibers/thermoset polymer composites." *Carbohydr. Polym.* 109: 102-117.

[7] NagarajaGanesh, B., and Rekha, B., (2013) "A Comparative Study on Tensile Behaviour of Plant and Animal Fiber Reinforced Composites," *International Journal of Innovation and Applied Studies*, vol. 2, no. 4, pp. 645–648.

[8] Hussain Barbhuiya, A., and Ismail. K., (2016): Characterization of Hybrid Epoxy Composites Reinforced by Murta and Jute Fibers, *Int. J. Polym. Anal. Charact*, DOI: 10.1080/1023666X.2016.1183276.

[9] Harish, S., D. Peter Michael, A. Bensely, D. Mohan Lal, and A. Rajadurai. (2009). Mechanical property evaluation of natural fiber coir composite. *Mater. Character.* 60 (1): 44-49.

[10] NagarajaGanesh, B., and Muralikannan, R., (2016): "Physico-chemical, thermal, and flexural characterization of *Cocos nucifera* fibers." *Int. J. Polym. Anal. Charact*, 21, no. 3: 244-250.

[11] Valcineide O. A. Tanobea, Thais H. D. Sydenstrickera, Marilda Munarob, Sandro C. Amicoa. (2005) A comprehensive characterization of chemically treated Brazilian sponge-gourds (*Luffa cylindrica*) *Polym. Test*. 24 (4): 474–482.

[12] NagarajaGanesh, B., and R. Muralikannan, R., (2016): "Extraction and characterization of lignocellulosic fibers from *Luffa cylindrica* fruit", *Int. J. Polym. Anal. Charact*, 21, no. 3: 259-266.

[13] Karbowiak, T., Ferret, E., Debeaufort, F., Voilley, A., Cayot, P., (2011) Investigation of water transfer across thin layer biopolymer films by infrared spectroscopy. *J. Membr. Sci.* 370 (1): 82-90.

[14] Lasagabaster, Aurora, María José Abad, Luis Barral, and Ana Ares. (2006) FTIR study on the nature of water sorbed in polypropylene (PP)/ethylene alcohol vinyl (EVOH) films. *Eur. Polym. J.* 42 (11): 3121-3132.

[15] Alemdar, Ayse, and Mohini Sain. (2008) Isolation and characterization of nanofibers from agricultural residues–Wheat straw and soy hulls. *Bioresour. Technol.* 99 (6): 1664-1671.

[16] Lionetto, Francesca, Roberta Del Sole, Donato Cannoletta, Giuseppe Vasapollo, and Alfonso Maffezzoli.(2012). Monitoring wood degradation during weathering by cellulose crystallinity. *Materials* 5 (10): 1910-1922.

[17] Kim, Jun Tae, and Anil N. Netravali. "Mercerization of sisal fibers (2010): Effect of tension on mechanical properties of sisal fiber and fiber-reinforced composites." *Composites Part A.,* 41 (9): 1245-1252.

[18] Ray, Dipa, and B. K. Sarkar (2001) "Characterization of alkali‐treated jute fibers for physical and mechanical properties." *J. Appl. Polym. Sci.* 80, no. 7: 1013-1020.

[19] Priya Dasan, K., "Nanoclay/Polymer Composites: (2015) Recent Developments and Future Prospects." In Eco-friendly Polymer Nanocomposites, pp. 561-579. Springer India.

[20] White, John E., W. James Catallo, and Benjamin L. Legendre. (2011). Biomass pyrolysis kinetics: a comparative critical review with relevant agricultural residue case studies. *J. Anal. Appl. Pyrolysis* 91 (1): 1-33.

[21] Nair KM, Thomas S and Groeninckx G. (2001) Thermal and dynamic mechanical analysis of polystyrene composites reinforced with short sisal fibres. *Compos. Sci. Tech.* 61 (16): 2519–2529.

[22] Monteiro, Sergio Neves, Verônica Calado, Rubén JS Rodriguez, and Frederico M. Margem. (2012) Thermogravimetric stability of polymer composites reinforced with less common lignocellulosic fibers–an Overview. *J. Mater. Res. Technol.* 1 (2): 117-126.

[23] Kim, Hee-Soo, Sumin Kim, Hyun-Joong Kim, and Han-Seung Yang.(2006). Thermal properties of bio-flour-filled polyolefin composites with different compatibilizing agent type and content. *Thermochimica Acta* 451 (1): 181-188.

[24] Ishak. H., Sapuan S, Leman, Z., Rahman, H. and Anwar, U. (2012) Characterization of sugarpalm (Arenga pinnata) fibers. *J. Therm. Anal. Calorim.* 109 (2): 981-989.

[25] Atiqah, A., M. A. Maleque, Mohammad Jawaid, and Mohammad Iqbal (2014)."Development of kenaf-glass reinforced unsaturated polyester hybrid composite for structural applications." *Composites Part B:* 56: 68-73.

[26] Mohanty S, Verma SK, Nayak SK. (2006). Dynamic mechanical and thermal properties of MAPE treated jute/HDPE composites. *Compos*. *Sci*. *Technol*. 66 (3): 538-47.

[27] Ramanaiah, K., Ratna Prasad, A. V., and Hema Chandra Reddy, K., (2013). Mechanical and Thermo-Physical Properties of Fish Tail Palm Tree Natural Fiber–Reinforced Polyester Composites. *Int. J. Polym. Anal. Charact.* 18 (2): 126-136**.**

[28] Mohanta, N., and S. K. Acharya. (2015), "Fiber surface treatment: Its effect on structural, thermal, and mechanical properties of Luffa cylindrica fiber and its composite." *J. Compos. Mater.*: 0021998315615654.

[29] Sgriccia N, Hawley MC, Misra M. (2008) Characterization of natural fiber surfaces and natural fiber composites. *Composites Part A*. 39 (10): 1632-37.

[30] Toth A, Faix O, Rachor G, Bertoti I, Szekely T. (1993), ESCA (XPS) study on light-induced yellowing of thermomechanical and chemothermomechanical pulps. *Appl. Surf. Sci.*; 72 (3): 209-13.