Eng. Sci., 2022, **20**, 80–99

Engineered Science

DOI: https://dx.doi.org/10.30919/es8d713

A Review on Processing Methods and Characterization Techniques of Green Composites

Nithesh Naik,[1] B. Shivamurthy,[1,*] B. H. S.Thimmappa,[2] Aman Gupta,[1] John Zhanhu Guo[3,*] and Ilwoo Seok[4,*]

Abstract

In today's modern world, extensive research has been performed to apply new materials to various components. With stiff competition in the market, industrialists are keen to use tailor-made materials to provide a cutting edge to their product. Out of all the materials available worldwide, composites can be tailor-made to satisfy one's needs. Therefore, compounds have played a pivotal part in the revolution of the material industry. Composites have high durability, ratio of power to mass, and high efficiency. These advantages have made composites as a core material for the aerospace, automobile, and defense sectors. In bio-composites, carbon fiber composites and glass fiber composites have been used to reduce cost and energy consumption. Natural fibers traditionally used as fillers have now been extensively used as reinforcements in bio-composites. The adoption of green technology aids in overcoming environmental pollution challenges. It can achieve sustainable development by substituting petroleum-based composites with bio-based composites. This review discusses the various commonly used matrices and biopolymers in Green Composites. The article highlights characterization techniques and suitable processing techniques that are used to manufacture green composites.

Keywords: Natural Fibers; Processing methods; Characterization techniques; Green composites.
Received: 21 January 2022; Revised: 18 May 2022; Accepted: 21 May 2022.
Article type: Review article.

1. Introduction

Advanced polymer composites comprising glass and carbon fibers are used extensively in aerospace companies, construction, and automotive. Since the fiber and the matrices reinforcements in these advanced compounds are derived from mineral properties, their sustainability in the long term is a concern. These advanced composites are challenging to recycle due to their inherent morphology. Green composites are environmentally friendly and may be landfilled or destroyed at the end of their use cycle as they are biodegradable.[1] Natural fibers have some benefits over synthetic fibers such as high impact strength, low intensity, biodegradability, reduced respiratory and thermal irritation, less expensive, high resilience, high specific strength, and less usage on non-renewable resources. Usually, polyolefin thermoplastics like $(C_2H_4)n$ and $(C_3H_6)n$ were used as the matrix polymer. But, the combination of bio-fibers with bio-based polymers provides added environmental benefits.

Nevertheless, bio-based composites' usage compared with synthetic composites and isotropic structural materials has been restricted because of their lower thermo-mechanical and physical properties.[2,3] Various authors worldwide have done studies on bio-fiber-reinforced composites. Different natural fibers such as the Kapok, ramie, sisal, abaca, jute, hemp, and bamboo are mainly used due to their structural property for reinforcement. However, many positive remarks, a certain level of negative aspect, are also found in the natural fiber simultaneously. It must be ignored in the case of replacing natural fiber in the place of glass fibers. Moisture absorption level, absence of a link between matrix and fiber were the main reasons for poor affinity with the hydrophobic matrix[4]. It must be sorted out to attain uniform property based on the climatic condition for cultivation, thermal stability, *etc*. These are the certain drawbacks of the bio-fibers, making them achieve a low level of attention by the manufacturer for

[1] *Department of Mechanical & Manufacturing Engineering, Manipal Institute of Technology, Manipal Academy of Higher Education, Manipal-576104, Karnataka, India.*
[2] *Department of Chemistry, Manipal Institute of Technology, Manipal Academy of Higher Education, Manipal-576104, Karnataka, India.*
[3] *Department of Chemical and Biomolecular Engineering, University of Tennessee, Knoxville, TN 37996, USA.*
[4] *College of Engineering and Computer Science, Arkansas State University, Jonesboro, AR 72401, USA.*
**Email:* shiva.b@manipal.edu (B. Shivamurthy); zguo10@utk.edu (Z. Guo); iseok@astate.edu (I. Seok)

manufacturing purposes.[5] Peponi *et al.*[6] state the poor consistency of the natural fiber by pointing out small plants. The required fiber properties are high modulus, tensile strength, lesser bulk density, high durability, and recycling property.[7] With the help of renewable polymer, several bio-composite has been manufacturedfille.[8] And the things of these bio-compounds are characterized depending on the fiber property.[9]

1.1 Matrices for green composites

Holding the emphasizing things of composites by surface linking is the purpose of the matrix. Tolerances from the material reliability, environmental, and surface appearance are the critical functions of the matrix. When the matrix was under stress, it passes the load to the fibers evenly to prevent any development or spreading of cracks and damage.[10] The study has been undertaken to replace the fuel-based conventional matrices because of the inadequate fossil fuel sources and their environmental impacts.[11] The classification of polymer matrices based on their degradability used in green composites is shown in Fig. 1.

1.2 Petrochemical based resins

Such products are petroleum-derived products from fossil fuels like gas and coal.[11] Petrochemical matrices are classified as thermoplastics and thermosets when used for green composites. The thermoplastic matrices are polypropylene, polyethylene, polyvinyl chloride (PVC), and polystyrene, while epoxy, polyester, vinyl ester, and phenolic are analyzed thermoset conditions.

1.2.1 Thermoplastics

Polymers such as thermoplastic can be quickly modified and shaped when heated and are solid at room temperature. Better impact resistance, greater reformability, higher tolerance to damage, and higher process temperatures and pressure than thermoset resins are found in thermoplastic resins.[10] When the thermoplastic resin is melted, the viscosity is about 500-1000 times larger than the thermoset resins. The advantages and disadvantages of thermoplastic resins can be shown in Table 1.

1.2.2 Thermoset resins

The thermoset resins are infusible, insoluble, heat, or catalyst cured materials.[12] It is impossible to shape or reform these resins.[13] The Thermoset resins have a higher modulus, better resistance to creep, enhanced thermal stability, and increased resistance to chemicals than thermoplastic resins because of the 3D covalent connections in the polymer chains.[11] Table 2 displays the petrochemical resins' disadvantages and advantages.[14,15]

1.2.3 Biodegradable based resins

Polymers like biodegradable depend resins are derived partially or entirely from renewable sources. Production of bio-based polymers is proceeded by the plants (*i.e.*, cellulose and starch). Polymerization with plant sugar and oils such as polyethylene terephthalate, polylactic acid, and polypropylene) is also used. They can be classified into fully biobased and biodegradable, partially biological and biodegradable, biobased, and non-biodegradable based on their physical properties.[10,11] The disadvantages and the advantages of the types of biobased resins are shown in Table 3.

Table 1. Disadvantages and advantages of petrochemical thermoplastic resins.[12]

Resin	Advantages	Disadvantages
$(C_2H_4)n$	High impact strength and ductility Good exhaustion Light in weight Low immersion of moisture Inexpensive High confrontation to temperatures	Poor weather resistance Combustible High thermal expansion Difficulty in processing Costly
$(C_3H_6)n$	High dielectric resistance Excellent opposition to chemicals Good exhaustion Good resistance to chemicals Resistance to stress cracking	Combustible Low confrontation to impact
$(C_8H_8)n$	Low humidity absorption Weather High resistance to impact Versatile	Hard in nature Poor UV resistance Insufficient confrontation at high and low temps
$(C_2H_3Cl)n$	Good resistance to chemicals Fire retardant Inexpensive Good dimensional strength	

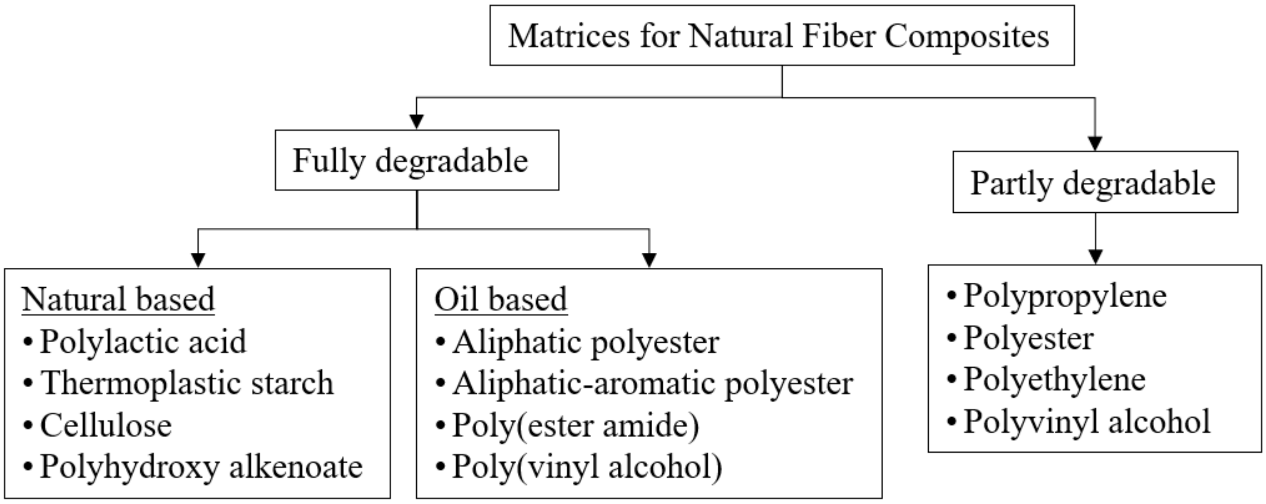


Fig. 1 Polymer matrix classification process utilized in Green Composite. Reproduced with the permission from [10], Copyright CRC Press 2019.

Table 2. Disadvantages and advantages petrochemical of thermoset resins.[14,15]

Resin	Advantages	Disadvantages
Epoxy	High mechanical and thermal properties High resistance to water Low shrinkage during curing Long working time ability	Expensive compared to vinyl ester Corrosive amine hardener Difficulty in processing
Polyester	It can be used easily Inexpensive	High shrinkage during curing The working times range is limited Adequate mechanical components High discharges of styrene in open mold
Vinyl ester	Excellent environmental and chemical composition. Advanced mechanical elements compared to polyester	Reduction during curing Expensive compared polyester A high content of styrene Post-remedial requirement for things.
Phenolic	High opposition to fire	Difficulty in processing

Table 3. Advantages and disadvantages of Bio-based Resins.

Resin	Advantages	Disadvantages
Starch	Completely environmental Inexpensive	Hard in nature Difficulty in processing Sensitive to water
PLA	High strength and modulus Relatively inexpensive Non-toxic	Brittle in nature Low thermal deprivation temperature Comparatively poor strength
PHA	Molecular mass is high Completely environmental	Relative low decomposition temperature Less stable Hard in nature Low deformability Costly compared to other bio-based polymers
Cellulose	Expensive Relatively inexpensive It can be modified easily Moderate impact resistance Moderate resistance to heat	Humidity absorption Relatively low decomposition temperature

Biodegradable polymers and fully bio-based and have renewable that, when the polymer decomposes, is removed from the environment and then added to the atmosphere. The polymers commonly used for not reusable cutlery or reusable food packet, with plastics disposed of with the excess food, can acquire this renewable carbon. This resin has several benefits over petrochemical depend on resins: less energy is required while in production (65% less energy used for production), are safe and recyclable (require less energy with fast break down), renewable, and eco-friendly (68% fewer greenhouse gases are generated).[16] However, the cost of production of petrochemical resins is approximately 10% higher.[17]

2. Matrix modification

There is a big concern in moisture absorption in the usage of natural fibers when biological resin matrices such as soy proteins are used. R. Kumar & Zhang improved this problem by introducing the various superplasticizers of benzylic acid.[18,19] The soy resin with 61% of superplasticizer thiodiglycol drops to 76% for resins' humidity absorption under the immersion in 0.5% benzyl acid during 27 hours. When thiodiglycol is used as a superplasticizer, nanospheres of widths 250–600 nm are detected, as shown in Fig. 2. These nanospheres are suspected to rise in tensile strength and other mechanical properties.[20]

Newill *et al*. demonstrated that polymeric coatings might reduce the humidity immersion in polymer and polymer compound structures.[20] It shows that the saturation level, thickness, and diffusion constant of the covering relative to the substrate are essential to the coating's effectiveness. A polymer coating of 0.254 mm thickness reduced the moisture absorbance of the matrix by about 47%, as shown in Fig. 3.

Doherty *et al*. proposed a framework using water-based lignin coating on the corrugated and cardboard sheets.[21] Through which the wettability of the layer can be determined using water absorption test and contact angle. From the contact angle increment, in the lignin films, it is found that the lignin is having adequate water barring capacity. The lignin films have decreased using water absorption levels for about 2-folds than the wax-based coating concerning the wetting characters. The thick layer of lignin coating absorbed only less amount of water. This study clearly shows that the natural lignin-based coating with 0.204 millimeters of thickness has reduced the matrix's moisture absorption by 62 percent.

3. Processing techniques

The choice of manufacturing methods is usually dependent on the form and ways in which the material is treated. The quality of the part, the level of complexity in the part's design, processing efficiency, and the criteria for capital costs are factors.[22] Also, moisture, type of fiber, fiber volume fraction, and material temperature are the factors for natural fiber composite manufacturing. Other essential impacts on the production, durability, and performance of composite materials are the aspect ratio, length, and chemical composition of fibers.[10]

Improvement of the fiber composite's volume fraction increases the composite's stiffness, water intake, and material strength and reduces its deformation.[10] Often the processing temperature is an important aspect. There is a maximum temperature limit. Temperatures beyond this level might cause many problems: the deprivation and reduction of the fiber, small-sized composites, recrystallization, decarboxylation, depolymerization, hydrolysis, dehydration, and oxidation.[23] Table 4 displays the disadvantages and advantages of various dispensation methods used to manufacture natural fiber compounds.

3.1 Compression molding (CM)

Preheated materials are put in the molding cavity initially in this technique. These are then compressed, deformed, and the cavity is exposed to high pressure by the mold on the core side. High pressure is retained to solidify the composite before the mold was opened to eliminate the composite. The critical limitations to be accounted for in this method are material quantity, heating period, mold pressure applied, and cooling time. Reddy and Yang used water as a plasticizer to produce

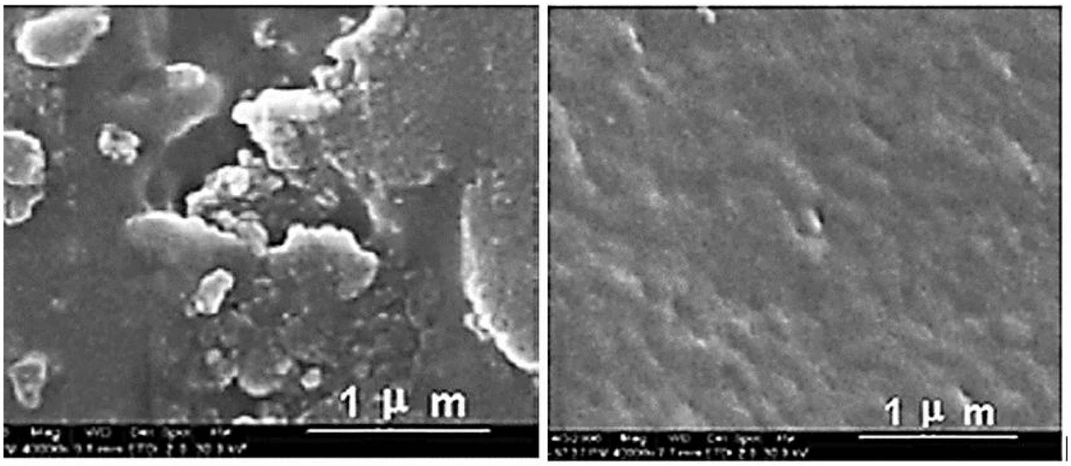


Fig. 2 Scanning electron microscope photographs of exterior (left) and cross-section (right) of Soy protein with thiodiglycol as superplasticizer. Reproduced with the permission from [19], Copyright Elsevier 2016.

Table 4. Benefits and disadvantages of various dispensation composites.[24-36]

Technique	Advantages	Disadvantages
Density Moulding	Low time to setup Low wastage of things The low expense for complex and large compounds Perfect completion at the surface Pressure supply is even on the compounds	Manufacture speed is low Moderately curved or flat composite shapes are only suitable for this process
Sheet Moulding	Appropriate part for reproducibility The volume production ability is very high Fewer requirements for labor on a manufacturing level	Suitable for low fiber composites processing
Extrusion Moulding	Fast to setup Low costs for beginning setup Lower manufacturing costs	Reasonable speed for production Only appropriate for the composite preparation with cross-section being uniform Precision is ordinary
Injection Moulding	Lower cost of operational Low mass fabrication costs High output Elasticity to make complex shape portions Highly precise	High prices for initial setup
Resin Transfer Moulding	Better consistency of the product compared to compression molding Tight tolerances and more intricate parts than injection molding low cost for setup and fast time to setup	More wastage of material compared to compression molding Slower production speed than that of injection molding
Thermoforming	Low conservation cost Continuous, higher production volumes, many types of raw materials	High wastage of material
Pultrusion	Good usage of material, good contents of resin, high throughput can be achieved	Accumulation of the resin and fiber at the die opening. Decrease in the strength of the part when the resin is used excessively

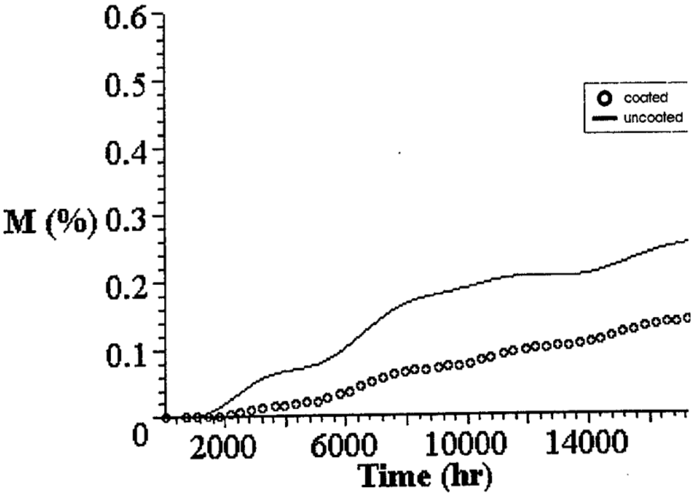


Fig. 3 Moisture responses to coating with a thickness of 0.254 mm.

wheat gluten composites using the CM technique to prevent chemicals, maintaining the composite's mechanical properties. The wheat gluten plasticized with water has a significant melting point, 110 & 170 ℃ with a melting enthalpy of 1.2 KJ /g. The wheat gluten of 60 percent and jute content of 40 percent placed at 160 ℃ for about fifteen mins has optimum property. Also, specific mechanical properties were identified to have tensile strength, flexural strength, and tensile modulus (20 MPa, 69 MPa, and 7.7 GPa). All these components are more substantial than the PP (Polypropylene Composites). It was found that the composites made of wheat gluten are having a less tensile and flexural property with 90 percent humidity when compared to the properties of 65 percent humidity. They are the same as that of the PP (Polypropylene Composites).

Sheet molding is one of the production methods used in the composite, a type of compression molding technique. The specific resin of measured quantity is distributed onto a plastic carrier film in this process with a paste reservoir placed under a chopper. The fibers are then cut onto the surface.[37] After the fibers have been put in the resin paste, another sheet is applied to the top layer. The sheets are taken for material storage to the

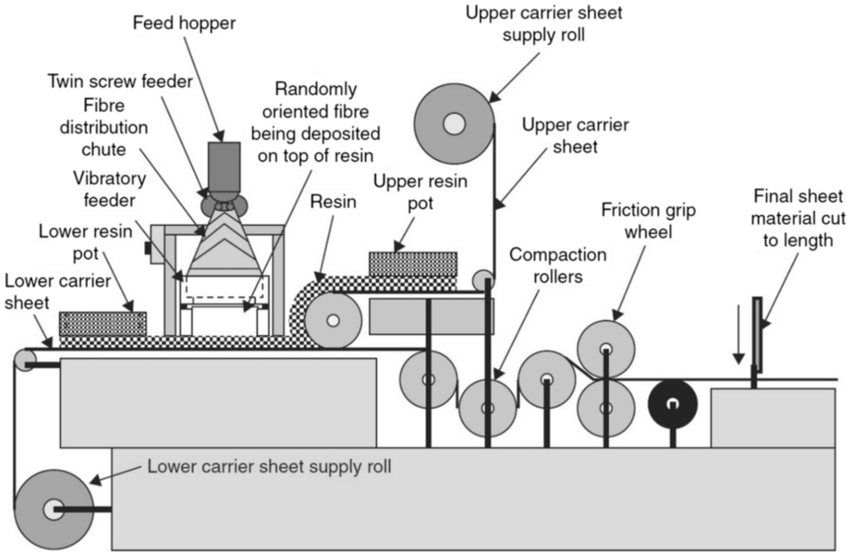


Fig. 4 Sheet molding Process flow-illustration based on the production of composite. Reproduced with the permission from [10], Copyright CRC Press 2019.

take-up roll after compacting them. The material is molded and cut under high pressure and heat after stripping the carrier film to create a composite with the desired form. The resin is then removed from the mold until the resin is entirely cured.[10] Fig. 4 shows the process diagram of the sheet molding technique.

3.2 Extrusion molding

This technique is preferable due to the high strength and high stiffness of the composites, and they are straightforward to form with this method.[10] The procedure begins with the preservation in a hopper of the thermoplastic material like pellets or granules. They are then sent to be molten in a hot barrel. Subsequently, the molten plastics are then used for the appropriate composite structure. The last step is product cooling. Extrusion of one and two screws is used widely to apply natural composites, including the PLA matrix. Substantial compounding and good functional versatility are the key advantages of this method.

3.3 Injection molding

It is used mainly for the manufacturing of plastic parts but also for the fabrication of natural fibers. Short fibers are used as particles or powders in this process. The process begins by placing the polymers of granules and pellet forms into the hopper and then heating them till they are in their molten state. Molten polymers are inserted in a chamber consisting of a separated die mold where it is kept for a while. The mold is opened, allowing the material to be cooled down to solidify.

Lou *et al*. proposed a framework where the wood plastic composite is injected through the molding process.[38] The sawdust of 5, 10, and 15 grams is melted and blended with PP using injection and a single-screw machine. By this process where a sawdust or PP composite will be formed. The screw extruder tank temperature is set for all three tanks at 175 ℃ with a speed of 26 rpm. The injection molding machine for the three tanks is set at 190 ℃ and 200 ℃ for die. From the outcome, it is understood that the wood-plastic is slightly lighter than that of the PP composite. The wood plastic composite is 5 percent more lightweight, and this property adds more convenience for usage.

Mohanty *et al.* developed industrial hemp fibers and soy-based bioplastics by injection molding to find wood plastics' mechanical and thermal properties. As shown in Fig. 5, the composites and compressive strength module improved as the hemp content of fiber content was enhanced. Fig. 6 shows that with 30-wt% fiber loading, ductility strength and stiffness increased by 3 and 10 times, respectively.[39]

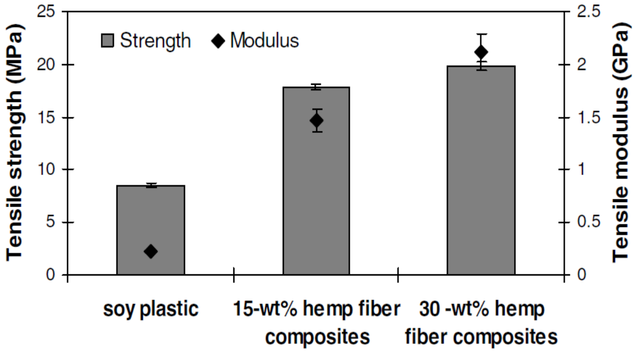


Fig. 5 Compressive properties of hemp FR soy-based environment compounds. Reproduced with the permission from [39], Copyright 2005 Springer Science Business Media, Inc.

Fig. 7 shows the notched Izod tensile strength of the compound. It was found that with the additional fiber, where the strength of the composite will increase. The increased impact strength was found to be 40 percent with 30 percent of fiber. Through this, the fiber has a significant impact on the strength of the soy-based bioplastic. It is attained due to the fiber reinforcement's enhancement utilizing 30 percent weight with 35 ℃. The following Fig. 8 describes the heat deflection temp value of the various bio-composites and bioplastics.[39]

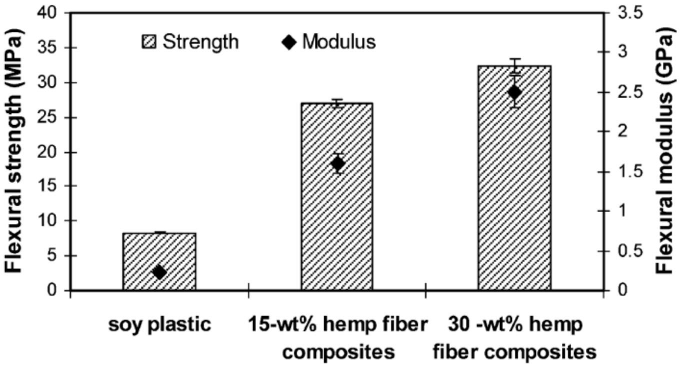


Fig. 6 Flexural properties of the hemp FR soy-based environment compounds. Reproduced with the permission from [39], Copyright 2005 Springer Science Business Media, Inc.

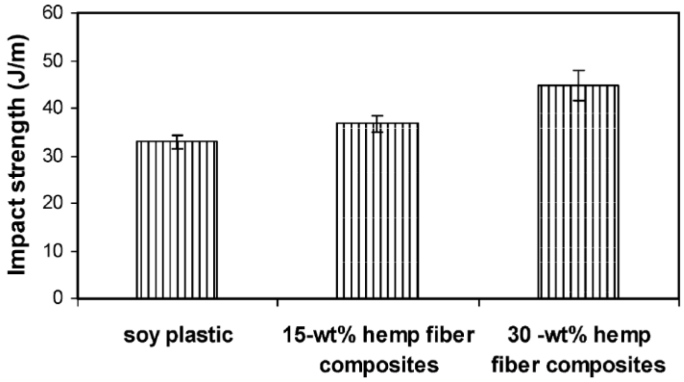


Fig. 7 Compressive strength of the hemp FR soy-based environment compounds. Reproduced with the permission from [39], Copyright 2005 Springer Science Business Media, Inc.

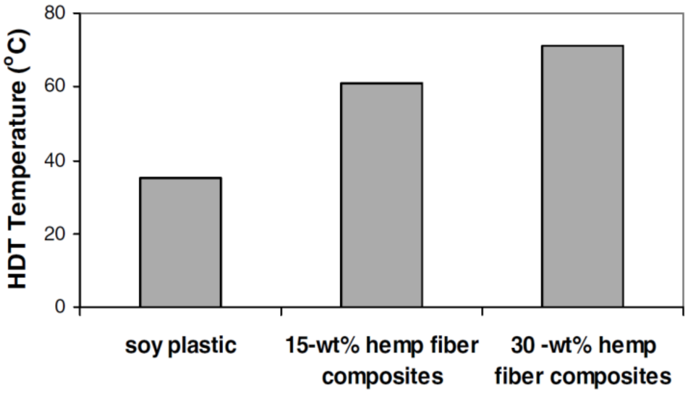


Fig. 8 Heat refraction temperature of the hemp FR soy-based environment compounds. Reproduced with the permission from [39], Copyright 2005 Springer Science Business Media, Inc.

Bamboo or polybutylene succinate (PBS) was manufactured by injection molding, and a study of the composite's mechanical properties was done.[40] For manufacturing, the use of a twin-screw extruder was made, and PBS was mixed with pellets of 40% wt. to prepare the specimen where they were charged into an injection mold. For fabrication, the specimens are placed under Izod impact, immersion analyses, and tensile behavior. The injection mold process was done at 180 ℃. The thermostat is utilized for controlling the mold temp at 40 ℃. After carrying out the injection molding, the specimens are placed under a temperature of 23 ℃ with a moisture level of 50 percent for about seven days. The impact strength gets increased along with increasing BF.

The investigators explored dynamic mechanical properties and surface morphology of short sisal fiber reinforcements with injection-molded PLA and PP composites.[41,42] The findings from scanning electron microscopy (SEM) have shown that PLA has more outstanding fiber communication than PP.

3.4 Thermoforming (TF)

This technique involves heating and then shaping the material according to the appropriate geometric parts at a flexible temperature.[43] The composite sheets according to their thickness are then pre-cut and using a clamping system to prevent warping and torsion. Next, the sheets are heated at the softening temperature with heaters to avoid the risk of material shearing and fracture during deformation. FR PP sheets were manufactured by the TF method. The sheet was heated to place in an oven for about 20 mins, which helps form temperature, then it is cooled to form die. The manufacturing process of PLA is done with the help of TF to the foams, films, and fibers. All these helped in combining the applications. Lim *et al.* suggested that the Al molds are the best for TF cycles.[44]

3.5 Resin transfer molding

The resin transfer method is used to produce continuous mats, woven roving, and discontinuous lignocelluloses.[45] After preheating, the resin is loaded in a holding chamber rather than using an exposed molding. This method was ideally suited for average capacity in production. Textile layers are pre-arranged in the resin, and solid mold was inserted to saturate the process. The essential advantage of the use of this approach is that the fiber orientation can be controlled easily.

The chemical method was used for treating the Abaca fiber and blended using epoxy resin with the help of resin transferring mold.[46] The component and structure analysis is used to decrease the lignin, deterioration of the lumen structure, functional alternation in surface, chemical, and hemicellulose content. With the help of the mentioned modification, the weak links and void areas are used for influencing and transferring the heat flow and tensile flow.

3.6 Vacuum resin transfer molding

Vacuum bags are used to facilitate the transition of resin flow instead of a mold device.[47] The used resin's viscosity should keep below as the vacuum pump's pressure is limited. That is a high-end component production method. Using vacuum-assisted resin molding for manufacturing composite flax fiber specimens and comparing the results is done with the reference data of various components.

The manufacturing of the panels is specially done through the VARTM process.[47] The specimens follow the rules and regulations of the American Society of testing and materials.

Moisture must be removed to increase the composite strength of flax fibers.[48] The fibers are covered with the help of the panel molds. It is completed after drying the fibers in the oven for about fifteen minutes at 110 ℃. A pressure of 1.6 bar is used for injecting the resin. The constant curing temp is used every two hours for about 180 ℃. the curing temp post the process was maintained at 120 ℃ for about one hour. Through which the fault in the panel can be determined. The panel manufacturing is done with the help of an Ultrasonic C-Scan. With the use of this process, the defect in the panel area is identified and cured. These test outcomes revealed that the specimens used for preparing the composite have high mechanical properties, which showed they are higher than the reference test outcomes.

3.7 Pultrusion process

The main difference between the protrusion and extrusion process is in pultrusion, in which the material is pulled rather than pushed through. The continuous length of the fiber tapes is drawn through the thermosetting polymer resin bath, with the exit die cross-section providing its final structure. The shape of the products depends on the die shape, in the form of a bar or rods. Based on the length requirement, the end products are cut, preserving that compounds were needed in the die. This method has the crucial benefit of constantly working for the manufacture of parts of constant transverse composites.

3.8 Filament winding

The fiber components are unfounded, unceasingly, and distributed through a tank of resin in this method. These components are then distributed through a revolving mandrel and are coiled around the mandrel in a definite direction in a controlled manner.

Le *et al*. investigated the physical components of yarn-reinforced natural fiber materials.[49] The samples were made using filament winding processes for flax and kenaf fiber tubular, and their components are contrasted with composites of electronic glass fibers. An analysis revealed that some of the characteristics of electric glass fiber compounds are enhanced than flax fiber compounds. Still, the flax fiber compound's tensile strength was comparable to composites of electronic glass fiber.

The wound composite's filament has enhanced property as follows: less expensive, low density, and less solidity than the property of the wound's composite products of the synthetic filament.[50] And at the same time, the natural fiber-based reinforcement for PC (Polymer Composite) also positively impacts the polymer's mechanical property. The bio-composite materials are the ones found with significant property with the present wound product of the filaments.

4. Characterization techniques

Characterization techniques are essential to categorize whether the natural fibers are suitable for reinforcement for the polymer composites. Fig. 9 shows different characterization techniques used in green composites and are discussed below.

4.1 FTIR (Fourier-transformed infrared spectroscopy)

FTIR gives qualitative and quantitative information about natural fibers. The frequency of a spectrum of infrared absorption is obtained within the 40-400 cm^{-1} range from the organic structure of natural fibers.[27] FTIR results reveal propionyl & acetyl presence (acetylation & propionylation) on processed wood, hemp, and flax fibers. An increase in the amorphous content is seen due to the esterification process leading to a progressive decrease in the fiber crystallinity.[51]

FTIR offers a qualitative description of date palm fibers' chemical composition change. The broad absorption spectrum of the 3500 cm^{-1} regions results from hydrogen-bonded, oxygen-hydrogen bonding and aliphatic carbon-hydrogen bonding vibration gives a peak of approx. 2900 cm^{-1}.

According to the carbonyl clutch widening, the immersion band in the area of 1740 cm^{-1} was obtained. The bending vibration of the CH_3 group has an amplitude of around 1375 cm^{-1} band. FTIR has demonstrated that raw and treated fibers' chemical composition would not be altered significantly.[51] The FTIR spectrum shows that, after NaOH treatment, wax and fat content were removed from jute fibers mat reinforced

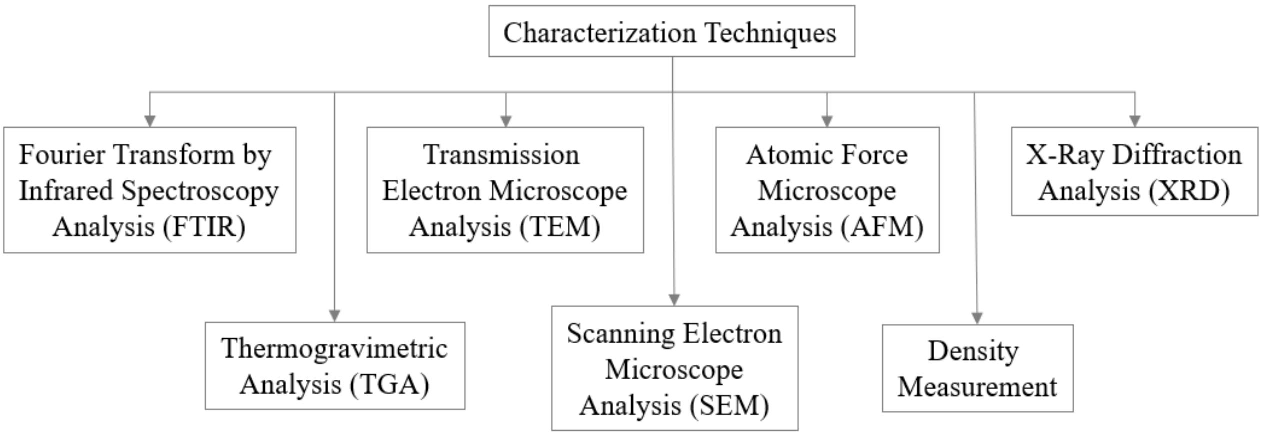


Fig. 9 Characterization techniques used in green composites.

polypropylene composites, and those improvements happened at the fiber surface topography.[52] FTIR spectroscopy shows that in the band region of 1505-1600 cm^{-1}, softwood polymers reveal high strength ratios for two lignin tops. Still, for hardwood polymers, the intensity proportion of carbonyl immersion will be higher at bands 1740 cm^{-1} to lignin tops.[53]

FTIR spectra revealed the lack of lignin and hemicellulose in NaOH-treated Kenaf- fibers, whereas untreated fibers show more hydrogen connection than processed fibers.[54] The bandwidth size intensity at 1731 cm-1 decreased, implying that Sterculia urens fibers had reduced hemicellulose content.[55] Ficus religiosa leaf fibers cooled cryogenically after they are powdered. Then, using potassium bromide (KBr), these powders were thinned up to 1%. Using the Perkin Elmer 16PC FTIR instrument, 32 scans at a resolution of 4 cm^{-1} were reported in the range of 4000–500 cm^{-1} for unprocessed and alkali samples.[56]

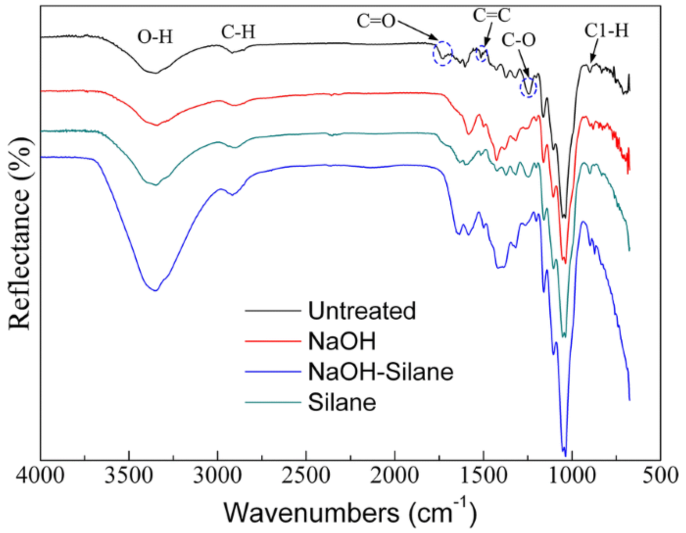


Fig. 10 FTIR spectra for treated and untreated corn stem fibers. Reproduced with the permission from [57], Copyright 2019, Springer Nature B.V.

The spectrum of FTIR corn stem fibers processed with various natural substances compared to the unprocessed fiber can be shown in Fig. 10.[57] The peaks at about 2917 cm^{-1}, 3349 cm^{-1}, 1513 cm^{-1}, 1723 cm^{-1}, 898 cm^{-1}, and 1243 cm^{-1} are the C–H, O–H, C=C, C=O, C1–H, and C–O bonds. When alkali treated, the C=O bonds widening from the pectin and hemicelluloses of the fresh corn stem fibers at the peak of 1723 cm^{-1} were no longer visible. A weak peak gave the C=C group's elongation shaking in the carbonyl of hemicellulose, and its absorbance at about 1513 cm^{-1} was lesser than the untreated material. Furthermore, when alkali-treated corn stems were processed or processed in NaOH- silane solution, the untreated fiber peak at around 1243 cm^{-1} was invisible for a long time due to C–O bond elongating. It was because of the absence of hemicellulose and lignin in the processed fibers. When silane treatment was conducted on corn stem fibers, it was suspected that stretching vibrations of Si–C and Si–O–Si in the spectral curve should occur. The peak observed at about 1243 cm^{-1} was lesser than the unprocessed fiber, suggesting the elimination of a portion of the hemicellulose during the silane process because of the influence of the mixture of ethanol or water.[58]

4.2 TGA (Thermogravimetric analysis)

This process measures the thermal decomposition, change of mass, and the composite material's thermal stability.[54] Physical and chemical properties may be determined as a temperature while maintaining a steady heating rate. The Thermogravimetric Analysis is done with a sample pan's help, which is supported using a balance. A furnace occupies the pan. The cooling and the heating process occur during the time of the research. The quantity example was controlled at the time of the investigation. The purge gas is used for controlling the environment, which is surrounded by the example.[57]

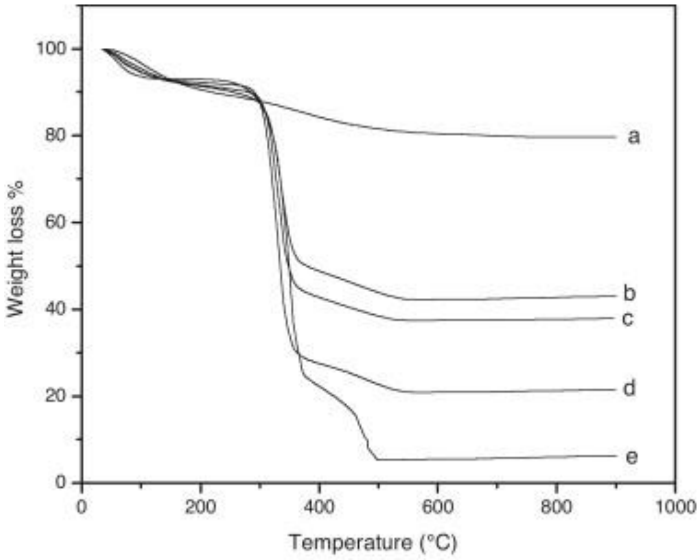


Fig. 11 TGA analysis curve of b) cellulose, c) cellulose with 16.67 in wt%, e) hybrid cellulose with 50wt%. Reproduced with the permission from [59], Copyright Elsevier 2012.

The TGA is used for characterizing the cellulose, hybrid cellulose, and hydrous niobium oxide. This process is done with Shimadzu TGA's help with fifty thermal analyzers with a temp of about 900 °C at room temperature by heating it at 29 °C for a minute in nitrogen. Following Fig. 11 describes the hydrous niobium oxide TGA analysis. In the TGA analysis, the weight loss takes in 2-stage with the temp range of 25 to 800 °C. The cellulose TG curve in Fig. 11b indicates three weight loss measures. The first is caused by water evaporation from the polysaccharide surface. The decomposition of lignin and cellulose can be attributed to the 2nd phase (182–404 °C). The residual lignin decomposition reactions are degraded in the 3rd stage (404–736 °C).[59] The existence of inorganic material had a minor effect on the sustainability of hybrid material, as shown in Fig. 11. The TG curves reveal that the prototype value is 50.0 wt% because it shows the minor dm from the 2nd level. It is due to the increased interaction between hydrated niobium oxide and cellulose.

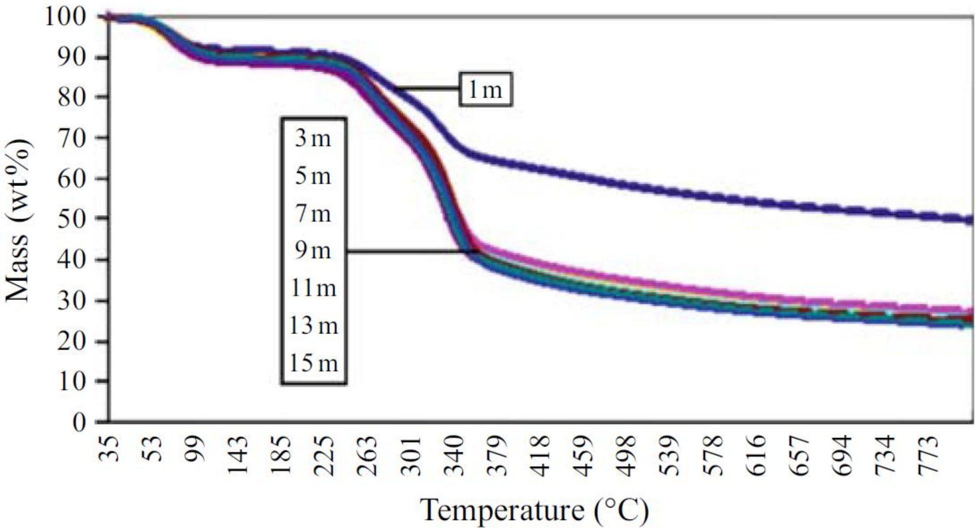


Fig. 12 Thermogravimetric analysis of sugar PF existed from various heights of sugar PT. Reproduced with the permission from [60], Copyright Springer 2012.

Thermogravimetric analysis of sugar PF was conducted, with the outcome of temperature increase from derivative quantity and loss rate of sugar PF is illustrated in Figs. 12 & 13. The thermal decomposition of sugar palm fiber takes place in four phases. The first phase was due to the moisture evaporation and then corrosion of lignocellulosic components of ash, lignin, cellulose, and hemicellulose. The derivative curves of thermogravimetric analysis showing the 1-meter fibers have better thermal stability than the 3-15 meter fibers. The high content of ash is the explanation for the improved thermal stability.[60]

Guimaraes *et al.* showed that the lignocellulose fibers such as sponge gourd, sugarcane bagasse, and banana have the highest thermal strength at a temperature of 200 ℃ while hemicellulose and cellulose decompose and reduce at 300 and 400 ℃ as shown in Fig. 14.[61] All the strands have a considerable loss at 100℃ with mainly the water loss because of the humidity level obtained in all the fibers. The fibers showcase thermal stability at the presence of 100 and 200 ℃. For banana fibers, the temperature should be considered at 200 ℃, which is the highest temperature for the natural fibers. If the temperature used above this level, the loss in all the fibers would be increased. For the case of above 300 ℃, the sample may showcase the highest level of failure, mainly because of the decomposition process of the hemicellulose and the fibers' cellulose. If the temperature is above 400 ℃, the fiber's performance degradation will be more, leading to bread in the bonds of the fibers.[61]

4.3 Differential thermal analysis

DTA is a technique for defining and quantifying the physical properties of substances by monitoring the sample's dynamic properties as it was heated. The method is based on the reality that when the material was heated, it goes through phase changes and reactions that include heat emission and absorption.

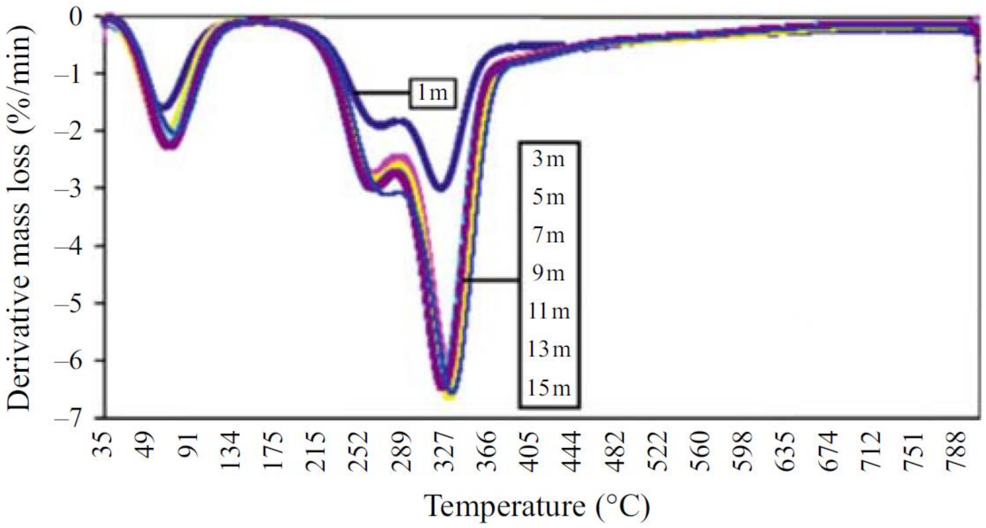


Fig. 13 DTG of sugar PF existed from various heights of sugar PT. Reproduced with the permission from [60], Copyright Spring 2012.

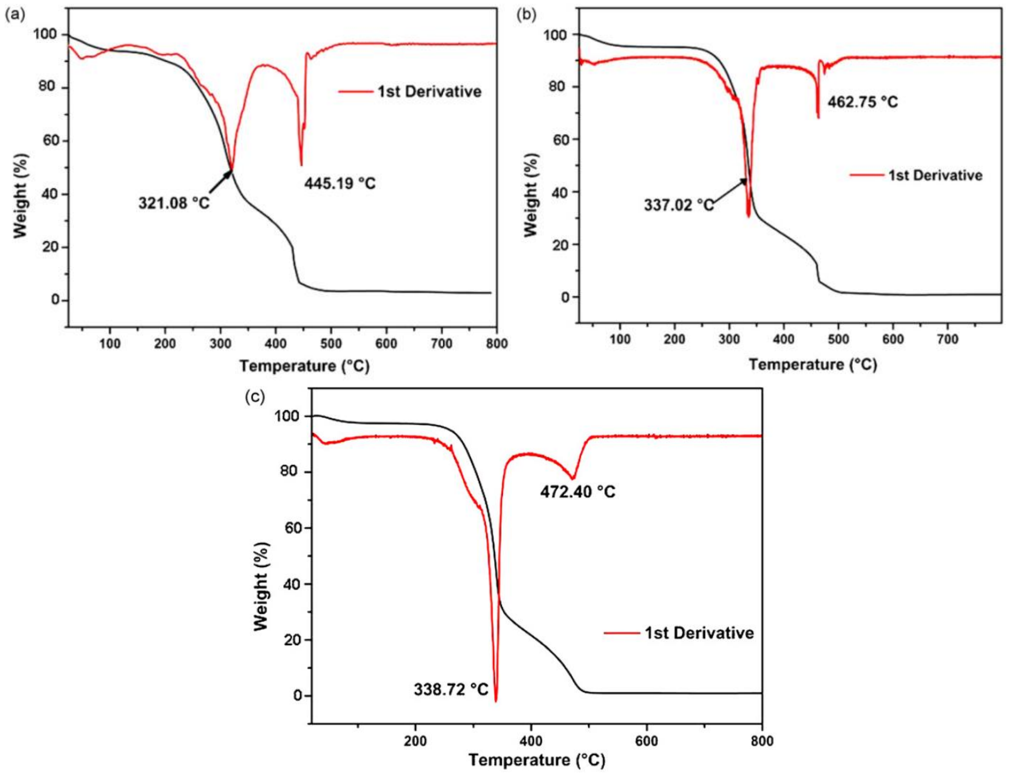


Fig. 14 Thermogravimetric analysis curves: (a) banana fiber, (b) sugarcane bagasse fiber, and (c) sponge gourd fiber. The dynamic flow of air, 20 mL min^{-1} 10 °C min^{-1}. Reproduced with the permission from [61], Copyright 2009 Elsevier Ltd.

This method measures the test material's temperature compared to that of inert material. One thermocouple is inserted into the piece to be tested, and another is integrated into the inert material. These are linked, so that heat difference produced through the heating process is recorded graphically on a moving chart as a series of peaks. The level of temperature and heat present in the fiber leads to the change. This change mainly occurs in the element characteristics, substance identification. These changes can be cured by dealing with the fibers with the help of DTA curves with both the known and the unknown compound elements. The substance present in the composite will be established with the help of the graph with the use of peak point in the graph. The level may be noted by associating it with the peak area. The DTA analysis mainly analyzes the mineral and its mixtures present in the composite combination of the fiber.[57]

The temperatures at which these changes occur and the amount of heat involved are typical for the individual compounds and elements. Therefore, a substance is identified by analyzing DTA curves and comparing them with the DTA curves from known compounds or elements. Also, in the composite sample, the quantity of a substance is related to the area under the peaks, which can be found by comparing the specific peak area to the standard samples evaluated under the same conditions.[62]

The treated and unprocessed thermograms filaments are analyzed in Fig. 15. A broad endotherm between 50 and 200 ℃ shows the water atoms' occurrence in unprocessed and treated kenaf filaments. No endothermic or exothermic responses are seen somewhere between 180 and 280 ℃, recommending that the filaments are steady between these temperatures. As per the principal deterioration temperature, the untreated fiber gives a higher worth than the treated fiber showing that it is thermally steady. Nonetheless, the later three disintegration temperatures demonstrate that the treated fiber is thermally more steady than the untreated one.[63]

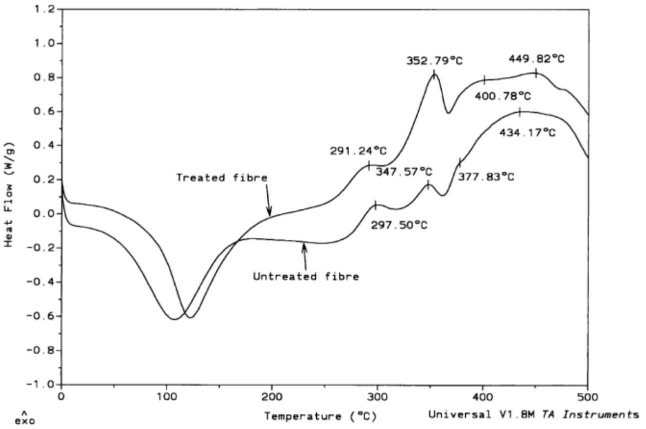


Fig. 15 Differential thermal analysis of untreated and treated (6% NaOH) kenaf fiber. Reproduced with the permission from [63], Copyright 2003 Elsevier Ltd.

Differential warm investigation bends of unprocessed and preserved hemp strands appear in Fig. 16. A wide endotherm saw in the heat scope of 50–175 ℃ in both the unprocessed and processed hemp fiber shows the water particles present in the filaments. The exothermic pinnacles, which are more significant than 200 °C, show the deterioration temperatures of the fiber in the filaments. The locale somewhere in the range of 150 and 240 °C reveals no endothermic and exothermic responses that recommend that the filaments are steady in this high temperature. The exothermic tends to be derived that the processed hemp fiber was contrasted with the unprocessed fiber since the untreated hemp filaments have lower decay temperatures.[63] DTA was performed on kenaf and hemp bast fiber, showing extensive endothermic DSC crests in the thermograms, indicating moisture. An occurrence of water in compounds causes the material to become plastic, reducing its tensile strength.[63]

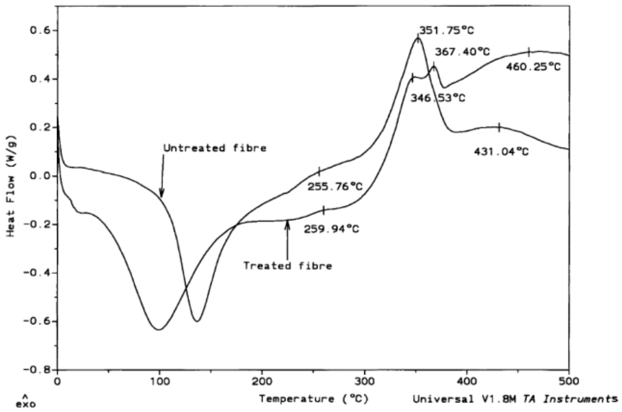


Fig. 16 Thermal analysis of the treated and the untreated hemp fiber with NaOH. Reproduced with the permission from [63], Copyright 2003 Elsevier Ltd.

4.4 Transmission electron microscope (TEM) analysis

This approach can calculate the diameter of the fibers, monitor specific sublayers, and measure thicknesses.[64] Minute measurements with TEM micrographs may also be measured. However, this requires a detailed follow-up of the surface preparation process. This assumes boiling the fibers in a 1% NaOH solution twice for approximately 3 hours. Processing of the fibers is then done with 0.05 mol/liter hydrochloride solution followed by washing with liquid. These fibers are then located for 4 hours in purified water to generate an example that enables the fibers' structure to be analyzed in TEM carefully. Fig. 17 illustrates the TEM analysis of the standard fiber and the lotus root fiber.[65]

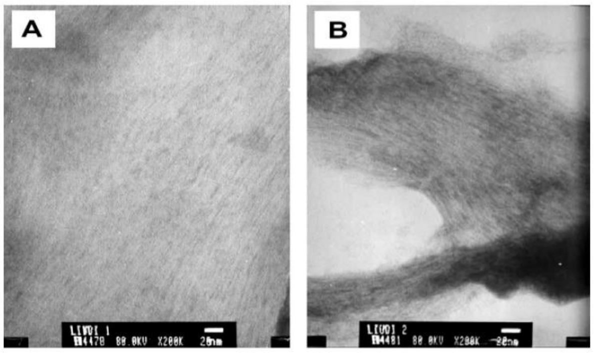
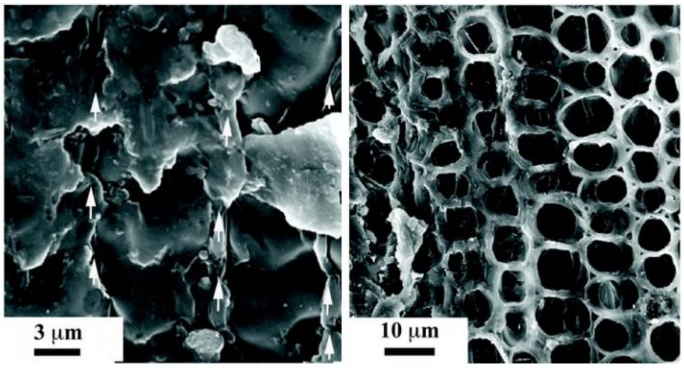


Fig. 17 TEM images of the two fibers: standard fiber (left) and Lotus root fiber (right). Reproduced with the permission from [65], Copyright 2008 Elsevier Ltd.

4.5 Scanning electron microscope (SEM) analysis

SEM is the essential method for understanding the distribution of every fiber in a network. It gives high-resolution images of the fibers when the electron beam is concentrated on the surface, and the backscattered signal is observed. SEM analysis has shown that raffia textilis fibers are made up of superimposed layers. The top layer comprises longitudinally aligned fibers with the rulers, whereas the lower layer looks like the honeycomb model. Thus, these honeycomb structures and the scales support bond the compound milieu mechanically.[66] The SEM micrographs of raffia textilis are shown in Fig. 18.

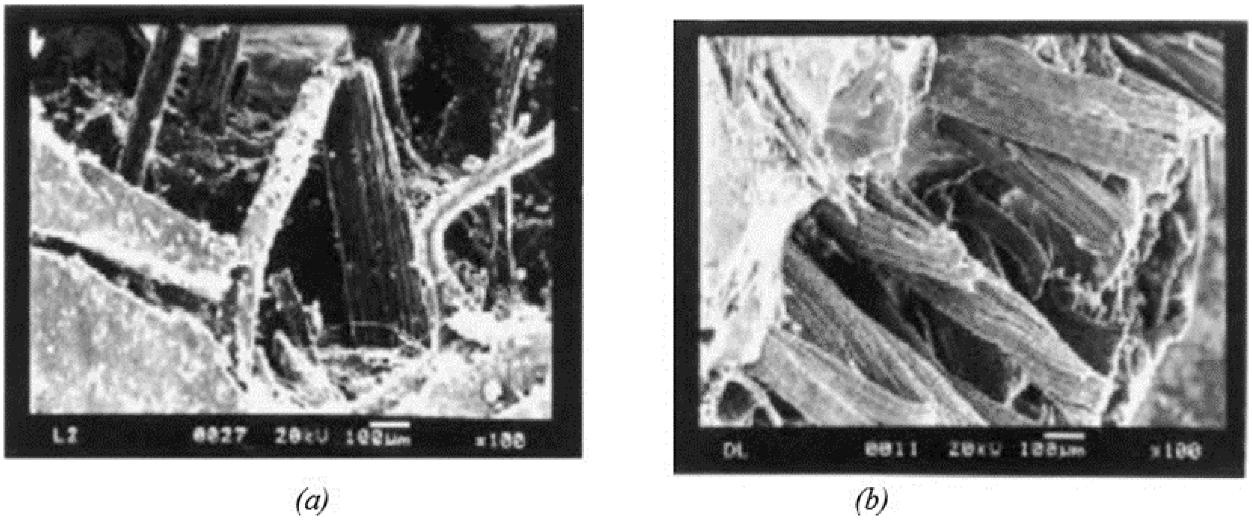


(a) (b)

Fig. 18 (a) Scanning electron microscope micrograph of the top layer of raffia textilis fiber display three columns of tiles accidents isolated by a slight vertical interface (white bolts) lined up with the longitudinal hub of the fiber. (b) Scanning electron microscope micrograph of the base layer of raffia textilis fiber, Reproduced with the permission from [66], Copyright 2003 Elsevier Ltd.

The SEM shows that the presence of treated fibers and bonding agents means that a higher adhesion between oil palm and sisal fibers is possible. The wetting process of the fibers is done using the rubber matrix. The significant adhesion between the palm oil fibers and the sisal oil fibers can be identified with the Scanning Electron Microscope's help. Fig. 19 describes the palm oil fiber's tensile strength and the sisal oil fiber-filled using natural rubber composites. Few holes can be found in the rubber matrix after the fiber is removed from the matrix, clearly showing the wetting process's low level between the matrix and the fiber. Better adhesion can be found by using a significant bonding agent shown in Fig. 19(b) in the rubber matrix.

Fig. 20 shows the SEM analysis of the untreated and the treated Populifolia with the alkali at 5%. Fig. 20(a) describes the organic fabric where a white coat was formed on the fabric's external layer. It is due to the joined components of lignin and hemicellulose; however, it may be neglected in the alkali process in Fig. 20(b), confirming the elimination of the hemicellulose content in the fabric. These pictures help to see the fabric's knit form due to void areas in the fibers. Specific geometric structure helps in penetrating the resin along with the void areas. The outcome shows the significant attachment



(a) (b)

Fig. 19 (a) SEM malleable break surface of oil or sisal PF filled regular elastic compounds at 30 phr stacking amplification 100. (b) Scanning electron microscope tractable break exterior of oil or sisal PF filled standard elastic composites at 30 phr stacking in occurrence. Reproduced with the permission from [65], Copyright 2008 Elsevier Ltd.

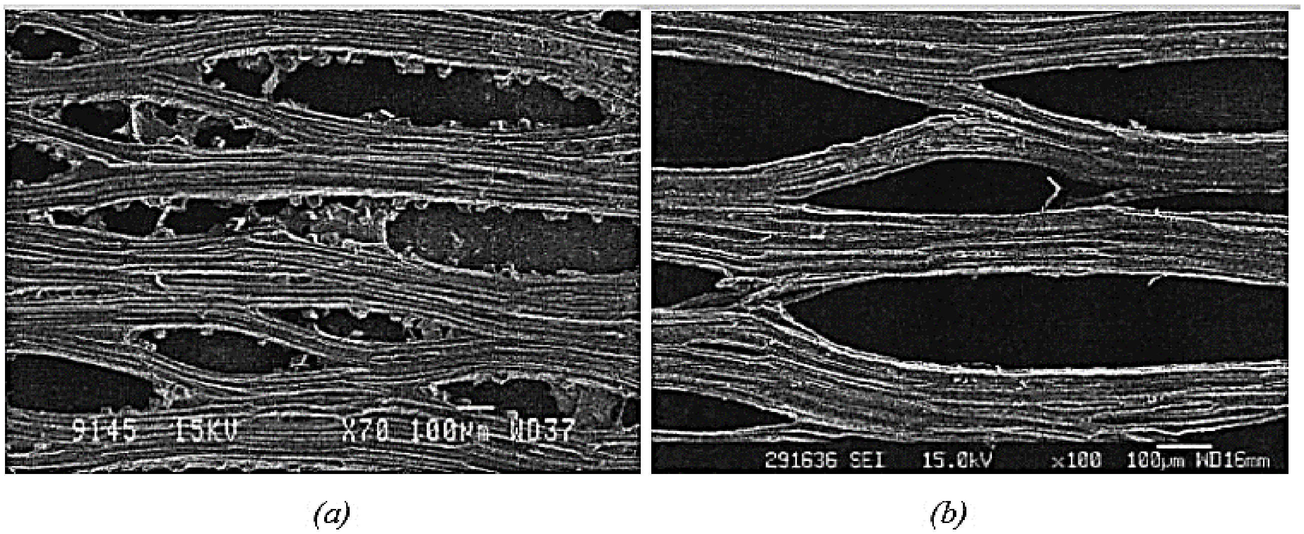
level in the matrix and the fabric, which attains an expected level.[63]

SEM has been used to consider sisal fiber fracture mode, which is shown in Fig. 21. The study examined both primary and tertiary wall delamination and fiber-cell delamination. The minor nonlinear section in the stress delamination in the fiber cell curves was reported when delamination between the fiber cells before the strength collapsed.[67]

4.6 Atomic force microscope (AFM) analysis

This technique can measure the surface profile of the fibers up to the sub-nanometer levels of resolution. AFM measures repulsive and attractive forces between the fiber and the cantilever tip. The forces governing the adhesion phenomena are also measured explicitly by AFM. A high peak beam needs to be scanned over the fiber in this process. The repulsive forces produced by the fiber deflect the cantilever tip when it contacts the fiber. The amount of cantilever deflection can be detected and tracked using a photodiode detector and a laser beam. The roughness parameters obtained by this measurement are the 10 points R_z, R_t, R_a, R_q, R_{ku}, and R_{sk}.

Natural flax fibers are primarily used for mechanic separation and dewretted after this process. They are sent for the 3-different class of chemical processing. The first level of chemical processing is done using subjecting the fibers with 10 g/l of NaOH and ethanol solution for about two hours at 78 °C. After this process, the fibers are submerged with H_2O ethanol and then dried under 20 °C with a relative humidity of 50% at room temperature. In the second chemical processing, the fibers are subjected to 10 g/l of NaOH and ethanol solution for about two hours at 78 °C. They are then rinsed with water, ethanol and dried under room temp. Then the fibers are immersed in AA (Acetic Anhydrite) for about an hour at 20 °C. In the final chemical processing, the fibers are immersed with 99% formic acid for about forty mins at 20 °C. After this process, they are rinsed with H_2O, then dried at room



(a) (b)

Fig. 20 Scanning electron microscope of (a) unprocessed and (b) 5% sodium hydroxide treated H. Populifolia fabrics. Reproduced with the permission from [68], Copyright 2006 Wiley Periodicals, Inc.

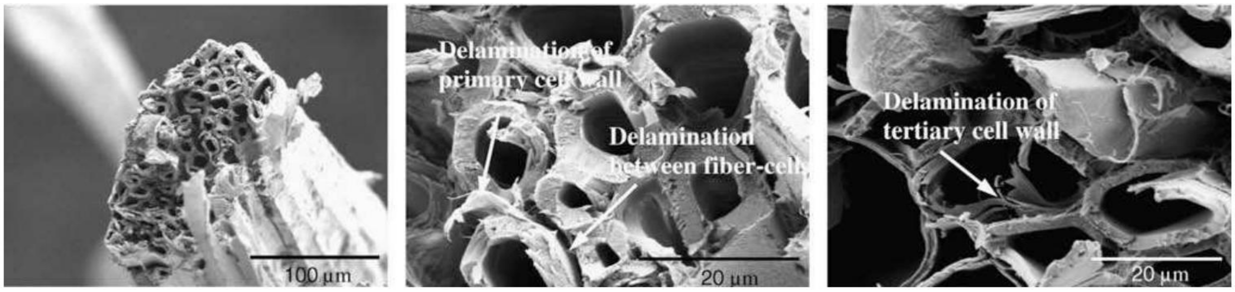


Fig. 21 SEM of the sisal fibers:(a) total view, (b) and (c) particulars of fiber-cell, fiber-wall, and fiber-cell fracture. Reproduced with the permission from [67], Copyright 2008 Elsevier Ltd.

temperature with 20% humidity for about 24 hours. The above-mentioned chemical processing is done to modify the fibers based on their design slightly and separate them from other impurities through the retained fiber's mechanical performance. The basic glue level is applied to the fiber through cyanoacrylate glue before the Atomic Force Microscopy observation process.[68]

The following Fig. 23 describes the AFM images in contact mode of various flax fibers. These images are the representation before and after chemical processing. The illustration of raw fiber image under AFM where one could see the fibers' heterogeneous surface almost covered by grains for about 500 to 1000 nanometer and in height level of 150 to 300 nanometers. Figs. 22(b) and (c) describes the fiber's efficiency after chemical processing, especially on the roughness of the raw flax fibers are noted carefully. It was found that the grain sizes and numbers are reducing due to chemical processing. It brings a positive result on its surface because they are smooth and uniform. The topography demonstration of fibers treated using NaOH is shown in Figs. 22(b) and (c). The fibers are found with a certain level of variation in height, and only a tiny amount of grain was identified on the fiber surface. Fig. 22(d) shows the fiber treated using formic acid in this process. The initial amounts of grains are not found on the fiber surface. With the help of the alkali process, the surface contaminations present in the fibers responsible for creating surface roughness are wholly eradicated.[68]

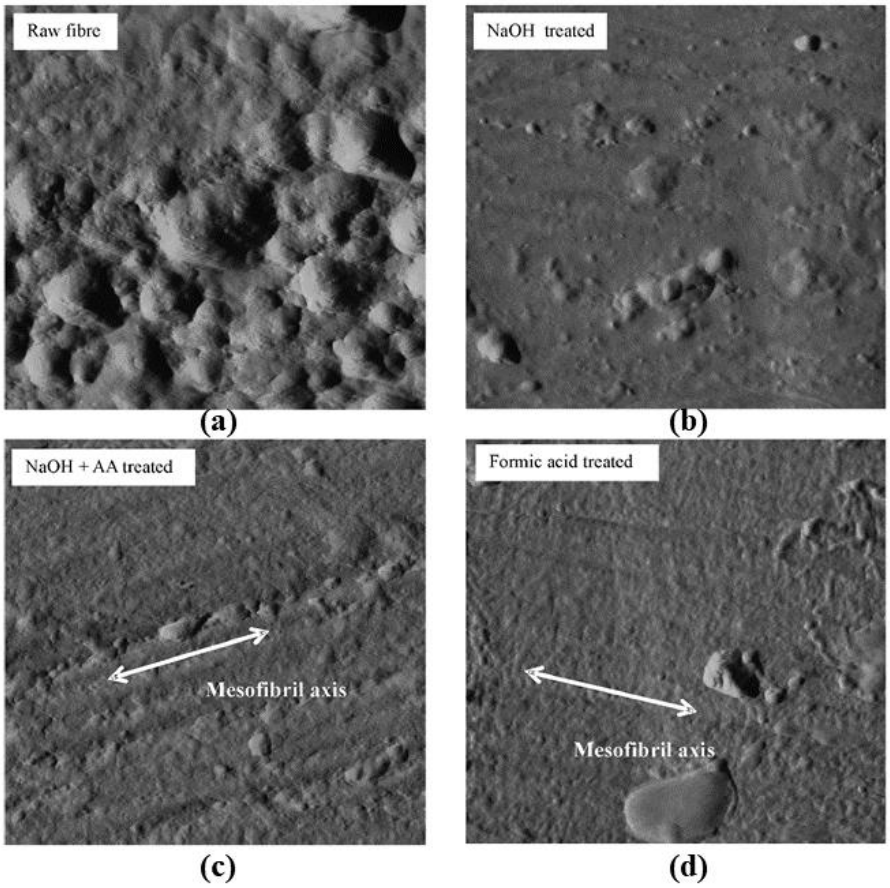


Fig. 22 Deflection process of the Atomic Force Microscopy image on various flax-fibers, a) image of raw flax-fiber, b) flax-fiber treated in sodium hydroxide, c) falx-fiber treated in sodium hydroxide + acetic anhydride, and d) flax-fiber treated with formic acid. Reproduced with the permission from [68], Copyright VSP 2007.

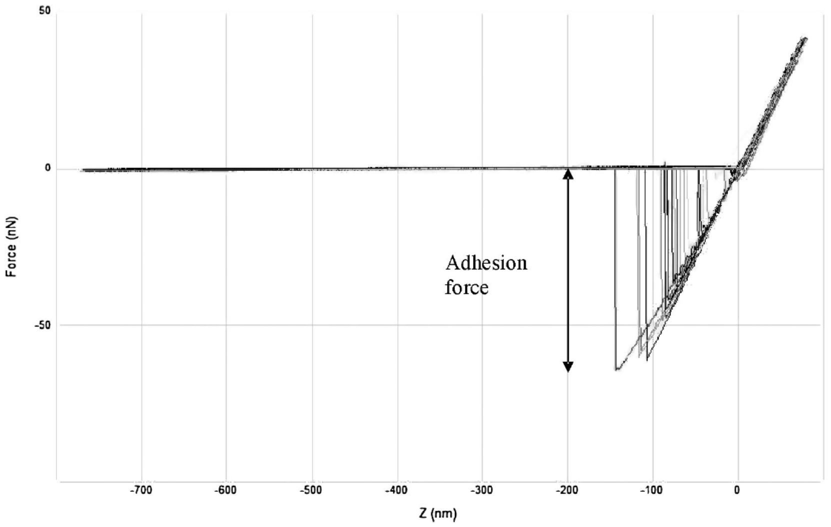


Fig. 23 Illustration of force curve between the Atomic Force Microscope and the raw flax fiber. Reproduced with the permission from [68], Copyright VPS 2007.

The fibers force is measured using regular Si3N4 (silicon nitride) in the atomic force microscopy under 20 °C and 50% RH. According to the manufacturers, the peak takes a curvature range valued to be ten to thirty nanometers. The chosen cantilever spring is scaled through the cantilever's reference of the forced constant.[68]

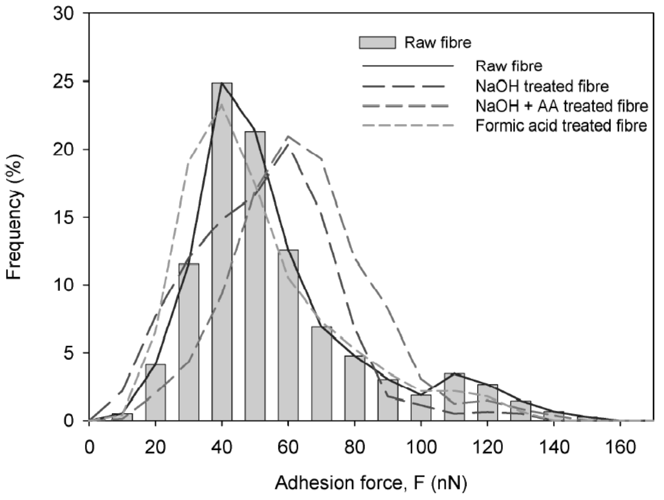


Fig. 24 Measurement of adhesion force between the fibers. Reproduced with the permission from [68], Copyright VPS 2007.

Fig. 23 describes the illustration of the atomic force microscopy curves found between the raw fiber flax and the silicon nitride Atomic Force Microscopy. Fig. 24 illustrates the adhesion force measured between the converted force and the distance curves done with the histogram's help. The wide circulation of the bonding powers per fibers should be credited to the compound heterogeneity of flax filaments in the nanoscale and at the same time firmly reliant on the harshness of example.[16] It is the reason that the space of the link between the surface and AFM peak provides an impact on the subsequent grip power in the two items. The attachment power corresponds to an external territory, which was impacted through the inclination surface at the connection point. In any case, a reasonable variety in the grip powers amid the various examples. Fig. 25 shows the middle estimation of the combined bends for different strands. These bends uncover the attachment powers and changes as F (formic acid) < F (raw fiber) < F (NaOH) < F (NaOH + acetic anhydride).

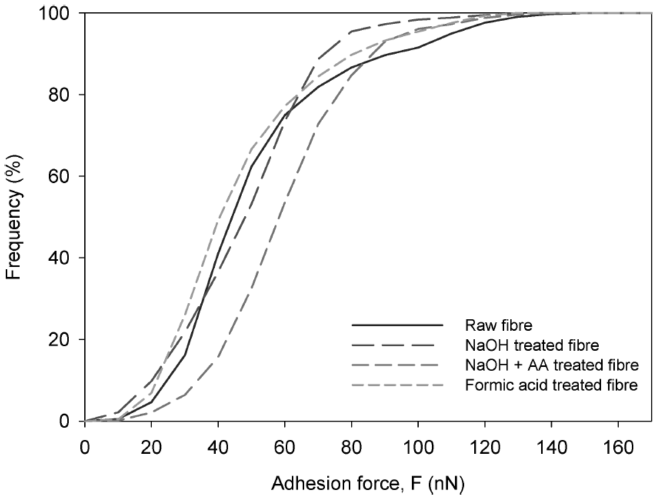


Fig. 25 Cumulative adhesion force curves for the different fibers. Reproduced with the permission from [68], Copyright VPS 2007.

4.7 Density measurement

The fibers need to be dried in a calcium chloride-containing non-hygroscopic desiccator for 48 hours to measure the density of fibers.[69] Subsequently, the fortitude was saturated for 2 hours in toluene to extract microbubbles from the fiber. The fibers are then trimmed to 5-10 mm in length and stored in the pycnometer.[64,70,71] The mass of the fibers may be measured through the following equation,

$$\rho = \left(\frac{(m_2 - m_1)}{(m_3 - m_1)\big((m_4 - m_2)\big)}\right) \times \rho T$$

where, m_1 was the quantity of unfilled pycnometer, m_2 is the total quantity of the pycnometer packed with fibers, and m_3 is the quantity of the filled toluene fiber and pycnometer m_4 as the fiber quantity occupied with toluene.[72-74]

4.8 X-ray diffraction analysis (XRD)

This method depends on X-rays' particle nature or dual wave to determine the natural fiber's chemical structure and crystallographic design.[75,76] The identification and representation of mixtures based on their diffraction design is a critical application of the strategy. When an episode light discharge X-ray cooperates with an unbiased material, the dissipation of those X-rays from molecules within the detached material is the most significant effect. In materials with an ordinary design that is glasslike, the dissipated X-rays go through the productive and damaging obstruction. This was the interaction of deflection.[57]

This method permits the scanning of natural fibers under the two theta (2θ) range. The higher the crystallinity index, the more will be the current solidity.[77] The determination of CI can be done by the following equation:

$$CI = \left(1 - \frac{I_{AM}}{I_{00}}\right) \times 100$$

where, I_{00} is the strength of the crystalline phase, and I_{AM} is the strength of the amorphous phase.[78] The CS is determined by the following equation:

$$CS_{000} = \frac{0.0893\lambda}{\beta cos\theta}$$

where, β was the width at half-maximum and θ was the Bragg angle.[79]

XRD study shows that closer cellulose packing increases the CI of A. leucophloea fibers when alkali-treated and enhances the mechanical things such as elongation, compressive, and modulus strength.[80] An XRD study displays that the Althaea Officinalis L. fiber CI value was 68%, while the crystallite size was 2.4 µm.[81] X-ray diffractogram states that micro-fibrils in Agave leaves have improved CI than the untreated fiber and regenerated cellulose composite films primarily because non-cellulosic materials are not present.[82] XRD showed roughness on Hildegardia lignocellulosic fibers' surface and improved crystallinity after eight hours of alkali process.[83] X-ray diffractogram shows that mercerized nanofibers' crystallinity because of high sodium hydroxide concentrations was lower.[62] In the Sansevieria cylindrical fibers, XRD shows cellulose Iβ and has a CI of 70%.[84] Fig. 26 shows XRD diffractograms of some natural fibers used to calculate their crystalline index. The peaks displayed to the corresponding crystalline planes: 1st peak (1–10), 2nd peak (110), 3rd peak (200), and 4th peak (1004). The planes relate to the Iβ-monoclinic crystalline form, and the peaks (1–10) and (110) are most probably overlapping for higher plant cellulose, such as cotton and curaua. During the assessment, the 1st and 2nd levels of intensity may be evaluated and representative of cellulose content. It can be seen that these peaks for curaua are more pronounced than for buriti; *e.g.*, these suggest more fibers ordered for curaua. The determination of the fibers crystallinity index is done by taking the sum of 1st area (2h & 15), 2nd area (2h & 22), 3rd area (2h & 35), which is then being divided by the whole integral curve.[85]

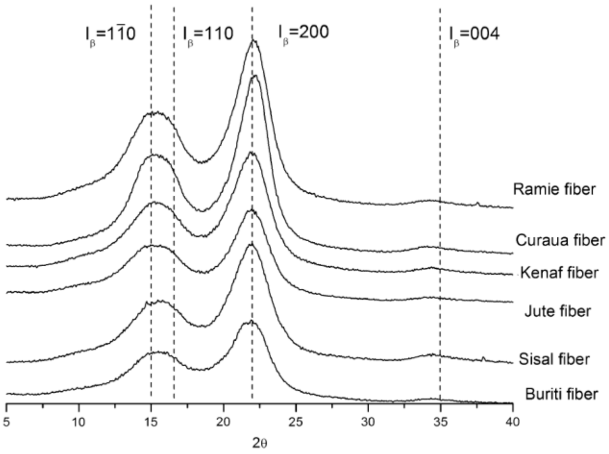


Fig. 26 XRD diffractograms for the studied natural fibers. Reproduced with the permission from [85], Copyright 2013, Springer Science Business Media Dordrecht.

5. Conclusion

Due to the present situation of ecological concerns like global warming and to satisfy the needs of the new requirement, scientists are producing novelistic materials. Especially efforts have been put much in the field of construction with Natural FCs. The natural fiber composite is utilized mainly in a commercial application such as interior panel manufacturing in the automobile sector. Biomaterials in recent times are obtained from localized resources such as polymer-based composite through they are trying to make people aware of environmental factors. These natural fibers and polymers are trying to reduce the consumption of unsustainable products such as synthetic materials. Also, it was found that the NF cost is much lesser when compared to unsustainable materials such as synthetic fibers and materials. This review paper was made by keeping in mind the generation process of the commonly used natural fibers, matrices, and bio-polymers in present use. Which also presents the processing method and characterization technique of the Green Composites manufacturing process. The FRPC mostly depends on the reinforced material. Its natural behavior when natural fibers used along with composite tries to reduce the processing Temp up to 15 to 20 percent, and the weight is reduced by 25 to 30 percent. These characters of natural fibers are making many manufacturing companies use them due to their eco-friendly nature. It was also found that the chemical process and fiber surface can also increase the thermo-chemical, physical, and chemical properties of the natural filaments. Proper identification and characterization of the natural fibers encourage it to give novelistic sustainable composites, which industries will highly use. Therefore, it is essential to identify the characterization method which occurs in the natural fibers through which the products and application made of natural compounds will be mandated in the future. The following conclusion was drawn from the natural fiber material and composites. First, the surface and morphological behavior of the fiber can be grouped based on the FTIR, XRD, thermogravimetric, and SEM analysis. Through which the nature of the fiber can be understood clearly. Second, the Fourier Transform Infrared Spectroscopy (FTIR) showcases the treated fiber's hydrogen bond. The hydrogen bond in the fiber is more robust. Also, it showed that the natural fiber is having significant mechanical behavior as well. Third, the X-ray Diffraction (XRD) Analysis showcase the closer cellulose packing enhances the Chloride bond of the treated fiber. Also, the mechanical property like tensile strength, elongation, and modulus of the fiber at breakage has been improved. Last, the thermogravimetric analysis (TGA) revealed that the accumulation of natural fiber to the FRC would enhance the thermal behavior of the composite material.

Conflict of Interest

The authors declare no conflict of interest.

Supporting information

Not applicable.

References

[1] T. Ohkita, S.-H. Lee, Thermal degradation and biodegradability of poly (lactic acid)/corn starch biocomposites, *Journal of Applied Polymer Science*, 2006, **100**, 3009-3017, doi: 10.1002/app.23425.

[2] R. Burgueño, M. J. Quagliata, A. K. Mohanty, G. Mehta, L. T. Drzal, M. Misra, Load-bearing natural fiber composite cellular beams and panels, *Composites Part A: Applied Science and Manufacturing*, 2004, **35**, 645-656, doi: 10.1016/j.compositesa.2004.02.012.

[3] E. Borrows, I. Pillin, N. Montrelay, C. Baley, Could biopolymers reinforced by randomly scattered flax fibre be used in structural applications? *Composites Science and Technology*, 2007, **67**, 462-470, doi: 10.1016/j.compscitech.2006.08.024.

[4] F. Carrillo, X. Colom, X. Cañavate, Properties of regenerated cellulose lyocell fiber-reinforced composites, *Journal of Reinforced Plastics and Composites*, 2010, **29**, 359-371, doi: 10.1177/0731684408097777.

[5] B. Wang, S. Panigrahi, L. Tabil, W. Crerar, Pre-treatment of flax fibers for use in rotationally molded biocomposites, *Journal of Reinforced Plastics and Composites*, 2007, **26**, 447-463, doi: 10.1177/0731684406072526.

[6] K. van de Velde, P. Kiekens, Thermoplastic pultrusion of natural fibre reinforced composites, *Composite Structures*, 2001, **54**, 355-360, doi: 10.1016/s0263-8223(01)00110-6.

[7] L. Peponi, J. Biagiotti, L. Torre, J. M. Kenny, I. Mondragòn, Statistical analysis of the mechanical properties of natural fibers and their composite materials. I. Natural fibers, *Polymer Composites*, 2008, **29**, 313-320, doi: 10.1002/pc.20408.

[8] X. Li, L. G. Tabil, S. Panigrahi, Chemical treatments of natural fiber for use in natural fiber-reinforced composites: a review, *Journal of Polymers and the Environment*, 2007, **15**, 25-33, doi: 10.1007/s10924-006-0042-3.

[9] F. M. AL-Oqla, Y. A. El-Shekeil, Investigating and predicting the performance deteriorations and trends of polyurethane bio-composites for more realistic sustainable design possibilities, *Journal of Cleaner Production*, 2019, **222**, 865-870, doi: 10.1016/j.jclepro.2019.03.042.

[10] S. Mishra, A. K. Mohanty, L. T. Drzal, M. Misra, G. Hinrichsen, A review on pineapple leaf fibers, sisal fibers and their biocomposites, *Macromolecular Materials and Engineering*, 2004, **289**, 955-974, doi: 10.1002/mame.200400132.

[11] R. D. Campilho, Natural fiber composites, *CRC Press*, 2019.

[12] O. Faruk, A. K. Bledzki, H.-P. Fink, M. Sain, Biocomposites reinforced with natural fibers: 2000-2010, *Progress in Polymer Science*, 2012, **37**, 1552-1596, doi: 10.1016/j.progpolymsci.2012.04.003.

[13] E. Lokensgard, Industrial plastics: theory and applications. *Cengage Learning*, 2016.

[14] R. Sinha, Outlines of polymer technology: manufacture of polymers. Prentice-Hall India, *New Delhi*, 2002.

[15] T. Iijima, T. Tochimoto, M. Tomoi, Modification of epoxy resins with poly(aryl ether ketone)S, *Journal of Applied Polymer Science*, 1991, **43**, 1685-1692, doi: 10.1002/app.1991.070430911.

[16] G. Pritchard, Developments in reinforced plastics 4, *Springer Science & Business Media*, 2012.

[17] T. L. Sporleder, P. D. Goldsmith, J. Cordier, P. Godin, *International Food and Agribusiness Management Review*, 2011, **14**, 35-50.

[18] Z. Kuruppalil, Green Plastics: An Emerging Alternative for Petroleum Based Plastics? *International Journal of Engineering Research and Innovation*, 2011, **3**, 59-64.

[19] R. Kumar, L. Zhang, Water-induced hydrophobicity of soy protein materials containing 2, 2-diphenyl-2-hydroxyethanoic acid, *Biomacromolecules*, 2008, **9**, 2430-2437, doi: 10.1021/bm800396x.

[20] R. Kumar, N. Na, Soy protein films with the hydrophobic surface created through non-covalent interactions, *Industrial Crops and Products*, 2009, **29**, 485–494, doi: 10.1016/j.indcrop.2008.09.010.

[21] J. R. Newill, S. McKnight, C. P. Hoppel, G. Cooper, Effects of coatings on moisture absorption in composite materials, Army Research Lab Aberdeen Proving Ground MD, 1999.

[22] W. Doherty, P. Halley, L. Edye, D. Rogers, F. Cardona, Y. Park, T. Woo, Studies on polymers and composites from lignin and fiber derived from sugar cane, *Polymers for Advanced Technologies*, 2007, **18**, 673-678, doi: 10.1002/pat.879.

[23] K. O. Niska, M. Sain, Wood-polymer composites. *Elsevier*, 2008.

[24] S. Yildiz, E. D. Gezer, U. C. Yildiz, Mechanical and chemical behavior of spruce wood modified by heat, *Building and Environment*, 2006, **41**, 1762-1766, doi: 10.1016/j.buildenv.2005.07.017.

[25] B. van Voorn, H. H. G. Smit, R. J. Sinke, B. de Klerk, Natural

fibre reinforced sheet moulding compound, *Composites Part A: Applied Science and Manufacturing*, 2001, **32**, 1271-1279, doi: 10.1016/s1359-835x(01)00085-9.

[26] D. Rouison, M. Sain, M. Couturier, Resin transfer molding of natural fiber reinforced composites: cure simulation, *Composites Science and Technology*, 2004, **64**, 629-644, doi: 10.1016/j.compscitech.2003.06.001.

[27] A. P. Kumar, R. P. Singh, B. D. Sarwade, Degradability of composites, prepared from ethylene-propylene copolymer and jute fiber under accelerated aging and biotic environments, *Materials Chemistry and Physics*, 2005, **92**, 458-469, doi: 10.1016/j.matchemphys.2005.01.027.

[28] N. Chand, U. Dwivedi, Effect of coupling agent on abrasive wear behaviour of chopped jute fibre-reinforced polypropylene composites, *Wear*, 2006, **261**, 1057-1063, doi: 10.1016/j.wear.2006.01.039.

[29] D. Rouison, M. Sain, M. Couturier, Resin transfer molding of hemp fiber composites: optimization of the process and mechanical properties of the materials, *Composites Science and Technology*, 2006, **66**, 895-906, doi: 10.1016/j.compscitech.2005.07.040.

[30] P. A. Santos, M. A. S. Spinacé, K. K. G. Fermoselli, M.-A. de Paoli, Polyamide-6/vegetal fiber composite prepared by extrusion and injection molding, *Composites Part A: Applied Science and Manufacturing*, 2007, **38**, 2404-2411, doi: 10.1016/j.compositesa.2007.08.011.

[31] J. R. Araújo, W. R. Waldman, M. A. de Paoli, Thermal properties of high-density polyethylene composites with natural fibres: coupling agent effect, *Polymer Degradation and Stability*, 2008, **93**, 1770-1775, doi: 10.1016/j.polymdegradstab.2008.07.021.

[32] E. Corradini, E. N. Ito, J. M. Marconcini, C. T. Rios, J. A. M. Agnelli, L. H. C. Mattoso, Interfacial behavior of composites of recycled poly(ethyelene terephthalate) and sugarcane bagasse fiber, *Polymer Testing*, 2009, **28**, 183-187, doi: 10.1016/j.polymertesting.2008.11.014.

[33] D. R. Mulinari, H. J. C. Voorwald, M. Odila H. Cioffi, M. Lúcia C. P. da Silva, T. Gouvêada Cruz, Clodoaldo Saro, Sugarcane bagasse cellulose/HDPE composites obtained by extrusion, *Composites Science and Technology*, 2009, **69**, 214-219, doi: 10.1016/j.compscitech.2008.10.006.

[34] Y. Zou, S. Huda, Y. Yang, Lightweight composites from long wheat straw and polypropylene web, *Bioresource Technology*, 2010, **101**, 2026-2033, doi: 10.1016/j.biortech.2009.10.042.

[35] X. Peng, M. Fan, J. Hartley, M. Al-Zubaidy, Properties of natural fiber composites made by pultrusion process, *Journal of Composite Materials*, 2012, **46**, 237-246, doi: 10.1177/0021998311410474.

[36] M. Elkington, D. Bloom, C. Ward, A. Chatzimichali, K. Potter, Hand layup: understanding the manual process, *Advanced Manufacturing: Polymer & Composites Science*, 2015, **1**, 138-151, doi: 10.1080/20550340.2015.1114801.

[37] E. Ciupan, L. Lăzărescu, I. Filip, C. Ciupan, E. Câmpean, I. Cionca, E. Pop, Characterization of a thermoforming composite material made from hemp fibers and polypropylene, *MATEC Web of Conferences*, 2017, **137**, 08003, doi: 10.1051/matecconf/201713708003.

[38] A. Hodžić, R. Shanks, Natural fibre composites: materials, processes and properties, *Woodhead Publishing*, 2014.

[39] C. W. Lou, C. W. Lin, C. H. Huang, T. T. Li, J. M. Chen, J. H. Lin, Preliminary study of polypropylene/sawdust green composite, *Advanced Materials Research*, 2012, **557-559**, 334-337, doi: 10.4028/www.scientific.net/amr.557-559.334.

[40] A. K. Mohanty, P. Tummala, W. Liu, M. Misra, P. V. Mulukutla, L. T. Drzal, Injection molded biocomposites from soy protein-based bioplastic and short industrial hemp fiber, *Journal of Polymers and the Environment*, 2005, **13**, 279-285, doi: 10.1007/s10924-005-4762-6.

[41] K. Ohkita, H. Takagi, Study on fracture behaviors of injection-molded bamboo fiber/PBS composites, *Key Engineering Materials*, 2010, **452-453**, 229-232, doi: 10.4028/www.scientific.net/kem.452-453.229.

[42] M. T. Hayajneh, F. M. AL-Oqla, M. M. Al-Shrida, Hybrid green organic/inorganic filler polypropylene composites: morphological study and mechanical performance investigations, *e-Polymers*, 2021, **21**, 710-721, doi: 10.1515/epoly-2021-0074.

[43] J. P. Mofokeng, A. S. Luyt, T. Tábi, J. Kovács, Comparison of injection moulded, natural fibre-reinforced composites with PP and PLA as matrices, *Journal of Thermoplastic Composite Materials*, 2012, **25**, 927-948, doi: 10.1177/0892705711423291.

[44] P. Klein, Fundamentals of plastics thermoforming, *Synthesis Lectures on Materials Engineering*, 2009, **1**, 1-97, doi: 10.2200/s00184ed1v01y200904mre001.

[45] L.-T. Lim, R. Auras, M. Rubino, Processing technologies for poly(lactic acid), *Progress in Polymer Science*, 2008, **33**, 820-852, doi: 10.1016/j.progpolymsci.2008.05.004.

[46] G. Sebe, N. S. Cetin, C. A. Hill, M. Hughes, RTM Hemp Fibre-Reinforced Polyester Composites, *Applied Composite Materials*, 2000, **7**, 341-349, doi: 10.1023/A:1026538107200.

[47] K. Liu, X. Zhang, H. Takagi, Z. Yang, D. Wang, Effect of chemical treatments on transverse thermal conductivity of unidirectional abaca fiber/epoxy composite, *Composites Part A: Applied Science and Manufacturing*, 2014, **66**, 227-236, doi: 10.1016/j.compositesa.2014.07.018.

[48] C. Kong, H. Park, J. Lee, Study on structural design and analysis of flax natural fiber composite tank manufactured by vacuum assisted resin transfer molding, *Materials Letters*, 2014, **130**, 21-25, doi: 10.1016/j.matlet.2014.05.042.

[49] F. M. AL-Oqla, S. M. Sapuan, M. R. Ishak, A. A. Nuraini, A novel evaluation tool for enhancing the selection of natural fibers for polymeric composites based on fiber moisture content criterion, *BioResources*, 2014, **10**, 299-312, doi: 10.15376/biores.10.1.299-312.

[50] M. Le Troedec, D. Sedan, C. Peyratout, J. P. Bonnet, A. Smith, R. Guinebretiere, V. Gloaguen, P. Krausz, Influence of various chemical treatments on the composition and structure of hemp fibres, *Composites Part A: Applied Science and Manufacturing*, 2008, **39**, 514-522, doi: 10.1016/j.compositesa.2007.12.001.

[51] S. M. Ansari, C. M. R. Ghazali, K. Husin, in MATEC Web

of Conferences, *EDP Sciences*, 2017, **97**, 01018, doi: 10.1051/matecconf/20179701018.

[52] V. Tserki, C. Panayiotou, N. E. Zafeiropoulos, A study of the effect of acetylation and propionylation on the interface of natural fibre biodegradable composites, *Advanced Composites Letters*, 2005, **14**, 096369350501400, doi: 10.1177/096369350501400202.

[53] A. Alawar, A. M. Hamed, K. Al-Kaabi, Characterization of treated date palm tree fiber as composite reinforcement, *Composites Part B: Engineering*, 2009, **40**, 601-606, doi: 10.1016/j.compositesb.2009.04.018.

[54] X. Y. Liu, G. C. Dai, Surface modification and micromechanical properties of jute fiber mat reinforced polypropylene composites, *Express Polymer Letters*, 2007, **1**, 299-307, doi: 10.3144/expresspolymlett.2007.43.

[55] Y. A. El-Shekeil, S. M. Sapuan, A. Khalina, E. S. Zainudin, O. M. Al-Shuja'a, Effect of alkali treatment on mechanical and thermal properties of Kenaf fiber-reinforced thermoplastic polyurethane composite, *Journal of Thermal Analysis and Calorimetry*, 2012, **109**, 1435-1443, doi: 10.1007/s10973-012-2258-x.

[56] J. Jayaramudu, B. Guduri, A. V. Rajulu, Characterization of Natural Fabric Sterculia urens, International *Journal of Polymer Analysis and Characterization*, 2009, **14**, 115-125, doi: 10.1080/10236660802601415.

[57] K. O. Reddy, C. U. Maheswari, D. J. P. Reddy, B. Guduri, A. V. Rajulu, Properties of ligno-cellulose ficus religiosa leaf fibers, 2010.

[58] Y. Liu, J. Xie, N. Wu, Y. Ma, C. Menon, J. Tong, Characterization of natural cellulose fiber from corn stalk waste subjected to different surface treatments, *Cellulose*, 2019, **26**, 4707-4719, doi: 10.1007/s10570-019-02429-6.

[59] S. Thomas, R. Thomas, A. K. Zachariah, R. Kumar, Thermal and rheological measurement techniques for nanomaterials characterization, *Elsevier*, 2017.

[60] P. H. F. Pereira, H. J. C. Voorwald, M. O. H. Cioffi, M. L. C. P. da Silva, Novel cellulose/$NbOPO_{4.n}H_2O$ hybrid material from sugarcane bagasse, *BioResources*, 2011, **6**, 867-878, doi: 10.15376/biores.6.1.867-878.

[61] J. L. Guimarães, E. Frollini, C. G. da Silva, F. Wypych, K. G. Satyanarayana, Characterization of banana, sugarcane bagasse and sponge gourd fibers of Brazil, *Industrial Crops and Products*, 2009, **30**, 407-415, doi: 10.1016/j.indcrop.2009.07.013.

[62] Y. Li, G. Li, Y. Zou, Q. Zhou, X. Lian, Preparation and characterization of cellulose nanofibers from partly mercerized cotton by mixed acid hydrolysis, *Cellulose*, 2014, **21**, 301-309, doi: 10.1007/s10570-013-0146-6.

[63] S. H. Aziz, M. P. Ansell, The effect of alkalization and fibre alignment on the mechanical and thermal properties of kenaf and hemp bast fibre composites: part 1 - polyester resin matrix, *Composites Science and Technology*, 2004, **64**, 1219-1230, doi: 10.1016/j.compscitech.2003.10.001.

[64] A. Béakou, R. Ntenga, J. Lepetit, J. A. Atéba, L. O. Ayina, Physico-chemical and microstructural characterization of "Rhectophyllum camerunense" plant fiber, *Composites Part A: Applied Science and Manufacturing*, 2008, **39**, 67-74, doi: 10.1016/j.compositesa.2007.09.002.

[65] D. Liu, G. Han, J. Huang, Y. Zhang, Composition and structure study of natural Nelumbo nucifera fiber, *Carbohydrate Polymers*, 2009, **75**, 39-43, doi: 10.1016/j.carbpol.2008.06.003.

[66] R. G. Elenga, G. F. Dirras, J. Goma Maniongui, P. Djemia, M. P. Biget, On the microstructure and physical properties of untreated raffia textilis fiber, *Composites Part A: Applied Science and Manufacturing*, 2009, **40**, 418-422, doi: 10.1016/j.compositesa.2009.01.001.

[67] M. Jacob, S. Thomas, K. T. Varughese, Mechanical properties of sisal/oil palm hybrid fiber reinforced natural rubber composites, *Composites Science and Technology*, 2004, **64**, 955-965, doi: 10.1016/s0266-3538(03)00261-6.

[68] C.-Y. Hsieh, S.-L. Chung, High thermal conductivity epoxy molding compound filled with a combustion synthesized AlN powder, *Journal of Applied Polymer Science*, 2006, **102**, 4734-4740, doi: 10.1002/app.25000.

[69] F. de Andrade Silva, N. Chawla, R. D. de Toledo Filho, Tensile behavior of high performance natural (sisal) fibers, *Composites Science and Technology*, 2008, **68**, 3438-3443, doi: 10.1016/j.compscitech.2008.10.001.

[70] E. Balnois, F. Busnel, C. Baley, Y. Grohens, An AFM study of the effect of chemical treatments on the surface microstructure and adhesion properties of flax fibres, *Composite Interfaces*, 2007, **14**, 715-731, doi: 10.1163/156855407782106537.

[71] I. K. Varma, S. R. A. Krishnan, S. Krishnamoorthy, Effect of chemical treatment on density and crystallinity of jute fibers, *Textile Research Journal*, 1989, **59**, 368-370, doi: 10.1177/004051758905900609.

[72] T. P. Sathishkumar, P. Navaneethakrishnan, S. Shankar, R. Rajasekar, Characterization of new cellulose sansevieria ehrenbergii fibers for polymer composites, *Composite Interfaces*, 2013, **20**, 575-593, doi: 10.1080/15685543.2013.816652.

[73] P. Manimaran, P. Senthamaraikannan, K. Murugananthan, M. R. Sanjay, Physicochemical properties of new cellulosic fibers from Azadirachta indica plant, *Journal of Natural Fibers*, 2018, **15**, 29-38, doi: 10.1080/15440478.2017.1302388.

[74] H.-S. Kim, H.-S. Yang, H.-J. Kim, Biodegradability and mechanical properties of agro-flour-filled polybutylene succinate biocomposites, *Journal of Applied Polymer Science*, 2005, **97**, 1513-1521, doi: 10.1002/app.21905.

[75] K. M. M. Rao, K. M. Rao, Extraction and tensile properties of natural fibers: Vakka, date and bamboo, *Composite Structures*, 2007, **77**, 288-295, doi: 10.1016/j.compstruct.2005.07.023.

[76] M. Truong, W. Zhong, S. Boyko, M. Alcock, A comparative study on natural fibre density measurement, *Journal of the Textile Institute*, 2009, **100**, 525-529, doi: 10.1080/00405000801997595.

[77] Y. Liu, H. Hu, X-ray diffraction study of bamboo fibers treated with NaOH, *Fibers and Polymers*, 2008, **9**, 735-739, doi: 10.1007/s12221-008-0115-0.

[78] K. M. Mannan, X-ray diffraction study of jute fibres treated with NaOH and liquid anhydrous ammonia, *Polymer*, 1993, **34**, 2485-2487, doi: 10.1016/0032-3861(93)90576-v.

[79] M. Poletto, A. J. Zattera, M. M. C. Forte, R. M. C. Santana,

Thermal decomposition of wood: influence of wood components and cellulose crystallite size, *Bioresource Technology*, 2012, **109**, 148-153, doi: 10.1016/j.biortech.2011.11.122.

[80] L. Segal, J. J. Creely, A. E. Martin Jr, C. M. Conrad, An empirical method for estimating the degree of crystallinity of native cellulose using the X-ray diffractometer, *Textile Research Journal*, 1959, **29**, 786-794, doi: 10.1177/004051755902901003.

[81] Y. Seki, M. Sarikanat, K. Sever, C. Durmuşkahya, Extraction and properties of Ferula communis (chakshir) fibers as novel reinforcement for composites materials, *Composites Part B: Engineering*, 2013, **44**, 517-523, doi: 10.1016/j.compositesb.2012.03.013.

[82] V. P. Arthanarieswaran, A. Kumaravel, M. Kathirselvam, S. S. Saravanakumar, Mechanical and thermal properties of Acacia leucophloeafiber/epoxy composites: influence of fiber loading and alkali treatment, *International Journal of Polymer Analysis and Characterization*, 2016, **21**, 571-583, doi: 10.1080/1023666x.2016.1183279.

[83] Z.-G. Song, F.-M. Li, Aerothermoelastic analysis of nonlinear composite laminated panel with aerodynamic heating in hypersonic flow, *Composites Part B: Engineering*, 2014, **56**, 830-839, doi: 10.1016/j.compositesb.2013.09.019.

[84] K. O. Reddy, J. Zhang, J. Zhang, A. V. Rajulu, Preparation and properties of self-reinforced cellulose composite films from Agave microfibrils using an ionic liquid, *Carbohydrate Polymers*, 2014, **114**, 537-545, doi: 10.1016/j.carbpol.2014.08.054.

[85] A. V. Rajulu, L. G. Devi, G. B. Rao, R. L. Reddy, Chemical resistance and tensile properties of epoxy/unsaturated polyester blend coated bamboo fibers, *Journal of Reinforced Plastics and Composites*, 2003, **22**, 1029-1034, doi: 10.1177/0731684403024571.

[86] V. S. Sreenivasan, S. Somasundaram, D. Ravindran, V. Manikandan, R. Narayanasamy, Microstructural, physico-chemical and mechanical characterisation of Sansevieria cylindrica fibres - An exploratory investigation, *Materials & Design*, 2011, **32**, 453-461, doi: 10.1016/j.matdes.2010.06.004.

[87] H. L. Ornaghi Jr, M. Poletto, A. J. Zattera, S. C. Amico, Correlation of the thermal stability and the decomposition kinetics of six different vegetal fibers, *Cellulose*, 2014, **21**, 177-188, doi: 10.1007/s10570-013-0094-1.