Physicochemical and Structural Investigation of Argeli (*Edgeworthia gardneri*) Bast Fibers

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Abstract: The structure and some physicochemical properties of Argeli *(Edgeworthia gardneri)* bast fibers were investigated using Fourier Transform Infrared (FTIR) and Energy Dispersive X-ray (EDX) spectroscopies, Optical Microscopy (OM) and Scanning Electron Microscopy (SEM). The neat fibers were found to contain about 54.47% cellulose, 25.98% hemicellulose, 10.5 % lignin, 6.1% extractives, and about 2.9% ash. The fibers on chemical treatments changed several properties, some of them being quite significant. Fiber density was increased by 8.5% in the alkali-treated samples which may be due to the loss of less dense components such as lignin and hemicelluloses. The tensile strength of the fiber increased by 34 % and 61 %, respectively, after alkali and bleaching treatments. However, the thermal properties of treated samples did not change significantly. The mechanical properties of Argeli fiber were improved on chemical treatments making them attractive in the fabrication of polymer composites, textiles, and papers.

Keywords: Argeli fibers, Physicochemical properties, Structure, Thermal stability, Water absorption.

1. INTRODUCTION

Lignocellulosic fibers obtained from various plants are being explored as alternatives to traditionally used synthetic fibers. Flax, hemp, sisal, bamboo, cotton, jute, kenaf, pineapple, and ramie are just a few representative sources of lignocellulosic fibers in this regard [1,2]. These fibers are extracted from various parts of the plant such as bast, leaf, seed, stem, root, grass, and wood through chemical and mechanical processing [3]. The plant-based natural fibers, indeed, offer unique properties compared to synthetic fibers [4-6], in their nominal cost, satisfactory mechanical properties, renewability, biodegradability, non-abrasive behavior, and good thermal, electrical, and acoustic insulation properties being few of their advantageous features [7,8]. Due to these advantages, natural fibers are being extensively explored as an alternative material in several industrial sectors such as in paper, textiles, automotive, and construction industries.

Plant-based natural fibers are complex structural composites comprising cellulose, hemicellulose, lignin, wax, and pectin [9] in which cellulose microfibrils being embedded in an amorphous pool of lignin and hemicellulose matrix act as reinforcing filler [10]. The major constituents of natural fibers are mainly found in the primary cell walls of plants in different proportions and behave differently.

Fiber quality depends on chemical composition, microstructure, and processing techniques. It is known that the size of the microfibrils, their angles of orientation, the degree of crystallinity, and the chemical composition are other key parameters that determine the overall quality of the fiber bundle [11,12]. Each fiber bundle consists of a bunch of smaller structural units called fibrils and is strongly held by pectinous gum. Lignin and pectin are strongly bound to cellulose microfibrils and water-insoluble hemicelluloses engage the space between the fibrils [13]. Alkali treatment also called the *Mercerization Process*, eliminates the gluing components such as lignin and hemicellulose from the fiber, causing partial or complete dissociation of fiber bundles, their decreased width, and increased aspect ratio [14]. Mercerization, bleaching, silane treatment, benzylation, and acetylation are some chemical

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methods used to modify the fiber surface, which improves surface roughness, moisture absorption, and promotes adhesion [15].

Alkali treatment followed by bleaching is the most commonly used method to eliminate non-cellulose material and separate cellulose [16]. For the intended uses, such as papers, cardboard, and composites, this makes pulping simpler. Under optimal processing conditions, ultimate fiber qualities (such as thermal stability, mechanical strength, and water absorption capabilities) improved significantly.

Natural fibers derived from different plant species are examined by different researchers throughout the globe [16-19]. The majority of the studies are focused on their extraction and physical, chemical, mechanical, and water absorption properties. Lignocellulosic fibers are explored to fabricate lightweight, economical, and eco-friendly engineering materials. In this regard, the Nepalese paper bush, *Edgeworthia gardneri* (locally called Argeli) is one of the least-studied plant species of the *Thymelaeaceae* family. It grows as a deciduous evergreen shrub, 3-4 m tall, and native to the hilly region 1000 – 2500 m above sea level in Asian countries such as Nepal, China, India, and Bhutan and produces fibers from brownish-red stem bark. The present work aims at investigating different physicochemical, morphological, thermal, and mechanical properties of the Argeli bast fibers as a function of processing conditions.

2. MATERIALS AND METHODS

2.1. Materials

The raw materials were collected from Bhumika Handmade Paper, Udhyog Gokarneshwor-8, Kathmandu, Nepal. Acetic acid, sodium chlorite, sulfuric acid, acetone, and sodium hydroxide were purchased from Carl Roth GmbH, Germany, and used without further purification.

2.2. Fiber Extraction and Chemical Treatment

The sun-dried neat Argeli bast strip was chopped into about 5 cm long pieces, and immersed in a bucket with tap water for 10 days followed by a thorough cleansing with tap water to remove dirt and soluble impurities and subjected to oven-dry for 24 h at 105 °C. The extracted and dried fibers were ground using Retsch SK 100 and sieved through a 0.25 mm mesh. The dry powder was sealed in a plastic bag and stored in a dark place at 21 °C for further analysis.

The required quantity of dry powder was immersed in 5% NaOH solution for 4 h at 21 °C with a 'fiber weight to alkali solution' ratio of 1:25. Afterwards, to eliminate the excess alkali solution, acetic acid was added drop-wise. The fibers were washed thoroughly with distilled water until the pH of the filtrate reached a value of 7. The residue was dried for 24 h at 80 °C. Further, to remove the residual lignin, the alkali-treated dried fiber was immersed in 2% $NaClO_2$ solution for 2 h at pH of 4. The fibers were finally washed thoroughly with distilled water and dried for another 24 h in a hot air oven at 80 °C. Table **1** presents the overview of the samples studied in the present work.

Table 1: List of the Natural Fiber Samples used in this Work

S. No.	Sample code	Description
1	nAF	Neat Argeli Fiber obtained from the bast
2	tAF	5 % (w/v) NaOH-treated nAF
3	bAF	2% (w/v) $NaClO_2$ bleached tAF

2.3. Chemical Composition Analysis

The extractives, cellulose, hemicelluloses, lignin, and ash content of Argeli fiber were estimated gravimetrically using the methods described in the literature. Every experiment was conducted three times, and an average of the results were reported. The percentage composition was measured using eq. (i).

$$Composition\ (\%) = \frac{X}{Y} \times 100 \quad \text{(i)}$$

where, X = Final weight of the sample in grams, and Y = Initial weight of the sample in grams

Determination of Ash Content

The clean and empty crucible was heated to 575 °C in a muffle furnace for 30 min and cooled in a desiccator. A precision analytical balance was used to measure the weight of the samples. An electric muffle furnace was used to heat 1 g of the test specimen for 3 h at 575 °C. The crucible was then taken out of the furnace, and cooled in desiccators and the weight of the ash [20].

Determination of Hemicelluloses, Lignin, Cellulose, and Extractives

The quantification of the major ingredients of the natural fibers, namely hemicellulose, lignin, and cellulose, was performed.

For the determination of *hemicelluloses*, 1 g of oven-dried and extractives-free biomass was heated with 10 mL of 2%NaOH solution at 80 °C for 3.5 h. The residue was washed thoroughly with sufficient distilled water until the pH of the solution became 7. Then, it was dried at 105 °C until a constant weight was obtained. Hemicellulose content was determined by comparing the sample weight before and after the treatment [21].

For *lignin* determination, 3 mL of 72% H_2SO_4 was added into 0.3 g of dried, extract-free biomass and left at room temperature for 2 h. The mixture was carefully shaken every 30 minutes to ensure complete hydrolysis and 84 mL of distilled water was added. Finally, the slurry was hydrolyzed in an autoclave for 1 h at 121°C and allowed to settle at room temperature before being filtered. The amount of acid-insoluble lignin was determined after drying the residues at 105 °C [22].

The amount of *cellulose* was determined using the difference method, presuming that only elements of the whole biomass comprise extractives, ash, hemicellulose, lignin, and cellulose [20,23].

For *extractive* determination, 1 g of dried biomass was refluxed with 60 mL of acetone at 90 °C for 2 h. The biomass fiber was then dried at 105 °C until a constant weight was recorded. The amount of the extractives was calculated by contrasting the weights before and after extraction [21].

2.4. Physical Properties Characterization

Moisture Content

To determine the moisture content of fiber at ambient temperature, 5 g of each sample was conditioned in a moisture chamber at 23°C and 65% relative humidity (RH) for 24 h before being dried for 3 h at 105 °C in an oven. The fibers were weighed after being dried, cooled, and desiccated. The degree of wetness of the fiber was determined by comparing the weight of the fibers before and after drying [24]. Three replicates were used to determine the moisture content of the fiber.

Density

The density of the sample was measured gravimetrically at 23 °C using canola oil (with a density of 0.9135 g/cm^3) as the submersion liquid with the help of Mettler Toledo XSR225DU analytical balance. The average apparent density was recorded from five independent measurements.

Fiber Length and Diameter

The diameter of 50 fiber bundles from the images recorded using a KEYENCE VHX-950F microscope, assuming the fibers have cylindrical geometry. The length was measured by hand stapling method according to ISO 6989 standard and average fiber length was calculated as an average value of 20 different fibers.

Determination of Water Absorption

The water absorption properties of the sample were determined using a water absorption test. About 1 g of each sample was dried at 105 °C for 8 h. It was then cooled in desiccators and immersed in distilled water at room temperature. The specimens were taken out of the water and wiped with tissue paper. The weight gain of each specimen was measured at different time intervals. Three independent experiments were used to express the percentage of average water absorption (WA), eq. (ii).

$$WA = \frac{W2 - W1}{W1} \times 100 \quad \text{(ii)}$$

where, W1= initial weight of fiber, W2= final weight of fiber.

2.5. Structural, Thermal, and Mechanical Analyses

Optical Microscopy (OM) and Scanning Electron Microscopy *(SEM)*

A VHX-950F optical microscope was used to measure the microscopic structure of the samples. The surface morphology of each specimen was investigated using a JEOL JSM-6360 SEM operated at an accelerating voltage of 15 kV. The fibrous specimen was attached to the SEM specimen holder using a doubly carbon-coated tape followed by coating the sample with a thin conducting carbon film.

Fourier Transform Infrared (FTIR) Spectroscopy

FTIR spectra of the fibers were recorded using Tensor 27 Bruker Optics GmbH & Co. KG (Billerica, USA) in attenuated total reflection (ATR) mode between the wave numbers 4000 cm^{-1} and 400 cm^{-1} with a resolution of 4 cm^{-1} and 16 scans per sample.

Thermogravimetric Analysis (TGA)

The thermal stability of Argeli fiber samples was analyzed by using a NETZSCH TG209 F3 Tarsus (NETZSCH-Gerätebau GmbH, Selb, Germany) analyzer at a heating rate of 10 K/min over the

temperature range of 40 °C to 700 °C under nitrogen atmosphere with the gas flow rate of 40 mL/min.

Mechanical Testing

The tensile tests of the fibers were performed using a Universal Testing Machine (Zwick Roell GmbH & Co. KG, Ulm, Germany) following the ISO 527 standard with a gauge length of 20 mm and a crosshead speed of 2 mm/min at 23 °C. For this purpose, a bundle of definite mass of each fiber type was firmly fixed to the clamps of the tensile machine. The mechanical parameters, including tensile strength (the maximum attainable strength), elongation at break (the maximum strain attained at fracture of the specimen), and tensile modulus (calculated by the slope of the initial part of the stress-strain curves), were assessed. Five experiments were carried out for each sample type and the average value was calculated.

3. RESULTS AND DISCUSSION

3.1. Physicochemical Properties of Neat Argeli Fibers

The experimental results have demonstrated that the Argeli fibers comprise cellulose, hemicelluloses, lignin, extractives, and ash content which were found to be approximately 54.47, 25.98, 10.5, 6.1, and 2.9 wt.-%, respectively. The results are compared with the literature values [25-29] for some commonly studied natural fibers and presented in Table **2**.

Notably, like several other natural fibers reported in the literature, the Argeli fiber is a good source of cellulose content (~ 55 wt.-%) which is comparable to other fibers such as flax, hemp, kenaf, and jute. It can be deduced also based on the works reported in the literature that the cellulosic framework of Argeli fiber contributes to both its strength and durability [30]. The highly hydrophilic character of neat Argeli fiber could be due to the presence of a high amount of hemicellulose. It can be observed in Table **2** that the Argeli fiber typically has higher lignin content compared to kenaf, flax, and cotton fibers. Higher lignin content can impart them with lower flexibility or enhanced stiffness [20]. However, the chemical composition of fiber and hence their application windows may also depend upon the plant species, methods of extraction, and parts of the plants from which the fibers are extracted [9].

Some of the important physical properties of the Argeli fibers are presented in Table **3** comparing them with the corresponding values reported in the literature for different natural fibers [19,25,31].

The average diameter, density, and length of the neat Argeli fibers were found to be 13.57 µm, 1.28 g/cm^3, and 10.87 mm; respectively.

It can be noted that both the diameter and the density of the Argeli fibers are found to be lower than that of most of the fibers indexed in Table **3**. The former observation can be associated with its particular organization of the cellulose types [32] which, however, needs more investigations for confirmation. The lower density in Argeli fibers could be attributed to the presence of low-density non-cellulosic components in their microscopic composite framework [33].

3.2. Effect of Chemical Treatment on Properties of Fibers

Another important property of the Argeli fiber is moisture content and water absorption (see Table **4**). The higher moisture content and water absorption of Argeli fiber could be related to the porous structure of the fiber bundles (see Figure **4**, in the next section), the

Table 2: Comparison of Percentage Composition of Argeli Fiber with that of other Natural Fibers

Fiber Type	Cellulose (wt.-%)	Hemicellulose (wt.-%)	Lignin (wt.-%)	References
Argeli	54.47 ± 2.29	25.98 ± 1.15	10.5 ± 0.78	Current study
Lokta	36.8	44	14.3	[25]
Flax	64-71	18.6-20.6	2-5	[26]
Cotton	85-90	1-3	7-16	[26]
Jute	61-71.5	12-13	13.6 -20.4	[27]
Hemp	70.2-74.4	3.7-5.7	17.9 -22.4	[27]
Bamboo	46.91	17.29	21.30	[28]
Kenaf	66.47	9.43	2.39	[29]

Table 3: Some Physical Properties of Neat Argeli Fiber Compared to other Reported Natural Fibers

Fiber Type	Density (g/cm^3)	Diameter (µm)	Moisture content (%)	Water absorption (%)	References
Argeli	1.28± 0.01	13.57±1.25	10.9 ± 0.62	195.33± 4.04	Current study
Flax	1.52	16.66	7.63	132.4	[19]
Cotton	1.55	14.5	8.59	-	[31]
Jute	1.4	18.4	17.0	-	[31]
Lokta	1.1	-	13.3	506.2	[25]
Kenaf	1.3	19.8	17.0	-	[31]

Table 4: Effect of Chemical Treatment on some Physical Properties of the Argeli Fiber

Sample Code	Density (g/cm^3)	Diameter (µm)	Weight loss (%)	Water Absorption (%)
nAF	1.28 ± 0.01	13.57 ± 1.25	-	196
tAF	1.39 ± 0.03	11.19 ± 1.05	17.08	258
bAF	1.27 ± 0.01	10.78± 1.12	10.96	293

Note: The water absorption percentage reported here was for 12 h.

higher concentration of hydrophilic components hemicelluloses, and extractives. These findings conclude that fibers with high moisture content have low density and are ideal for the development of lightweight bio-based products. Chemical treatment changes the composition and properties of the fibers [34], as will be demonstrated also in the current study. The effect of chemical treatment on some physical properties of Argeli fibers is presented in Table **4**. It should be noted that the weight loss in the fibers after each stage of chemical treatment correlates with the removal of the soluble matter.

By the alkali treatment, the fiber lost its weight by 17.08% causing a decrease in average fiber diameter by ~8.5% (from 13.57 µm to 11.19 µm) as a result of the removal of non-cellulosic, amorphous material from the fiber bundle. The result is consistent with the literature [35]. The increase in apparent density of the fiber accompanied by highly enhanced water absorption can be attributed to the alkali treatment-induced removal of low-density non-cellulosic components. Both the kinds of effects discussed above were further magnified by bleaching the alkali-treated Argeli fibers. Hashim *et al*. [35] reported similar results that the density of kenaf fiber increased with the decrease in fiber diameter due to the alkali treatments.

The overall effect of the treatment of the fibers on their physical properties can be exemplified by a plot of weight gain by allowing the fibers to soak in water for varied times. The quantities measured can be further utilized to estimate the biodegradation behavior of the fibers and composites thereof [36]. The results of the tests are presented in Figure **1**.

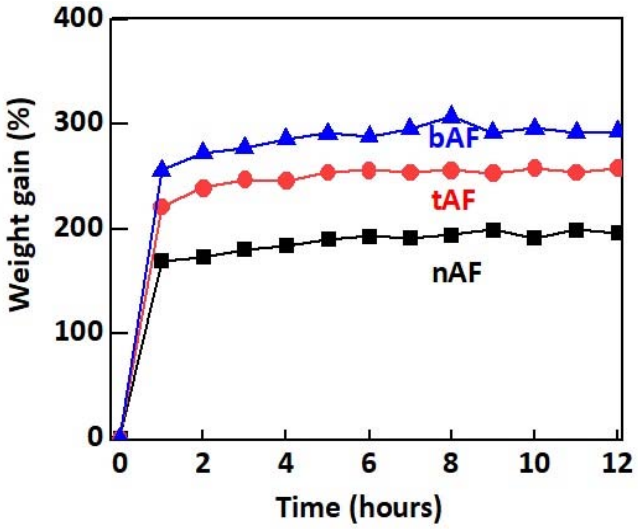

Figure 1: Comparison of water absorption behavior of treated (tAF) and bleached (bAF) Argeli fibers with the pristine neat (nAF) fibers.

When Argeli fibers were immersed in water for a specific time, the weight of the fiber rapidly increased up to a certain limit (more than 190%) and then leveled off. It was found that on chemically treating the fibers, the water absorption capacity increased significantly. For instance, the water absorption after 6 h increased by ~ 32% in alkali-treated fibers as compared to

untreated ones, and on further bleaching, the fibers could hold more than 50% more water relative to the neat fibers. It would been interesting to weigh the samples at shorter time intervals for the first few hours and record the trend of water uptake to precisely define when the levelling off of the curves takes place. We leave this experimental procedure for future work.

The enhanced water absorption properties of treated fibers can be attributed to the loss of adhesive coating materials (such as oil and wax) present in the fiber bundle allowing the fiber to be penetrated by water molecules. This result is in agreement with the water absorption properties of alkali-treated kenaf fibers reported by Fook and Yatim [18]. Further, the surface texture of the fibers was found to be smoothened with clearly distinguished microstructures on alkali treatment resulting from losing lignin, wax, pectin, hemicellulose, and other impurities that covered the external fiber surfaces [37]. In this process, it has been suggested that the existing H- bonds break down, and a new H- bond is formed between water molecules and the hydroxyl group of the fiber. Such interplays in chemical bonding observed in cellulosic frameworks are strongly affected by the presence of impurities [38].

The results presented in Figure **1** further show that alkali-treated and bleached fibers absorbed more water as compared to neat ones as the treatment removed the amorphous, non-cellulosic impurities from the fibers' surface leaving behind the crystalline cellulosic compounds thus decreasing their diameter [17,36,39].

Further, the sodium chlorite bleached fiber shows the highest water absorption, which could be due to the elimination of residual amorphous lignin from the fiber surface. The removal of excessive amorphous constituents during treatment may increase fiber roughness, cellulose content, pores, and voids. There are several advantages of the increased purity of the fiber and roughness including the increase in the water absorption capacity thereby increasing the ease of biodegradation [13]. Thus, the extent of water absorption of the bleached fiber increased with the change in surface roughness and morphology. It should be noted, however, that alkali and bleaching treatments may also make it more hydrophilic, which could be sometimes unfavorable (for applications with hydrophobic matrices, for instance). Excessive treatments may even damage the crystalline texture of cellulose, introducing defects that could potentially lead to decreased mechanical properties.

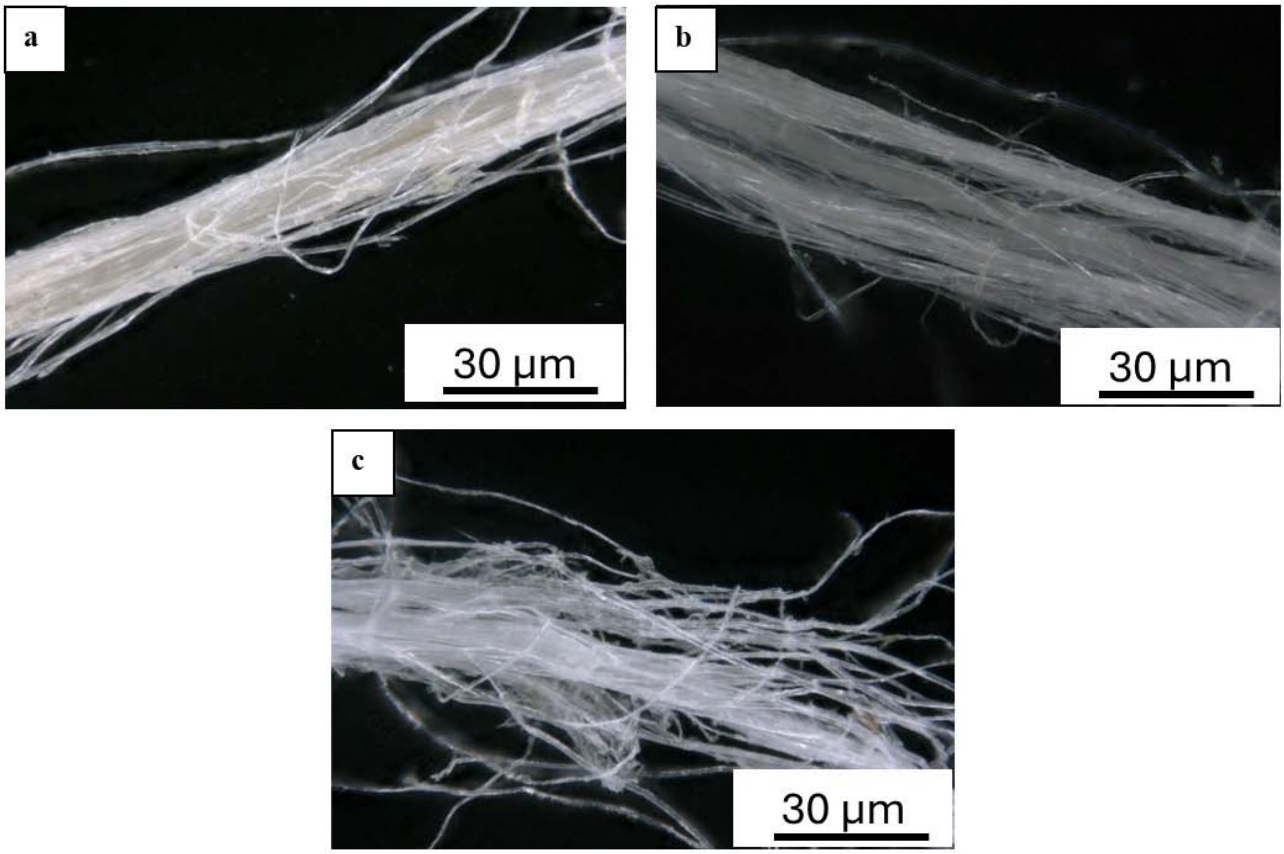

Figure 2: Optical micrographs of Argeli fibers depicting their typical morphologies: **a**) nAF, **b**) tAF, and **c**) bAF.

3.3. Effect of Chemical Treatment on Structure and Morphology

The structural features of the fibers were investigated by OM, SEM, and FTIR spectroscopy. The optical microscopic view of Argeli fibers, as presented in Figure **2**, shows that long, slender, and cylindrical threads are longitudinally aligned in a bundle.

Each bundle consists of several individual elementary fibers that are bonded together by natural adhesive materials [40]. The experimental results so far demonstrate a significant alteration of the surface morphology with a decrease in the fiber diameter. Similarly, on bleaching with sodium chlorite, the fibers become more distinct and decrease in diameter. The results showed that chemical treatment might have dissolved the compound that coated the outer surface of the fiber and reduced the inter-fiber bonding making it easier to separate them from the bundle [41]. The decrease in the diameter of the fibers on both treatments was also observed in the physicochemical studies discussed in previous sections.

The OM provided a quick overview of the fiber morphology, their diameter, and length as well as their orientation in bundles. However, the detailed morphology of the fibers cannot be ascertained. Result. Thus, to shed more light on the detailed morphological features of the fibers as a function of the chemical treatments, the samples were further investigated by SEM, see Figure **3**. The SEM images show that the neat Argeli fiber (see Figure **3a**) has smooth surface morphology, which could be ideal for the development of durable paper and comfortable fabrics [42]. The smooth surface morphology of the Argeli fiber is attributed to its cellular structure and composition. The uniform and dense arrangement of cells with minimal protrusions of irregularities on their surface could be the reason for the smooth surface. It should be noted that neat fiber bundles generally possess large amounts of non-uniform fibrils along with a considerable amount of non-fibrillar regions. The detailed discussion will be the object of a separate future paper.

After alkali treatment, the SEM images of the fiber surface appear porous and rougher (see Figure **3b**). On alkali treatment, the density of the fiber increased from 1.28 to 1.37 g/cm^3 while the diameter of the fiber decreased and surface roughness increased due to the loss of less dense materials as discussed in the

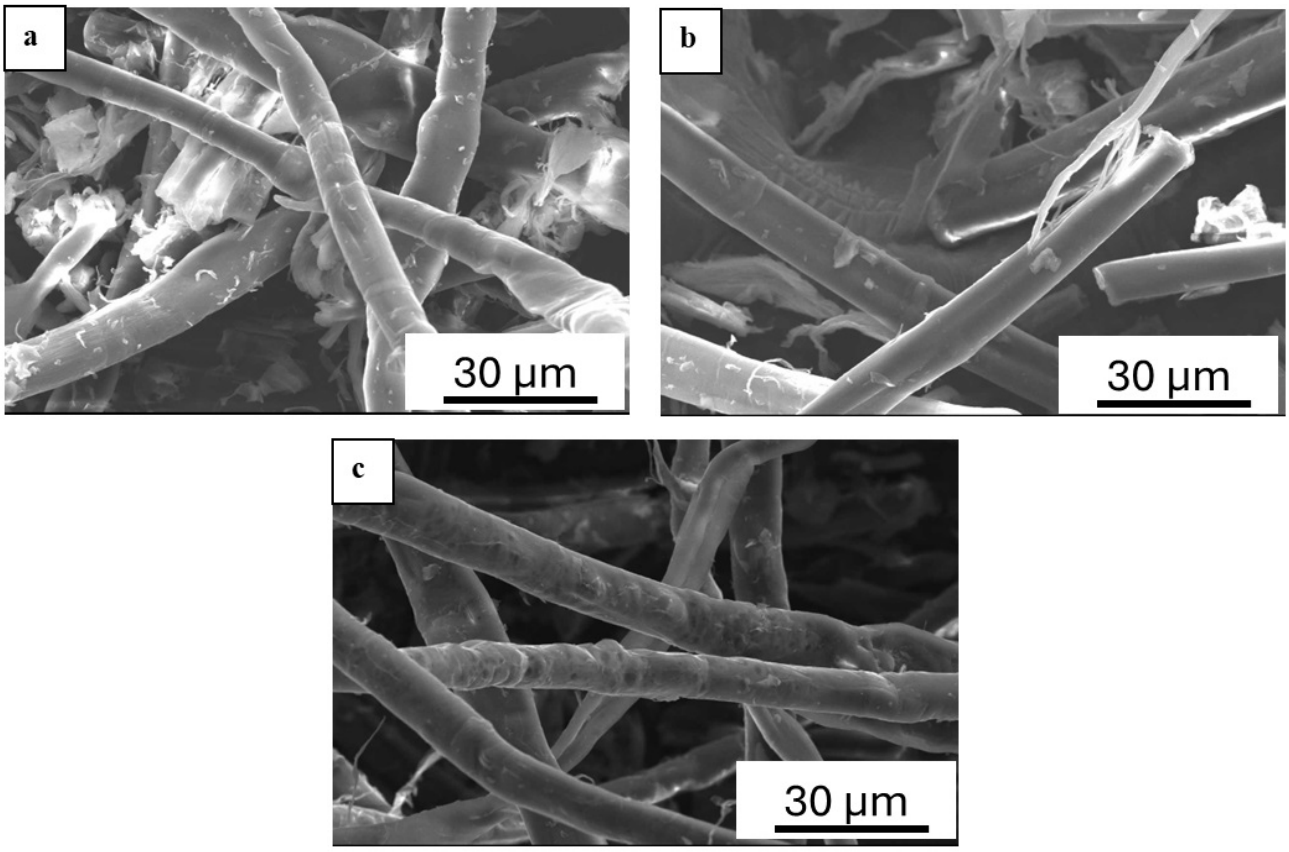

Figure 3: Representative SEM micrographs of the Argeli fibers: **a**) nAF, **b**) tAF, and **c**) bAF.

previous section (see Table **4**). The decrease in fiber diameter, weight, and disintegration of the bundles into individual fibers supported the removal of the loosely bound compounds from the lignocellulosic framework. Further, in treatment with $NaClO_2$ solution, the roughness of the fiber increased (see Figure **3c**) by dissolving more impurities but the density and diameter of fiber decreased implying that the dissolved compounds comprised had highly dense substances. Overall, the surface roughness of the fibers was found to increase on prolonged and multiple-stage chemical treatment.

The FTIR spectra of variously treated Argeli fibers are presented in Figure **4**. We discuss here the qualitative aspects of the peak positions relating them to the corresponding functional group as the spectra have been shifted vertically along the ordinate to ease the comparison between the spectra of different samples. The relative peak intensities hence do not necessarily reflect the amount of the substances absorbing at a corresponding wavelength.

The IR vibrational peak centered at 3314 cm^{-1} is due to the OH- stretching of H-bonded α - cellulose of hemicelluloses and cellulose. The peak centered at 2907 cm^{-1} is associated with the C-H stretching of $–CH_3$ and $–CH_2$- groups present in cellulose and hemicelluloses. The peak centered at 1729 cm^{-1} corresponds to the C=O stretching of the acetyl group present in lignin and hemicelluloses [43].

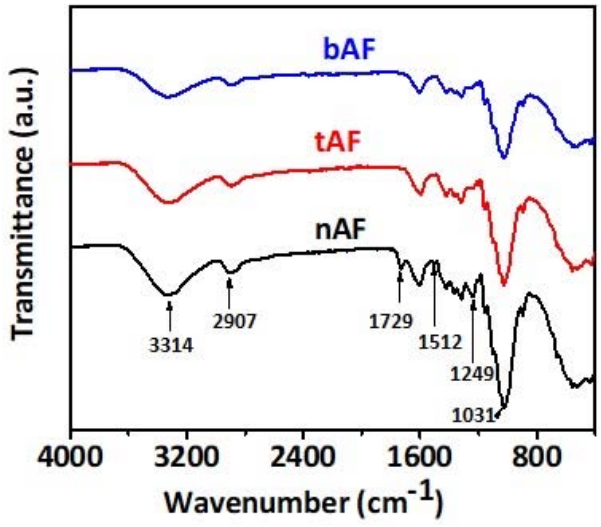

Figure 4: FTIR spectra of neat and chemically treated Argeli fibers: nAF, tAF, and bAF.

The disappearance of peaks at 1729 cm^{-1} and 1249 cm^{-1} on alkali treatment indicates the removal of lignin. The peaks located at 1249 cm^{-1} and 1031 cm^{-1} are representative of the $-OCH_3$ of lignin and C-O-C pyranose ring stretching of hemicellulose and cellulose, respectively [44]. The chemical treatments lead to a decrease in the intensity of these peaks leading to partial removal of non-crystalline compounds such as lignin, hemicelluloses, and pectin. The disappearance of both the peaks centered at 1729 and 1249 cm^{-1} indicates the removal of hemicelluloses, lignin, and waxes after chemical treatments.

3.4. Effect of Chemical Treatment on Thermal and Mechanical Properties

For several technical applications, the thermostability of the materials is an important aspect to consider. This is also true in the case of natural fibers that are used as they are or in combination with other materials. Figure **5** shows the TGA data (Figure **5a**) and corresponding derivative (DTG) (Figure **5b**) plots of the Argeli fibers as a function of chemical modifications as indicated. The curves in the DTG plot are vertically shifted removing the scale to ease the comparison of the peaks.

It can be observed that all three samples presented in Figure **5** have similar behaviors upon heating from 0 °C to 750 °C, with a broad peak around 100 °C and a significant degradation peak located between 340 °C and 370 °C. The peak at lower temperatures corresponds to the removal of water vapor and other volatile compounds while that at higher temperatures is done to the total degradation of the lignocellulosic framework. Nevertheless, it can be observed that the thermostability of the alkali-treated and bleached fibers have similar TGA curves with the highest degradation temperature located around 340 °C.

Overall, the thermostability of the natural fibers is not much affected by their chemical modification. However, the alkali-treated fibers have slightly lower thermal stability than the neat ones as it could lead to the degradation or depolymerization of cellulose chains [45]. This chemical breakdown of cellulose may result in a decrease in molecular weight leading to a decrease in thermal stability. In addition, alkali treatment may remove hemicelluloses and lignin which are the components of the plant fibers that have lower stability than the cellulose, but the removal of these components may alter the arrangement of the crystalline phase in the composite framework fiber and cause an overall reduction in thermal stability of the fiber [45].

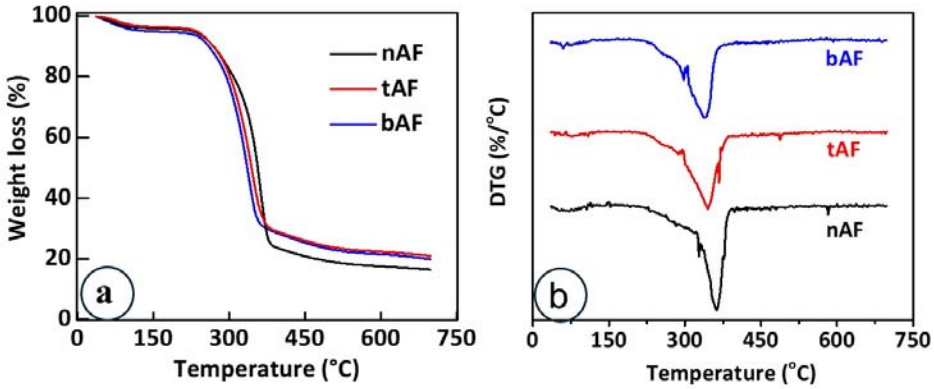

Figure 5: TGA (a) and DTG (b) curves of differently treated Argeli fibers depicting their thermal stability behavior.

The tensile stress-strain curves of the differently treated fibers are presented in Figure **6**. Each curve is the most representative one out of the plots of 10 experiments. It can be observed that there is a difference in the yield strength of the fibers. While the yield strength of the neat Argeli is about 190 MPa, the alkali-treated, as well as bleached fibers, exceed the properties of the neat fibers quite significantly, with the yield strengths of alkali-treated and bleached fibers measuring about 250 MPa and 360 MPa, respectively.

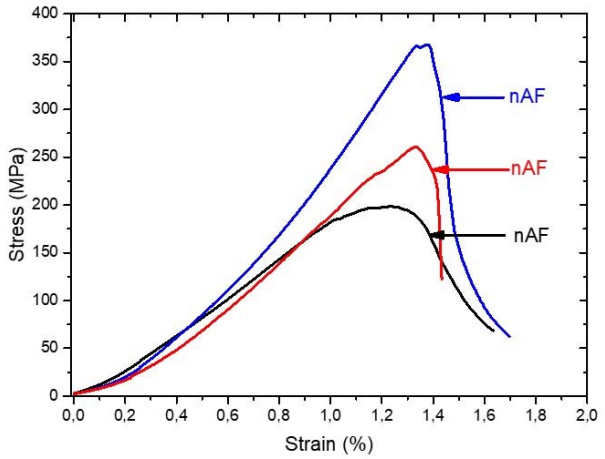

Figure 6: The tensile stress-strain curves of differently treated Argeli fibers, nAF, tAF, and bAF.

The increase in the tensile strength of fibers on chemical treatment can be correlated to the removal of loosely bound impurities (such as waxes, pectin, lignin, and hemicellulose) from the fiber surfaces. Thus, the alkali-treated fibers have more strength as compared to untreated fiber. Alkali treatment may activate the reactive sites on the fiber surface such as the hydroxyl group of cellulose fibers. These activated sites are more prone to forming stronger intermolecular bonds with other fibers enhancing thereby the tensile strength [46]. A comprehensive study of the mechanical properties, including Young's modulus and strain at break and micromechanical behavior of the fibers with sufficient consideration of the processing conditions is reserved for future publication.

4. CONCLUSIONS

The structural and morphological features of the fibers extracted from Argeli (*Edgeworthia gardneri*) bast were investigated using various advanced analytical techniques such as Fourier transform infrared (FTIR) spectroscopy, Energy Dispersive X-ray (EDX) spectroscopy, and Scanning Electron Microscopy (SEM). The fibers were treated with various concentrations of sodium hydroxide and sodium chlorite solutions to study the effect of the treatments on their physical, morphological, thermal, and water absorption behavior. The major outcome of the research can be summarized as follows:

- Cellulose, hemicelluloses, and lignin are the main constituents of Argeli fibers. Externally, the fiber surfaces were coated with hydrophobic extractives such as wax, oil, and pectin. On chemical treatments, the amorphous impurities are removed with improved surface roughness, mechanical strength, and water absorption ability of the fibers.
- The neat Argeli fibers contain about 54.47% of cellulose, 25.98% of hemicellulose, and 10.5% of lignin, with an average diameter and density of 13.57 µm and 1.28 g/cm^3, respectively.
- The chemical treatments, both with caustic soda and sodium chlorite, enhance the surface roughness, cleanliness, and mechanical performance of the fibers while the thermostability is found to remain nearly constant.

- The chemical treatment is found to partially dissolve the 'natural glues', hemicellulose, and lignin, and hence enhance the water absorption behavior of the fibers.

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AUTHORS' STATEMENT AND COMPETING INTERESTS

All the authors have read and approved the manuscript. They declare that they have no conflicts of interest.

AUTHOR'S CONTRIBUTION

PG: Performing the experiments, analysis of data, and drafting the first manuscript.

LG: Laboratory arrangements, analysis of data, and supervision.

SP: Supervision of the work, manuscript editing.

NLB: Supervision of the work, manuscript editing.

MN: Funding acquisition, conceptualization, supervision, and editing the manuscript.

RA: Funding acquisition, conceptualization, supervision, revising, and editing the manuscript.

REFERENCES

[1] Karimah A, Ridho MR, Munawar SS, Ismadi, Amin Y, Damayanti R, Lubis MA, Wulandari AP, Nurindah, Iswanto AH, Fudholi A. A comprehensive review on natural fibers: technological and socio-economical aspects. Polymers 2021; 13(24): 4280. https://doi.org/10.3390/polym13244280

[2] Reddy KO, Guduri BR, Rajulu AV. Structural characterization and tensile properties of borassus fruit fibers. Journal of Applied Polymer Science 2009; 114(1): 603-611. https://doi.org/10.1002/app.30584

[3] Rowell RM. Natural fibres: types and properties. In Properties and performance of natural-fibre composites 2008; 3-66. Woodhead publishing. https://doi.org/10.1533/9781845694593.1.3

[4] Mohammed L, Ansari MN, Pua G, Jawaid M, Islam MS. A review on natural fiber reinforced polymer composite and its applications. International Journal of Polymer Science 2015; 1: 243947. https://doi.org/10.1155/2015/243947

[5] Thyavihalli Girijappa YG, Mavinkere Rangappa S, Parameswaranpillai J, Siengchin S. Natural fibers as sustainable and renewable resource for development of eco-friendly composites: a comprehensive review. Frontiers in Materials 2019; 6: 226. https://doi.org/10.3389/fmats.2019.00226

[6] Madhu P, Sanjay MR, Senthamaraikannan P, Pradeep S, Saravanakumar SS, Yogesha B. A review on synthesis and characterization of commercially available natural fibers: Part-I. Journal of Natural Fibers 2018; 16(8): 1132-1144. https://doi.org/10.1080/15440478.2018.1453433

[7] Gholampour A, Ozbakkaloglu T. A review of natural fiber composites: Properties, modification and processing techniques, characterization, applications. Journal of Materials Science 2020; 55(3): 829-92. https://doi.org/10.1007/s10853-019-03990-y

[8] Jawaid MH, Khalil HA. Cellulosic/synthetic fibre reinforced polymer hybrid composites: A review. Carbohydrate Polymers 2011; 86(1): 1-8. https://doi.org/10.1016/j.carbpol.2011.04.043

[9] Komuraiah A, Kumar NS, Prasad BD. Chemical composition of natural fibers and its influence on their mechanical properties. Mechanics of Composite Materials 2014; 50: 359-76. https://doi.org/10.1007/s11029-014-9422-2

[10] Bledzki AK, Gassan J. Composites reinforced with cellulose-based fibres. Progress in Polymer Science 1999; 24(2): 221-274. https://doi.org/10.1016/S0079-6700(98)00018-5

[11] Tanpichai S, Sampson WW, Eichhorn SJ. Stress-transfer in microfibrillated cellulose reinforced poly (lactic acid) composites using Raman spectroscopy. Composites Part A: Applied Science and Manufacturing 2012; 43(7): 1145-1152. https://doi.org/10.1016/j.compositesa.2012.02.006

[12] Kiruthika AV. A review on physico-mechanical properties of bast fibre reinforced polymer composites. Journal of Building Engineering 2017; 9: 91-109. https://doi.org/10.1016/j.jobe.2016.12.003

[13] Pejic BM, Kostic MM, Skundric PD, Praskalo JZ. The effects of hemicelluloses and lignin removal on water uptake behavior of hemp fibers. Bioresource Technology 2008; 99(15): 7152-7159. https://doi.org/10.1016/j.biortech.2007.12.073

[14] Tuğluca MS, Özdoğru E, İ lcan H, Özçelikci E, Ulugöl H, Şahmaran M. Characterization of chemically treated waste wood fiber and its potential application in cementitious composites. Cement and Concrete Composites 2023; 137: 104938. https://doi.org/10.1016/j.cemconcomp.2023.104938

[15] Aliotta L, Gigante V, Coltelli MB, Cinelli P, Lazzeri A, Seggiani M. Thermo-mechanical properties of PLA/short flax fiber biocomposites. Applied Sciences 2019; 9(18): 3797. https://doi.org/10.3390/app9183797

[16] Pereira PHF, Ornaghi Jr HL, Arantes V, Cioffi MOH. Effect of chemical treatment of pineapple crown fiber in the

production, chemical composition, crystalline structure, thermal stability and thermal degradation kinetic properties of cellulosic materials. Carbohydrate Research 2021; 499: 108227.
https://doi.org/10.1016/j.carres.2020.108227

[17] Begum HA, Tanni TR, Shahid MA. Analysis of water absorption of different natural fibers. Journal of Textile Science and Technology 2021; 7(4): 152-160.
https://doi.org/10.4236/jtst.2021.74013

[18] Fook LT, Yatim JM. An experimental study on the effect of alkali treatment on properties of kenaf fiber for reinforced concrete elements. International Journal of Research in Engineering and Technology 2015; 4(8): 37e40.
https://doi.org/10.15623/ijret.2015.0408007

[19] Page J, Khadraoui F, Boutouil M, Gomina M. Multi-physical properties of a structural concrete incorporating short flax fibers. Construction and Building Materials 2017; 140: 344-353.
https://doi.org/10.1016/j.conbuildmat.2017.02.124

[20] Liu, K. Effects of sample size, dry ashing temperature and duration on determination of ash content in algae and other biomass. Algal Research 2019; 40: 101486.
https://doi.org/10.1016/j.algal.2019.101486

[21] Di Blasi C, Signorelli G, Di Russo C, Rea G. Product distribution from pyrolysis of wood and agricultural residues. Industrial and Engineering Chemistry Research 1999; 38(6): 2216-2224.
https://doi.org/10.1021/ie980711u

[22] Ayeni AO, Adeeyo OA, Oresegun OM, Oladimeji TE. Compositional analysis of lignocellulosic materials: Evaluation of an economically viable method suitable for woody and non-woody biomass. American Journal of Engineering Research 2015: 4(4): 14-19.

[23] Li S, Xu S, Liu S, Yang C, Lu Q. Fast pyrolysis of biomass in free-fall reactor for hydrogen-rich gas. Fuel Processing Technology 2004; 85(8-10): 1201-1211.
https://doi.org/10.1016/j.fuproc.2003.11.043

[24] Carneiro J, da S, Nogueira RM, Martins MA, Valladão DM. de S, Pires EM. The oven-drying method for determination of water content in Brazil nut. Bioscience Journal 2018; 3: 595–602.
https://doi.org/10.14393/BJ-v34n3a2018-37726

[25] Aryal GM, Kandel KP, Bhattarai RK, Giri B, Adhikari M, Ware A, Han S, George G, Luo Z, Gautam BR, Neupane BB. Material properties of traditional handmade paper samples fabricated from cellulosic fiber of Lokta bushes. ACS Omega 2022; 7(36): 32717-32726.
https://doi.org/10.1021/acsomega.2c04398

[26] Oktaee J, Lautenschläger T, Günther M, Neinhuis C, Wagenführ A, Lindner M, Winkler A. Characterization of willow bast fibers (Salix spp.) from short-rotation plantation as potential reinforcement for polymer composites. BioResources 2017; 12(2): 4270-4282.
https://doi.org/10.15376/biores.12.2.4270-4282

[27] Goda K, Cao Y. Research and development of fully green composites reinforced with natural fibers. Journal of Solid Mechanics and Materials Engineering 2007; 1(9): 1073-1084.
https://doi.org/10.1299/jmmp.1.1073

[28] Martijanti M, Juwono AL, Sutarno S. Investigation of characteristics of bamboo fiber for composite structures. IOP Conf. Series: Materials Science and Engineering; 2020, 850: 012028.
https://doi.org/10.1088/1757-899X/850/1/012028

[29] Andilolo J, Nikmatin S, Nugroho N, Alatas H, Wismogroho AS. Effect of kenaf short fiber loading on mechanical properties of biocomposites. InIOP Conference Series: Earth and Environmental Science 2017; 65(1): 012015. IOP Publishing.
https://doi.org/10.1088/1755-1315/65/1/012015

[30] Jakob M, Mahendran AR, Gindl-Altmutter W, Bliem P, Konnerth J, Mueller U, Veigel S. The strength and stiffness of oriented wood and cellulose-fibre materials: A review. Progress in Materials Science 2022; 125: 100916.
https://doi.org/10.1016/j.pmatsci.2021.100916

[31] Petroudy SD. Physical and mechanical properties of natural fibers. InAdvanced high strength natural fibre composites in construction 2017; 59-83. Woodhead Publishing.
https://doi.org/10.1016/B978-0-08-100411-1.00003-0

[32] Somerville C. Cellulose synthesis in higher plants. Annual Review of Cell and Developmental Biology 2006; 22(1): 53-78.
https://doi.org/10.1146/annurev.cellbio.22.022206.160206

[33] Bekele K, Linders TE, Eschen R, Shiferaw H, Haji J, Legesse B, Choge S, Eckert S, Mbaabu PR, Schaffner U. How well do local stakeholders' perceptions of environmental impacts of an invasive alien plant species relate to ecological data? Ecological Indicators 2022; 137: 108748.
https://doi.org/10.1016/j.ecolind.2022.108748

[34] Sawpan MA, Pickering KL, Fernyhough A. Effect of various chemical treatments on the fibre structure and tensile properties of industrial hemp fibres. Composites Part A: Applied Science and Manufacturing 2011; 42(8): 888-895.
https://doi.org/10.1016/j.compositesa.2011.03.008

[35] Hashim MY, Amin AM, Marwah OM, Othman MH, Yunus MR, Huat NC. The effect of alkali treatment under various conditions on physical properties of kenaf fiber. InJournal of Physics: Conference Series 2017; 914(1): 012030. IOP Publishing.
https://doi.org/10.1088/1742-6596/914/1/012030

[36] Islam MS, Ahmad MB, Hasan M, Aziz SA, Jawaid M, Haafiz MM, Zakaria SA. Natural fiber-reinforced hybrid polymer nanocomposites: effect of fiber mixing and nanoclay on physical, mechanical, and biodegradable properties. BioResources 2015; 10(1): 1394-1407.
https://doi.org/10.15376/biores.10.1.1394-1407

[37] Yang JM, Her YC, Han N, Clauer A. Laser shock peening on fatigue behavior of 2024-T3 Al alloy with fastener holes and stopholes. Materials Science and Engineerin A 2001; 298(1-2): 296-309.
https://doi.org/10.1016/S0921-5093(00)01277-6

[38] Scatolino MV, de Melo RR, de Oliveira Paula EA, Martins MA, Tonoli DHD, Esposito A, Mendes LM. Mechanical and surface properties of standard writing and printing papers coated with alternating layers of Amazonian Paricá nanofibrils and cassava starch. Journal of Materials Science 2024; 59: 11362-11380.
https://doi.org/10.1007/s10853-024-09827-7

[39] Husnil YA, Handayani AS, Setiaji DA, Chalid M. The effect of alkalization and bleaching treatment of Sorghum fibre on the crystallinity index of PP composite. InIOP Conference Series: Materials Science and Engineering 2019; 509(1): 012016. IOP Publishing.
https://doi.org/10.1088/1757-899X/509/1/012016

[40] Singleton AC, Baillie CA, Beaumont PW, Peijs T. On the mechanical properties, deformation and fracture of a natural fibre/recycled polymer composite. Composites Part B: Engineering 2003; 34(6): 519-526.
https://doi.org/10.1016/S1359-8368(03)00042-8

[41] Mwaikambo L. Review of the history, properties and application of plant fibres. African Journal of Science and Technology 2006; 7(2): 121.

[42] Hu Z, Ma Y, Chen H, Wei L, Zhu G, Liu L, Yao J. Efficient, durable, and breathable flame retardant cotton fabric via a feasible surface finishing. Applied Surface Science 2023; 615: 156314.
https://doi.org/10.1016/j.apsusc.2022.156314

[43] Prithivirajan R, Balasundar P, Shyamkumar R, Al-Harbi NS, Kadaikunnan S, Ramkumar T, Narayanasamy P.

Characterization of cellulosic fibers from Morus alba L. stem. Journal of Natural Fibers 2019; 16(4): 503-511. https://doi.org/10.1080/15440478.2018.1426079

[44] Md Salim R, Asik J, Sarjadi MS. Chemical functional groups of extractives, cellulose and lignin extracted from native Leucaena leucocephala bark. Wood Science and Technology 2021; 55: 295-313. https://doi.org/10.1007/s00226-020-01258-2

[45] Chen H, Zhang W, Wang X, Wang H, Wu Y, Zhong T, Fei B. Effect of alkali treatment on wettability and thermal stability of individual bamboo fibers. Journal of Wood Science 2018; 64: 398-405. https://doi.org/10.1007/s10086-018-1713-0

[46] Adeniyi AG, Onifade DV, Ighalo JO, Adeoye AS. A review of coir fiber reinforced polymer composites. Composites Part B: Engineering 2019; 176: 107305. https://doi.org/10.1016/j.compositesb.2019.107305

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