

CHARACTERIZATION OF FLAX FIBRES MODIFIED BY ALKALINE, ENZYME, AND STEAM-HEAT TREATMENTS

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Flax fibres are being considered as an environmentally friendly alternative to synthetic fibres in fibre-reinforced polymer composites due to their low density, biodegradability, and high mechanical strength. Previous work has found that the surface properties of natural fibres can be modified by chemical treatment and other treatment methods. This study focused on the effect of different treatments using alkaline, enzyme, and steam-heat, respectively, on some surface characteristics of flax fibre, e.g. physical, chemical, and thermal stability. Using scanning electron microscopy (SEM), treated fibres were observed to have smoother surfaces than untreated fibres. Chemical composition of fibres was found to be modified after treatment as characterized by Fourier transform infrared spectroscopy (FTIR). The crystallinity index and thermal stability of flax fibres were increased after certain treatments as determined by X-ray diffraction (XRD) and thermogravimetric analysis (TGA), respectively. The wettability of treated fibre by water was improved compared to the untreated sample.

Keywords: Flax fibres; Surface treatments; Analysis; Properties

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INTRODUCTION

Fibres from the flax plant (*Linum usitatissimum* L.) are classified as bast fibres because they are found in a peripheral band around the circumference of the plant stem known as bast. Similar to most environmentally friendly natural fibres, flax fibres are often investigated as materials for reinforced composites due to desirable attributes such as high mechanical properties, low density, and biodegradability. In general, the mechanical performances of a composite with fibres strongly depend on the nature and orientation of fibres and the quality of the adhesion between fibres. However, the properties and adhesive bond quality of flax fibres can be negatively impacted by harvesting age, growth conditions of the plants, and processing of the plants for extracting the fibre (Davies and Bruce 1998; Jaymini *et al.* 2002; Norton *et al.* 2006). Furthermore, characteristics of flax fibres can vary from different cultivation regions (Booth *et al.* 2004). Flax fibres usually require some form of treatments before they can be incorporated in composites to ensure a minimum level of product quality.

Some research work has been conducted on surface characteristics of natural fibre modified by chemicals (Jähn *et al.* 2002; Van de Weyenberg *et al.* 2003), enzyme (Akin *et al.* 2001; Mooney *et al.* 2001; Sharma *et al.* 2005), heat (Kessler *et al.* 1998; Sapieha *et*

al. 1989), and other methods. The physico-chemical properties of natural fibres can be changed significantly, depending on the concentration of chemical agent and duration of processing time. Jähn *et al.* (2002) reported that alkali concentration determined the extent of the polymorphic transformation of cellulose I into cellulose II taking place within the crystalline domains of the cellulose fibre. Van de Weyenberg *et al.* (2003) described that the combined treatment of alkali and dilute epoxy can give an improvement of 40 to 60% in the longitudinal bending strength and stiffness. Baley *et al.* (2006) reported that chemical treatments by sodium hydroxide/acetic anhydride and formic acid, respectively not only separated the individual flax fibres by dissolving the hemicellulose and pectin matrix, but also improved the adherence between flax fibre and unsaturated polyester resin at 78°C for 2 hours. Arbelaiz *et al.* (2006) used 20 wt% sodium hydroxide solution to treat flax fibres for 1 hour at room temperature; the results showed that the thermal stability of flax fibres was improved and the hygroscopicity of treated fibres decreased about 8 to 40% as a function of treatment used. Saha *et al.* (2010) reported that the diameter of jute fibres decreased by 23.4% upon 4% alkaline treatment at room temperature. The uniaxial tensile strengths of alkaline-treated and alkaline-steam treated jute fibre increased by 50% and 65%, respectively, compared to untreated fibres (370 ± 134 Mpa) due to the removal of non-cellulosic substances such as lignin, pectin, and hemicellulose.

Enzyme retting treatment increased the separation of flax bundles from shives and epidermis/cuticle and also separation into fibres and bundles of smaller divisions (Akin *et al.* 2001). Mooney *et al.* (2001) also reported that the xyloglucan polymers and xylan in flax fibres were not accessible to the enzyme, but that material could be released by the hydrolysis of this cellulose. Kessler *et al.* (1998) reported that the degraded materials, such as pectin, hemicellulose, *etc.*, were extracted by an alkaline scouring at different temperatures of 20 to 120°C. Prasad *et al.* (2004) researched the effect of heat treatment on properties of hemp fibre with temperature ranging from 160 to 260°C. The authors indicated that heat treatment at 220°C under nitrogen seemed to provide enough fibre opening without affecting the tissues of the fibres. However, few researchers have reported about the effect of heat treatment on the properties of flax fibres.

In this study, in order to better understand the influence of different treatment methods on characteristics of flax fibres, alkaline, enzyme, and steam-heat treatments were used to treat flax fibres. The morphologies were observed by scanning electron microscopy (SEM, JEOL JSM-6400), and the chemical compositions of untreated and treated flax fibres were characterized by Fourier transform infrared spectroscopy (FTIR, Nexus 670). Furthermore, the crystallinity index was determined by X-ray diffraction (XRD, Phillips XPERT-PRO) analysis; the thermal stability of flax fibres was investigated by thermogravimetric analysis (TG, Shimadzu DTG-60); and the contact angle of water on untreated and treated flax fibres was measured by a dynamic contact angle machine (Krüss K 12/14 desktop).

The other main purpose of this study was to improve the surface properties of flax fibres so as to enhance the bonding strength between flax fibres and resins in subsequent work.

MATERIALS AND EXPERIMENTS

Materials

Scutched flax (*Linum usitatissimum* L.) fibres were supplied by Stemergy of Ontario, Canada. Analytical grade of sodium hydroxide pellets with 97% purity was used in the alkaline treatment of the fibres. Laccase (L2157, from *Rhus vernicifera*) with enzymatic activity of 120 u/mg (u, abbreviation for units, refers to enzyme activity. One unit is the amount of activity which under standard conditions oxidizes 1 μ mol of 2,2-zino-bis-3-ethyl-benzo-thiazoline-6-sulfonic acid per minute in sodium tartaric or tartar acid buffer at pH 4.0 and 30 °C) is copper-containing oxidase enzyme that was supplied by Sigma-Aldrich of Saint Louis, USA.

Fibre Treatment

Three treatment methods were evaluated in this study. They are alkaline, enzyme, and steam-heat treatment. Details of the treatment schedules are described below.

Alkaline treatment

Dried flax fibres of 5 g were immersed in 150 mL of sodium hydroxide solution in a flask at 23°C, 60°C, and 100°C each for 1 and 2 hours, respectively. The concentration of sodium hydroxide solution was 3% and 10% wt/v, respectively. A solution to fibre ratio of 30:1 (mL/g) was maintained for all experiments. Sodium hydroxide solution was prepared by mixing sodium hydroxide granules with distilled water. After the alkaline treatment, the fibres were rinsed with tap water until the pH value of solutions reached 7. Then the fibres were dried in an oven set at 60°C overnight. There were three replicates for each treatment combination.

Enzyme treatment

Enzyme treatment was performed under the following conditions: pH of 3 and 5, temperature of 23°C, 36°C, and 50°C, and duration of 1 and 2 hours. The enzyme was applied at two dosages: 15u/g and 30u/g fibre. A solution to fibre ratio of 30:1 (mL/w) was maintained for all experiments. Dried flax fibres of 5 g were treated in each experiment, and there were three replications for each treatment combination. Fresh air was continuously injected into the mixture of flax fibres and enzymatic solution in a flask by a plastic tube during the entire treatment process. Then the fibres were dried in an oven set at 60°C overnight.

Steam-heat treatment

Dried flax fibres of 15 g were treated by steam at the following conditions: temperature of 160°C, 180°C, 200°C, and 220°C, and duration of 0.5, 1, and 2 hours in an airtight chamber with an atmosphere containing less than two percent oxygen. Superheated steam was used as a heating medium and a shielding gas during the whole treatment process, then the fibres were taken out from the device. Three replications were performed for each treatment combination.

Measurement Methods

Morphology

Scanning electron microscopy (SEM) of the flax fibres after different combination of treatments was performed using a JEOL JSM-6400 microscope at the conditions of accelerating voltage of 15.0 kV and a working distance of 16 mm. Flax fibres were sputter-coated with carbon and gold.

Fourier Transform Infrared Spectroscopy (FTIR)

The FTIR spectra at room temperature were measured in the spectral range of 4000 cm^{-1} to 400 cm^{-1} using an FTIR spectrometer (Nexus 670). A total of 32 scans were taken for each sample with a resolution of 2 cm^{-1} . The samples were processed into potassium bromide (KBr) pellets. All spectroscopic measurements were performed for the pulverized samples, which are a mixture of all three replicated specimens under the same treatment condition, obtained by milling the dried flax fibres.

Crystallinity Index

Fibres were finely cut to produce a powder less than 60-mesh, placed into a convenient depression within a flake of glass, and analyzed using a Phillips XPERT-PRO system with a Cu anode operation at 40 kV and 40 mA. The scan range was 2θ values from 5° to 55°, the counting time was 80 s, and the scan axis was Gonio. The crystallinity index was determined by using the following equation (Segal *et al.* 1959),

$$C_r I = \frac{(I_{002} - I_{am})}{I_{002}} \times 100 \quad (1)$$

where I_{002} is the counter reading at peak intensity at a 2θ angle close to 22°, and I_{am} is the amorphous counter reading at a 2θ angle of around 18°. The results are presented in Table 1.

Thermal Stability

In order to study the difference of thermal stability between untreated and treated fibres, thermogravimetric analysis (TGA) was performed using a Shimadzu DTG-60 thermoanalyzer. Samples weighing between 3 and 10 mg were placed in aluminum jars, and tests were carried out in an air atmosphere between 20°C and 600°C. The scan rate was set at 10°C/min.

Wettability

The wettability of water on the fibres, before and after various treatments, was determined using the modified Washburn method. This is the measurement of an interaction force between a liquid and a solid as the liquid is brought into contact with the solid. The contact angle was determined from the interaction force, the geometry of the solid, and the surface tension of the liquid. The Krüss Laboratory Desktop (K12/14) was used for contact angle measurements. A sample of fibre with the target mass was aligned and loaded tightly into the sample holder. The sample holder was then hung on a micro-balance, and the weight of the fibre samples together with the holder was tared to zero. The test liquid, maintained at 20°C, was brought under the sample holder. The test liquid

reservoir rose up to contact with the fibre sample at a rate of 5 mm/min. The balance detected the weight increase as a function of time until steady state was reached.

The wetting of n-hexane on the samples was first performed to obtain the capillary constant. Subsequently, the wetting of water on the samples was performed. At least three replications were performed of this measurement. The contact angle was calculated from the rate of water absorbed onto the samples (slope of the plot of mass² versus time) from a modified version of the Washburn Equation.

RESULTS AND DISCUSSION

Fibre Morphology

SEM micrographs show a fibre bundle of untreated flax fibres covered by non-cellulosic materials so that its surface looks uneven (Fig. 1a). Retting removes waxy epidermal tissue, adhesive pectins, and hemicellulose that bind fibre bundles to each other and to the pectin and hemicellulose-rich sheaths of the core (Sumere 1992). Alkaline, enzyme, and steam-heat treatments can remove waxy epidermal tissue from flax fibre bundle, which is in a good agreement with other results reported by Van de Velde and Baetens (2001). Smoother surface and more surface area of flax fibres were obtained after different treatments, as shown in Fig. 1 b-d. These figures are also proof that alkaline and enzyme treatments were able to separate the flax fibre bundle remarkably, while steam-heat treatment had slight effect on separation of flax fibre. This result is in good agreement with a conclusion by Kessler *et al.* (1998), who reported that the flax fibres did not separate well by steam-heat treatment.

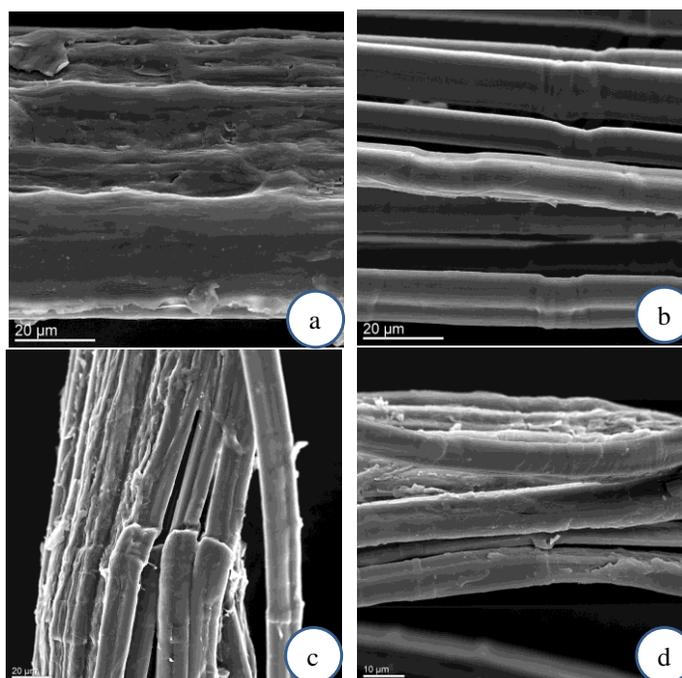


Fig. 1. SEM micrographs of untreated and treated flax fibres
a. Untreated b. Alkaline c. Enzyme d. Steam-heat

Fourier Transform Infrared Spectrometry (FTIR)

Single flax fibres were studied by FTIR microscopy. Different treatments led to significant differences in the infrared spectra as shown in Fig. 2. The FTIR spectra of treated and untreated flax fibres indicate that the intensity of the absorption band of $\nu(\text{O-H})$ stretching of hydrogen bond at 3419 cm^{-1} increased after steam-heat treatment, while it decreased slightly after alkaline and enzyme treatment. This is likely due to the breaking of hydrogen bonds between O-H groups of cellulose and hemicellulose molecules (Saha *et al.* 2010). The absorption peak at 1735 cm^{-1} associated to carbonyl (C=O) stretching of acetyl groups of hemicellulose (Haque *et al.* 2009), the absorbance bands at 1432 cm^{-1} and 1372 cm^{-1} (Saha *et al.* 2010) were assigned to $-\text{CH}_3$ asymmetric, and C-H symmetric deformations of lignin are seen in untreated, enzyme, and steam-treated flax fibres. However these peaks are detected in the alkaline-treated flax fibres very weakly; this weakening indicates the removal of the lignin after the treatment. These results show that alkaline treatment remarkably decreases certain chemical components, such as pectins and hemicellulose, while enzyme and steam-heat treatments showed no significant effect on the chemical components of flax fibres. Bhatnagar and Sain (2005) also reported that the alkaline treatment decreased the formation of hydrogen-type intermolecular bond due to ionization of pectin carboxylic groups and formation of sodium carboxylates. Furthermore, heating at elevated temperature led to degradation of pectic substances.

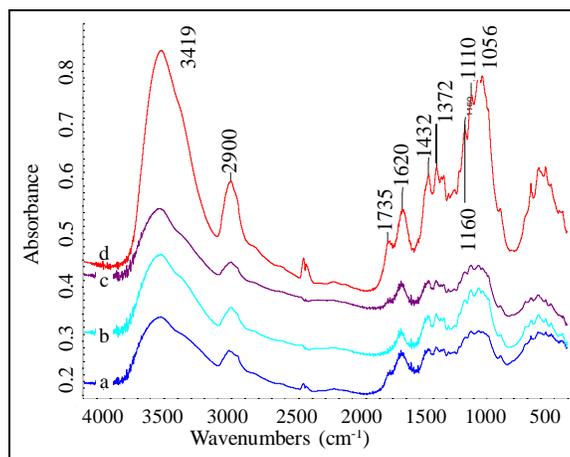


Fig. 2. FTIR spectra of untreated and treated flax fibres
a: untreated b: Alkaline: 100°C, 1h, 3% (NaOH) c: Enzyme:
50°C, 1h, pH 3, 30u/g fibre d: Steam-heat: 220°C, 2h

Crystallinity Index

The X-ray diffraction patterns of untreated and treated flax fibres are given in Fig. 3. These curves are examples of experimental conditions leading to an increase of the crystallinity index. The major peaks observed for all fibre samples were at 2θ diffraction angles of 18° to 22° .

The crystallinity index of untreated and treated flax fibre is shown in Table 1. The temperature, time, and concentration of NaOH solvent had significant effect on the crystallinity index of flax fibres. A 10% NaOH solvent decreased the crystallinity index

of flax fibre. Under the same treatment temperature and time, the crystallinity index was decreased with any increase in NaOH concentration. This result is in good agreement with results reported in the literature. Sharma *et al.* (2005) reported that when flax was mercerized with NaOH, crystallinity was decreased as shown by the increase in the amorphous cellulose content. Under the same treatment time and concentration of NaOH solvent, the crystallinity index increased with the increase of treatment temperature. Under the same temperature and concentration, crystallinity index decreased with the duration of treatment time. The X-ray diffraction analysis showed alkaline treatment improved the crystallinity index of flax fibres from 72 to 77, as seen in Table 1. The increase of crystallinity index of alkaline treated flax fibre occurred because of the removal of the cementing materials, which led to a greater packing of cellulose chains (Varma *et al.* 1984).

Table 1. Crystallinity Index Distribution of Untreated and Treated Flax Fibres

Treatment Methods	Treatment Parameters	Crystallinity Index	Treatment Methods	Treatment Parameters	Crystallinity Index
RAW		72.2			
Alkaline	23°C, 1H, 3%	72.1		23°C, 1h, pH 3, 15u/g	71.9
	23°C, 2H, 3%	73.0		23°C, 1h, pH 3, 30u/g	72.6
	23°C, 1H, 10%	61.4		23°C, 1h, pH 5, 15u/g	73.2
	23°C, 2H, 10%	60.9		23°C, 1h, pH 5, 30u/g	73.1
	60°C, 1H, 3%	74.0		23°C, 2h, pH 3, 15u/g	73.9
	60°C, 2H, 3%	71.6		23°C, 2h, pH 3, 30u/g	74.1
	60°C, 1H, 10%	67.7		23°C, 2h, pH 5, 15u/g	73.7
	60°C, 2H, 10%	67.2		23°C, 2h, pH 5, 30u/g	73.3
	100°C, 1H, 3%	77.4		36°C, 1h, pH 3, 15u/g	71.7
	100°C, 2H, 3%	76.8		36°C, 1h, pH 3, 30u/g	74.1
	100°C, 1H, 10%	67.6		36°C, 1h, pH 5, 15u/g	73.0
	100°C, 2H, 10%	68.8		36°C, 1h, pH 5, 30u/g	71.5
	Steam-heat	160°C, 0.5h	67.3	Enzyme	36°C, 2h, pH 3, 15u/g
160°C, 1h		68.1	36°C, 2h, pH 3, 30u/g		72.3
160°C, 2h		71.6	36°C, 2h, pH 5, 15u/g		73.7
180°C, 0.5h		67.2	36°C, 2h, pH 5, 30u/g		72.5
180°C, 1h		70.3	50°C, 1h, pH 3, 15u/g		73.0
180°C, 2h		69.8	50°C, 1h, pH 3, 30u/g		74.5
200°C, 0.5h		71.9	50°C, 1h, pH 5, 15u/g		72.7
200°C, 1h		72.6	50°C, 1h, pH 5, 30u/g		72.5
200°C, 2h		73.8	50°C, 2h, pH 3, 15u/g		72.0
220°C, 0.5h		71.2	50°C, 2h, pH 3, 30u/g		73.6
220°C, 1h		73.8	50°C, 2h, pH 5, 15u/g		73.4
220°C, 2h		74.3	50°C, 2h, pH 5, 30u/g		72.8

The crystallinity index of flax fibre was changed, depending on temperature and time of steam-heat treatment. Steam-heat treatment increased the crystallinity index of flax fibres from 72.2 to 74.3 in some treatment combinations. When the temperature was over 200°C and the treatment time was equal to or more than 1 h, the crystallinity index increased. Below 200°C, steam-heat treatment decreased the crystallinity index. Kalia *et al.* (2009) reported that hydrothermal treatment can increase the crystallinity of cellulose due to reducing moisture uptake. Moreover, a part of hemicellulose is extracted, thereby decreasing the moisture absorbance during hydrothermal treatment.

On the other hand, enzyme treatment had slight effect on the crystallinity index of flax fibres. Most combinations of enzyme treatment increased the crystallinity index. Enzyme treatment improved it from 72.2 to 74.5. A similar result was concluded by other researchers. Li and Pickering (2008) reported that the crystallinity index of enzyme-treated hemp fibres was increased to around 86, compared with untreated fibre index of 66.3. This increase could be attributed to the removal of amorphous non-cellulosic or amorphous cellulose compounds by enzyme treatment, which would allow better packing of the cellulose chains. Pickering *et al.* (2007) reported combined enzyme and alkali treatment increased crystallinity index of hemp fibre due to lignin removal.

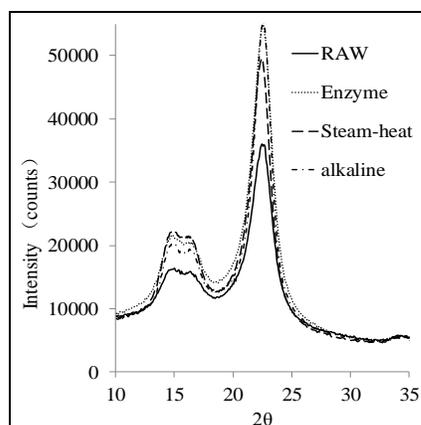


Fig. 3. Comparison of crystallinity index of untreated and treated flax fibres
Raw: untreated; Enzyme: 50°C, 1h, pH 3, 30 u/g fibre; Steam-heat: 220°C, 2h;
Alkaline: 100°C, 1h, 3% (NaOH)

Thermogravimetric Analysis

Generally speaking, there are three peaks in the derivative curve shown in Fig. 4. The first peak can be attributed to the release of water. The second peak is related to the degradation of cellulose and hemicellulose. The last peak is related to degradation of non-cellulosic substances (Arbelaiz *et al.* 2006). The curve of weight changes with temperature is shown in Fig. 5. An overview of weight loss rates of untreated and treated flax fibres is presented in Table 2.

The weight losses observed below 150°C are the pre-heating phase due to moisture loss from the minerals. Steam-heat treatment retained lower moisture in the flax fibre than alkaline-treated, enzyme-treated, or untreated flax fibre, as shown in Fig. 5. This result implied that steam-heat treatment reduced moisture remarkably relative to the untreated flax fibre, while alkaline or enzyme treatment had a slight effect on the

moisture of flax fibre. Thermogravimetric analysis revealed that untreated flax fibres achieved a 58% weight loss ratio at the first stage within the temperature range of 230°C to 348°C. In comparison, alkaline and enzyme treatments led to a higher reduction in weight loss ratio. This peak is supposed to be related to the degradation of cellulose and hemi-cellulose. On the other hand, similar weight loss occurred in the steam-heat treated flax fibre. This result indicated that steam-heat treatment had no significant effect on chemical compositions of flax fibres.

Table 2. Weight Loss Rate of Untreated and Treated Flax Fibres during Different Temperature

Treatment Methods	Treatment Parameters	Temperature Range °C	Weight Loss Ratio %	Temperature Range °C	Weight Loss Ratio %
RAW		230°C to 348°C	58	348°C to 465°C	38
	23°C, 2h, 3%		69		29
Alkaline	60°C, 1h, 3%		73		24
	60°C, 2h, 3%	256°C to 358°C	75	358°C to 470 °C	22
	60°C, 1h, 10%		73		23
	60°C, 2h, 10%		74		20
	100°C, 2h, 3%		79		15
	Enzyme	23°C, 2h, pH3, 30u/g		76	
36°C, 2h, pH3, 30u/g			76		19
50°C, 1h, pH3, 15u/g			75		20
50°C, 1h, pH3, 30u/g			74		19
50°C, 1h, pH5, 15u/g		244°C to 358°C	76	358°C to 460°C	21
50°C, 1h, pH5, 30u/g			77		18
50°C, 2h, pH3, 15u/g			76		18
50°C, 2h, pH3, 30u/g			76		20
50°C, 2h, pH5, 15u/g			75		21
50°C, 2h, pH5, 30u/g			75		19
Steam-heat	160°C, 2h		61		34
	180°C, 2h		59		36
	200°C, 0.5h	240°C to 348°C	63	348°C to 465°C	32
	200°C, 1h		59		36
	200°C, 2h		53		41
	200°C, 2h		46		47

At the second stage within the temperature range of 348°C to 465 °C, untreated flax fibre exhibited a 38% weight loss. In comparison, alkaline and enzyme treatments produced a lower reduction in weight loss rate. Steam-heat treated flax fibre generated a similar result to the untreated sample. These results imply that alkaline and enzyme treated flax fibres are more stable to thermal degradation than untreated flax fibre, which is in good agreement with the results reported by Arbelaiz *et al.* (2006). Steam-heat treatment has no significant effect on thermal stability of flax fibre.

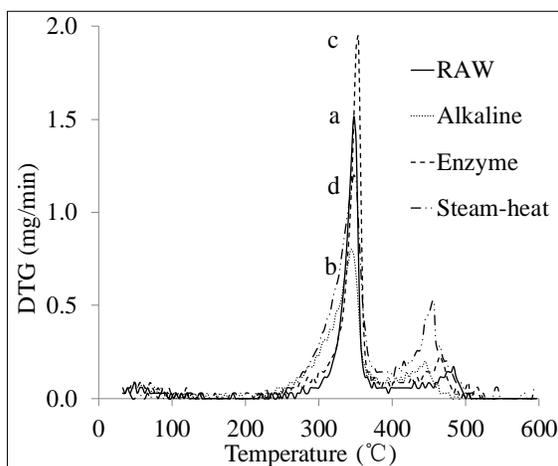


Fig. 4. DTG curve of untreated and treated flax fibres

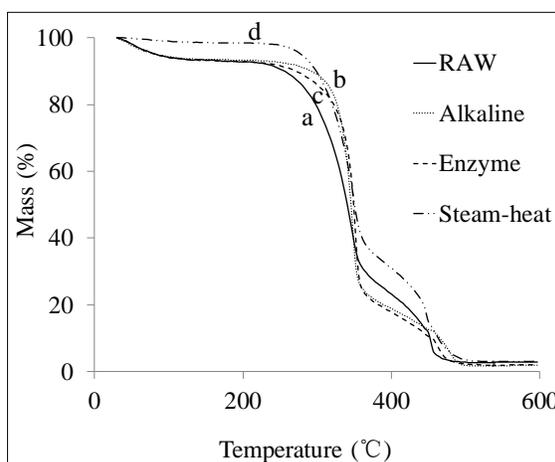


Fig. 5. TGA curve of untreated and treated flax fibres

a: Raw, untreated; b: Alkaline, 60°C, 2h, 10% (NaOH); c : Enzyme, 50°C, 2h, pH 5, 30u/g fibre; d: Steam-heat, 220°C, 2h

Wettability

Data for water contact angles on untreated and treated flax are shown in Table 3.

Table 3. Contact Angles of Untreated and Treated Flax Fibres

Specimens	Treatment Parameters	Contact Angle (degree)
Raw		89.99 ± 0.0
Alkaline	23°C, 2h, 3%NaOH	85.79 ± 0.3
	60°C, 1h, 3%NaOH	85.50 ± 1.0
	100°C, 1h, 3%NaOH	86.55 ± 0.8
Enzyme	23°C, 2h, pH5, 30u/g fibre	87.51 ± 0.7
	36°C, 2h, pH5, 30u/g fibre	87.76 ± 0.3
	50°C, 1h, pH3, 30u/g fibre	87.72 ± 0.8
	50°C, 2h, pH5, 30u/g fibre	86.58 ± 0.2
Steam-heat	160°C, 1h	88.68 ± 0.5
	180°C, 1h	88.65 ± 0.2
	200°C, 1h	89.45 ± 0.2

Alkaline, enzyme, and steam-heat treatments improved the wettability of flax fibre. However, steam-heat treatment showed slight effect on contact angle of flax fibre.

The contact angle of water on flax fibre was less than 90° , meaning that the fibre was hydrophilic in nature. The contact angles of the alkaline and enzyme-treated flax fibres were less than the contact angles of the untreated fibres. This signified that water can wet the treated fibres better than the untreated fibres. In other words, hydrophilicity of the fibres was significantly increased by alkaline and enzyme treatment. Figure 6 shows the water absorption of untreated and treated flax fibre. It implies that the three treatment methods accelerated the wettability compared with the untreated flax fibre. The difference in contact angle hysteresis between untreated and treated flax fibres can be explained by the difference in presence of higher surface energy components (Van de Velde and Kiekens 1999).

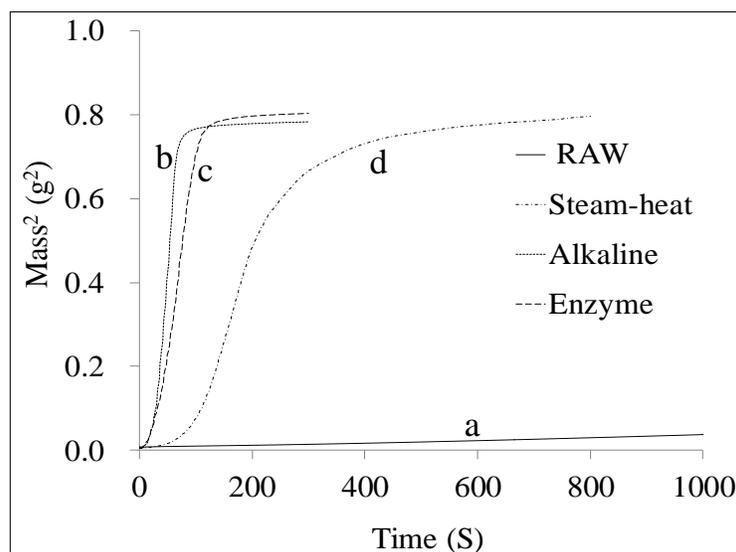


Fig. 6. Comparison of wettability of untreated and treated flax fibres
a: Raw: untreated b: Alkaline: 60°C , 1h, 3% (NaOH); c: Enzyme: 23°C , 2h, pH 5, 30u/g fibre; d: Steam-heat: 180°C , 1h

CONCLUSIONS

This study investigated the effects of three treatment methods on selected physical and chemical properties of flax fibres that may impact their adhesive bond quality. The following conclusions can be drawn from this study.

Compared to untreated flax fibres, a smoother surface and more surface area of flax fibres were obtained after any of the treatments considered, especially after alkaline and enzymatic treatments. Alkaline treatment decreases some chemical components on the surface of flax fibres, whereas enzyme and steam treatments have a slight impact on chemical components of flax fibres. Alkaline and enzyme treatments can remarkably improve the thermal stability of flax fibres, while steam-heat treatment has no significant effect on thermal stability. Alkaline treatment can enhance the crystallinity index of flax fibres, and enzyme and steam-heat treatments produce only a slight increase in crystallinity index. All three methods can improve water-wettability of flax fibres.

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REFERENCES CITED

- Akin, D. E., Foulk, J. A., Dodd, R. B., and McAlister III, D. D. (2001). "Enzyme-retting of flax fibre and characterization of processed fibers," *Journal of Biotechnology* 89 (2-3), 193-203.
- Arbelaiz, A., Fernández, B., Ramos, J. A., and Mondragon, I. (2006). "Thermal and crystallization studies of short flax fibre reinforced polypropylene matrix composites: Effect of treatments," *Thermochimica Acta* 440(2), 111-121.
- Baley, C., Busnel, F., Grohens, Y., and Sire, O. (2006). "Influence of chemical treatments on surface properties and adhesion of flax fibre-polyester resin," *Composites: Part A* 37(10), 1626-1637.
- Bhatnagar, A., and Sain, M. (2005). "Processing of cellulose nanofiber-reinforced composites," *Journal of Reinforced Plastics and Composites* 24(12), 1259-1268.
- Booth, I., Harwood, R. J., Wyatt, J. L., and Grishanov, S. (2004). "A comparative study of the characteristics of fibre-flax (*Linum usitatissimum*)," *Industrial Crops and Products* 20(1), 89-95.
- Davies, G. C., and Bruce, D. M. (1998). "Effect of environmental relative humidity and damage on the tensile properties of flax and nettle fibers," *Textile Research Journal* 68(9), 623-629.
- Haque, Md. M., Hasan, M., Islam, Md. S., and Ali, Md. E. (2009). "Physico-mechanical properties of chemically treated palm and coir fiber reinforced polypropylene composites," *Bioresource Technology* 100(20), 4903-4906.
- Jähn, A., Schröder, M. W., Fütting, M., Schenzel, K., and Diepenbrock, W. (2002). "Characterization of alkali treated flax fibres by means of FT Raman spectroscopy and environmental scanning electron microscopy," *Spectrochim Acta Part A* 58(10), 2271-2279.
- Jaymini, K., Roy, D. N., and Goel, K. (2002). "Effect of harvesting age on the chemical properties of hemp plants," *Journal of Wood Chemistry and Technology* 22(4): 285-293.
- Kalia, S., Kaith, B.S., and Kaur, I. (2009). "Pretreatment of natural fibers and their application as reinforcing material in polymer composites – A review," *Polymer Engineering and Science* 49(7), 1253-1272.

- Kessler, R. W., Becher, U., Kohler, R., and Goth, B. (1998). "Steam explosion of flax – A superior technique for upgrading fiber value," *Biomass and Bioenergy* 14(3), 237-249.
- Li, Y., and Pickering, K. L. (2008). "Hemp fibre reinforced composites using chelator and enzyme treatments," *Composites Science and Technology* 68(15-16), 3293-3298.
- Mooney, C., Stolle-Smits, T., Schols, H., and Jong, E. (2001). "Analysis of retted and non retted flax fibres by chemical and enzymatic means," *Journal of Biotechnology* 89(2-3), 205-216.
- Norton, A. J., Bennett, S. J., Hughes, M., Dimmock, J. P. R. E., Wright, D., Newman, G., Harris, I. M., and Edwards-Jones, G. (2006). "Determining the physical properties of flax fibre for industrial applications: The influence of agronomic practice," *Annals of Applied Biology* 149(1), 15-25.
- Pickering, K. L., Li, Y., Farrell, R. L., and Lay, M. (2007). "Interfacial modification of hemp fibre reinforced composites using fungal and alkali treatment," *Journal of Biobased Materials and Bioenergy* 1(1), 109-117.
- Prasad, B. H., Sain, M. M., and Roy, D. N. (2004). "Structure property correlation of thermally treated hemp fiber," *Macromolecular Material and Engineering* 289(6), 581-559.
- Saha, P., Manna, S., Chowdhury, S. R., Sen, R., Roy, D., and Adhikari, B. (2010). "Enhancement of tensile strength of lignocellulosic jute fibers by alkali-steam treatment," *Bioresource Technology* 101(9), 3182-3187.
- Sapieha, S., Pupo, J. F., and Schreiber, H. P. (1989) "Thermal degradation of cellulose-containing composites during processing," *Applied Polymer Science* 37(1), 233-240.
- Segal, L., Creely, J. J., Martin, A. E., and Conrad, C. M. (1959). "An empirical method for estimating the degree of crystallinity of native cellulose using the X-ray diffractometer," *Textile Research Journal* 29(10), 786-794.
- Sharma, H. S. S., Whiteside, L., and Kernaghan, K. (2005). "Enzymatic treatment of flax fibre at the roving stage for production of wet-spun yarn," *Enzyme and Microbial Technology* 37(4), 386-394.
- Sumere, C. F. (1992). "Retting of flax," in: H. S. S. Sharma, C.F. Sumere (eds.), *The Biology and Processing of Flax*, M. Publications, Belfast, UK, p. 157 Chapter 8.
- Van de Velde, K., and Kiekens, P. (1999) "Wettability of natural fibres used as reinforcement for composites," *Angewandte Makromolekulare Chemie* 272(1), 87-93.
- Van de Velde, K., and Baetens, E. (2001). "Thermal and mechanical properties of flax fibres as potential composite reinforcement," *Macromolecular Materials and Engineering* 286(1), 342-349.
- Van de Weyenberg, I., Ivens, J., De Coster, A., Kino, B., Baetens, E., and Verpoest, I. (2003). "Influence of processing and chemical treatment of flax fibres on their composites," *Composites Science and Technology* 63(9), 1241-1246.
- Varma, D. S., Varma, M., and Varma, I. K. (1984) "Coir fibers. Part I: Effect of physical and chemical treatments on properties," *Textile Research Journal* 54(12), 827-832.

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