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Optimisation of alkali treatment of banana fibres on lignin removal

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Alkali treatment of banana fibres has been used for lignin removal. The effects of various experimental parameters, such as alkali concentration, time and temperature, on lignin removal of banana fibres have been ascertained by response surface methodology using Box-Behnken design. The optimum conditions for lignin decomposition are identified as alkali concentration 11g/L, treatment time 150 min and temperature 90 ºC. The fibres treated under the optimum conditions are characterized based on chemical composition, diameter, density, moisture regain, strength, crystallinity and colour.

Keywords: Alkali treatment, Banana fibre, Lignin content

1 Introduction

Natural fibres, such as banana, flax, jute, ramie and hemp, offer several advantages such as low density, high stiffness, eco friendliness and sustainability. In the past, plant fibres were used for rope and twine manufacturing, fabric manufacturing in textile industry and paper industry[1-6]. The major constituents of all cellulosic fibres are cellulose, lignin and pentosan. The removal of lignin, hemicellulose and other contents by alkali treatment roughens the surface of fibre which is essential for it to be used as reinforcement in composites. It is known that high α-cellulose and low lignin content of a fibre are advantageous in textile application[5].

Banana fibre is a bast fibre which is obtained from the pseudo-stem of the plant. Fibres obtained are lustrous and fine with relatively good mechanical properties. The stem of banana plant is a major waste material which creates disposal problems. Banana fibre can be extracted from banana stem by manual, mechanical, chemical and biological methods. There is much variability in the banana fibres along the length and between fibres, which is a common characteristic of bast fibres[7,8]. The bast fibres have high cellulose content and a low micro-fibril angle which give them the desirable mechanical properties for it to be used in applications such as textile, composites, rope manufacture, etc. High holocellulose content and low lignin are indicated by chemical analysis of banana pseudo-stem fibre compared with some other non-wood fibre resource[9]. In textile processes, it is difficult to spin the fibre with 20-30% of adhered lignin. Hence, lignin needs to be removed by the process of degumming which is one of the most important sequences in processing of bast fibres; otherwise low-quality product will be the result[10-12].

Among all these treatments, mercerization or alkali treatment is a versatile one, which brings about changes in dimensions, fine structure, chemical composition, morphology, and crystalline component as well as improves the wettability, resin pick up of natural fibres like coir, sisal, flax, and cotton[12,13]. By boiling in optimum NaOH solution, lignin can be accessed and removed that allows rearrangement of molecules, which increases crystallinity and crystallite sizes[1,12]. Available literature reveals that alkali treatment removes the binding materials, depending on the treatment time, concentration of alkali used, temperature of treatment, liquor ratio, etc. Study on effect of alkali concentration on fibre yield and other properties of fibre has already been reported[14-17].

However, no research has been reported to find the optimum alkali treatment parameters. The main objective of this study is to optimize the lignin removal % of banana fibre with different concentrations of NaOH, time and temperature at three levels using Box- Behnken experimental design. The next objective is to determine the chemical composition and the other properties of fibre such as density, crystallinity percentage, moisture regain, colour of fibre and single fibre strength after treating the fibres in the optimized conditions.

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2 Materials and Methods

2.1 Materials

The banana fibres were mechanically extracted from the middle layers of the pseudostem from Nendran variety (Musa paradisiac) of banana plant grown in Kalapatti region of Coimbatore in Tamilnadu, India and dried in shade for 5 days. Sodium hydroxide, Sodium chloride, acetic acid, sulphuric acid, Xylene and carbon tetra chloride, obtained from Merck, India, were used for various methods for the determination of properties.

2.2 Methods

2.2.1 Experimental Design

Box Behnken design was used for experimental design and statistical analysis was performed using SYSTAT software. Three independent variables, namely NaOH concentration, time and temperature, were represented by X_1, X_2 and X_3 respectively. Each independent variable was coded in 3 levels (-1, 0 and +1). To study the effect of individual variables such as NaOH concentration, time and temperature on lignin decomposition separately, the process parameters for the various levels were fixed, as shown in Table 1 by conducting preliminary experiments.

2.2.2 Determination of Chemical Composition

2.2.2.1 Lignin Content

Two grams of extracted fibre sample were placed in a flask and 15 mL of 72% sulphuric acid was added. The mixture was stirred frequently for two and half hours at 25°C followed by addition of 200 mL of distilled water to the mixture. Then the mixture was boiled for next two hours and cooled. After 24 h, the lignin was transferred to the crucible and washed with hot water repeatedly until it becomes acid free. The collected lignin was dried at 105°C, cooled down in a desiccator and weighed. The drying and weighing were repeated till constant weight was achieved, and the lignin decomposition was calculated using the following equation:

$$\text{Lignin decomposition} = \frac{(M_0 - M_1)}{M_0} \times 100 \quad \ldots (1)$$

where M_0 is the lignin content of the untreated banana fibre; and M_1, the residual lignin content of the degummed banana fibre.

2.2.2.2 Determination of Holocellulose Content

Three grams of air dried fibre were weighed, placed in a flask, and then 160 mL of distilled water, 0.5 mL of glacial acetic acid and 1.5 g of sodium chloride were added successively. The flask was placed in water bath and heated up to 75°C for an hour and then an additional 0.5 mL of glacial acetic acid and 1.5 g of sodium chloride were added. The addition of acetic acid and sodium chloride was repeated two times hourly. The flask was placed in an ice bath and cooled down below 10°C. The holocellulose was filtered and washed with acetone, ethanol and water respectively. Finally, sample was dried in an oven at 105°C before it is weighed.

Table 1 – Process parameters and different levels for Lignin Removal

Factor	Levels		
	Low - 1	Medium 0	High +1
NaOH Concentration, g/L (X_1)	8	10	12
Time, h (X_2)	1	2	3
Temperature, °C (X_3)	70	80	90

2.2.2.3 Determination of α-Cellulose and Hemicellulose Contents

Two grams of holocellulose was placed in a beaker and 10 mL of sodium hydroxide solution (17.5%) was added. The fibres were stirred by glass rod so that they could be soaked with sodium hydroxide solution vigorously. Then sodium hydroxide solution was added to the mixture periodically (once in every 5 min) in 30 min and the mixture temperature was kept at 20°C. About 33 mL of distilled water was added in the beaker and kept for an hour. The holocellulose residue was filtered, transferred to the crucible and washed with 100 mL of sodium hydroxide (8.3%), 200 mL of distilled water, 15 mL of acetic acid (10%) and again water successively. The crucible with α-celluloses was dried and weighed.

The content of hemicelluloses of banana fibre was calculated from the following equation:

$$\text{Hemicelluloses} = \text{Holocellulose} - \alpha - \text{Cellulose} \quad \ldots (2)$$

2.2.3 Determination of Physical Properties

The surface morphology and diameter were measured using a polarised microscope (Lieca make) with a magnification of ×100 and interfaced with a PC. Fibres were measured for diameter in at least 10 different locations for 30 different fibres and the average of these measurements was calculated. Density and moisture regain were measured according to the ASTM D 1505 – 03 and ASTM D 2495 07 standards respectively.

Tensile properties of the untreated and treated banana fibres were measured according to the ASTM D 3822 – 01 standard test method for tensile properties of single textile fibres. The test was performed with 1kN loadcell at a crosshead speed of

5 mm/min, and approximately 20 fibres were tested for each sample, at 10mm gauge length in Zwick tensile tester. The colour of treated and untreated banana fibres was determined using the Premier Colour Scan, Mumbai, India. L, a, b and ΔE values were obtained from the instrument.

2.2.4 Crystallinity

X-ray diffractograms (scan range 2θ = 10 - 45°, θ -diffraction angle, scan speed 5.0 deg/min) of the untreated and treated fibres were obtained with a Shimadzu X-Ray diffractometer Model XRD600 having a X-ray tube producing monochromatic using Copper K alpha radiation at 30 kV and 20 mA.

3 Results and Discussion

3.1 Effect of Process Parameters on Lignin Decomposition

The independent variables, viz concentration of NaOH, treatment time and temperature were considered at three levels each. Based on Box-Behnken experimental design, fifteen combinations to perform the experiment were obtained. The experiment was performed and the influence of variables on the response (lignin decomposition %) was studied. (Table 2).

The impact of three process variables on lignin decomposition is very significant as confirmed from higher R^2 values (R=0.982). The regression equation gives the level of lignin removal as a function of different concentration, time and temperature. All the terms regardless of their significance are included in the following equation:

$$Y = 35.1 + 5.837X_1 + 6.362X_2 + 7.525X_3 - 6.725X_1^2 - 6.125X_2^2 - 5.15X_3^2 \quad \ldots (3)$$

Table 2 –Lignin decomposition of banana fibre samples using different process parameters

Run No.	NaOH conc. g/L	Time min	Temperature °C	Lignin decomposition, %
1	8	60	80	11.80
2	8	180	80	17.60
3	12	60	80	23.20
4	12	180	80	36.40
5	8	120	70	10.00
6	8	120	90	28.20
7	12	120	70	19.40
8	12	120	90	35.30
9	10	60	70	10.50
10	10	60	90	21.20
11	10	180	70	24.10
12	10	180	90	39.50
13	10	120	80	35.13
14	10	120	80	35.3
15	10	120	80	35.28

where *Y* is the lignin decomposition.

Results also show that the linear and quadratic effects of variables are significant ($p<0.05$), while there is no significant interaction ($p>0.05$). Each individual parameter has significant effect on lignin decomposition. The increase and decrease in these parameters show a change in lignin content of the fibre, whereas their interactions do not significantly affect the lignin removal. This is because, even though the concentration is high, the required amount of lignin will not be removed without particular treatment time or temperature.

3.2 Optimization of Lignin Removal using RSM

Contour plots were drawn to understand the influence of process parameters on lignin decomposition. Influence of process parameters on lignin decomposition has been elaborately discussed in three combinations of process parameters, such as influence of NaOH concentration and time, influence of NaOH concentration and temperature, and influence of time and temperature.

3.2.1 Effect of NaOH Concentration and Time on Lignin Decomposition

Figure 1(a) shows the influence of NaOH concentration and treatment time on lignin decomposition %. The low concentration of NaOH and lower treatment time does not influence lignin decomposition %. The lignin removal is found poor for lower treatment time even with the higher concentration of NaOH. This is because the fibre requires some time to swell so that the auxiliary agents can enter into the fibre. At lower time durations, whatever may be the concentration, the auxiliaries do not have space to attack and decompose the lignin. In the same way, with the lower concentrations and higher treatment time, the alkali is not efficient to decompose the lignin and other impurities from the fibre. Figure 1(a) shows that the concentration of 11g/L and treatment time of 2.5 h are found to be more efficient in lignin removal.

3.2.2 Effect of NaOH Concentration and Temperature on Lignin Decomposition

Figure 1(b) shows that low concentration of NaOH and lower treatment temperature do not influence lignin decomposition %. The lignin removal is slightly increased even at lower treatment temperature but with higher concentration of NaOH. Temperature is necessary to make the fibre to swell and alkali concentration is a must to decompose the impurities from the fibre. With high concentrations, the increase in treatment temperature linearly increases lignin decomposition% and it is nearly 40% at 11g/L of NaOH and 85-90^{0}C of treatment temperature.

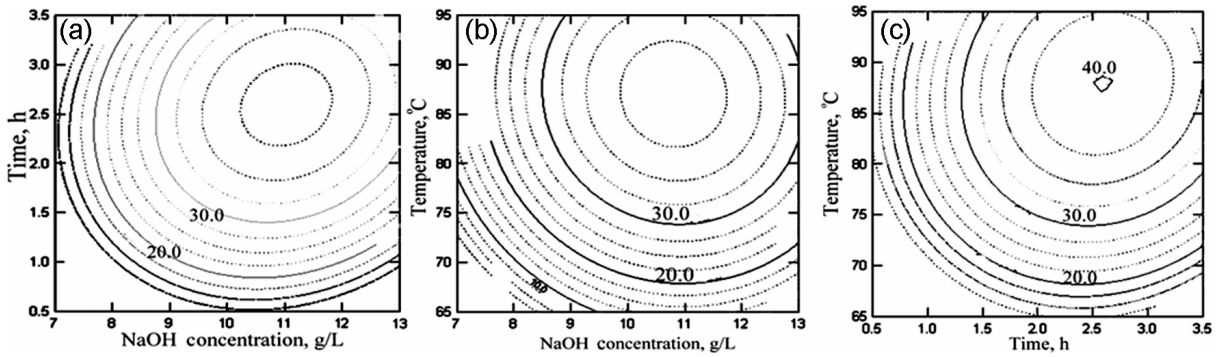

Fig. 1 –Influence of process parameters on lignin decomposition% (a) NaOH concentration and time, (b) NaOH concentration and temperature, and (c) time and temperature

3.2.3 Effect of Treatment Temperature and Time on Lignin Decomposition

Figure 1(c) shows the influence of treatment time and temperature on lignin decomposition %. At higher treatment time and lower temperature, the alkali action on fibre is not very fast. So, the lignin decomposition is very low. At longer treatment time and higher temperature the best results are obtained than at low temperature and higher treatment time and vice versa. Lignin removal is found high at 2.5 h of treatment time and 85-90^{0}C of treatment temperature.

From the responses, optimum concentration, time and temperature for effective lignin decomposition without affecting the cellulose fibrils of banana fibres are obtained. As observed from the results, the optimum conditions are 11g/L of NaOH concentration, 90^{0}C of treatment temperature and 2.5 h of treatment time.

3.3 Chemical and Physical Properties of Banana Fibre

The other chemical and physical properties of banana fibres have been obtained after treating them under optimum conditions and are discussed hereunder.

3.3.1 Chemical Composition

The major constituents of untreated and treated banana fibre have been assessed to analyze the effect of degumming on the fibre. It can be seen from Table 3 that the non-cellulosic constituents namely hemicellulose and lignin are obviously removed by degumming process. The lignin removal percentage is around 40%. The increase in α-celluloses content is almost 20% of the initial. The hemicelluloses content is reduced to 50% of its initial content in the untreated fibre.

3.3.2 Surface Morphology and Diameter

The untreated and treated (11 g/L, 90^{0}C and 150 min) fibres were examined under a light microscope with the magnification of × 100. Before any treatment, there is no visible separation of fibres, which are held together by lignin and pectin substances. After boiling the fibres in alkali, there is much less splitting and still some lignin on the surface of the fibres is observed.

Table 3 – Chemical composition of banana fibre

Sample	Holocellulose %	α-cellulose %	Hemicellulose %	Lignin %
Untreated	67.1	62.1	5.0	17.00
Treated	76.9	74.8	2.1	10.25

Table 4 – Properties of untreated and treated banana fibres

Sample	Diameter µm	Diameter CV%	Density g/cc	Moisture regain %	Single fibre strength g/tex
Untreated	213.000	49.0	1.35	13.23	30.66
Treated	131.767	44.8	1.44	14.47	33.312

3.3.3 Density

The untreated and the treated fibre densities are observed in Table 4. The densities of the untreated fibres are found to be lower (1.35 g/cc) and that of the optimally treated fibres showed higher values of density (1.44 g/cc). The increase in density is due to the removal of the low denser regions such as lignin and hemicellulose. The α-cellulose content has higher density and the increase in the density is due to the increase in α-cellulose % in the composition.

3.3.4 Moisture Regain

Table 4 shows 2% increase in moisture regains. It is known that the lignin is less hydrophilic than the cellulose content and the hemicelluloses contain more hydroxyl groups than the celluloses. Removal of lignin should have increased the moisture regain to a higher level but the simultaneous removal of hemicelluloses has decreased the moisture regain values. Also, the cellulosic regions may have become more accessible to the moisture for absorption.

Table 5 – Colour values of untreated and treated banana fibre

Sample	L	a	b	c	H	ΔE
Untreated	69.393	4.401	17.747	18.285	76.042	13.51177
Treated	56.385	4.877	21.371	21.92	77.114	

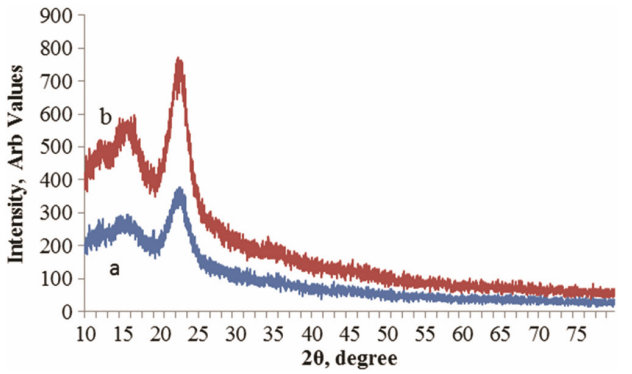

Fig. 2– XRD patterns of (a) untreated fibre, and (b) NaOH treated fibre

3.3.5 Single Fibre Strength

Single fibre strength of the fibres (Table 4) reveals that there is an increase in strength due to alkali treatment. The increase in tensile strength may be due to the action of the alkali on the non-cellulosic contents and their removal. In general, pectin, hemi-cellulose and lignin are amorphous and are very disorderly in nature. They form pools in between the cellulosic regions which prevent the α-cellulose chains from positioning themselves in the direction of loading. The removal of these pools makes the fibres α-cellulosic regions to withstand higher stresses comparatively.

3.3.6 Colour

The colours of treated and untreated banana fibres are shown in Table 5. It shows that there is significant change in colour which is evident with higher ΔE values. The decrease in L value of the treated fibre indicates that the fibres have become darker which was evident from the visual observation also. The fibres are towards redder side as the values of 'a' are positive and the redness has also increased. The increase in the 'b' value shows that the fibre is in the yellower side and the yellowness has increased. Also there is an increase in the hue value.

3.3.7 Crystallinity

The X-ray diffractogram patterns of the untreated and NaOH treated fibres are shown in Fig.2. The figure shows reveals that there is an increase of crystalline regions in the treated fibre samples, confirming the increase in cellulose content in the treated banana fibre.

4 Conclusion

In this study, analysis of alkali treatment on banana fibres revealed satisfactory results, and the conclusions are as follows:

4.1 According to the results of Box-Behnken method, the optimum conditions for higher lignin removal from banana fibres are NaOH concentration 11g/L, treatment time 2.5h and temperature 90^0C.

4.2 There are clear changes in fibre composition due to alkali treatment under optimum conditions. The removal of lignin from mechanically extracted banana fibre is at least 40% in the optimum conditions. There is 20% increase in the cellulose content. The hemicellulose contents are also reduced by half of the actual content.

4.3 The surface morphology reveals that there is a separation in the fibre bundles.

4.4 There is an increase in density due to removal of the low denser hemicellulose and lignin contents. There is an increase in moisture regain of the fibres, thus making the fibre more hydrophilic and suitable for the textile applications. It is a drawback when fibre is used as reinforcement in composite application since it reduces the wettability of the resin.

4.5 The single fibre strength and the crystallinity of the fibres have improved, which is an advantage in both textiles as well in composite applications.

4.6 The colour has become darker and the hue has increased which is a drawback from the aesthetic point of view.

References

1 Ramasamy N & Craft S, *Text Res J*, 65(1995) 765.
2 Khalil H P S A, Alwani M S & Omar A K M, *Bioresources*, 1(2006) 220-232
3 Bacci L, Lonardo S D, Albanese L, Mastromei G & Perito B, *Text Res J*, 81(2010) 827.
4 Bodros E & Baley C, *Mater Lett*, 62 (2008) 2143.
5 Das P K, Nag D, Debnath S & Nayak L K, *Indian J Tradit Know*, 9(2010)386.
6 Chawla K K, *Fibrous Materials* (Cambridge University Press, U.K), 2005.
7 Kulkarni A G, Sathyanarayana K G, Rohatgi P K & Kalyani Vijayan, *J Mater Sci*, 18(1983) 2290.
8 Mukhopadhyay S, Fangueiro R, Arpac Y & Senturk U, *J Eng Fiber Fabrics*, 3(2008) 39.
9 Li K, Fu S, Zhan H, Zhan Y & LuciaL A, *Bioresources*, 5(2010)576.
10 Jinqiu Z & Zhang Jianchun, *Text Res J*, 80 (2010)744.
11 Wang W M, Cai Z S & Yu J Y, *J Eng Fiber Fabr*ics, 3(2008)1.
12 Ray D, Das M & Mitra D, *Bioresources*, 4 (2009)730.
13 Oladele I O, Omotoyinbo J A & Adewara J O T, *J Miner Mater Charact Eng*, 9(2010) 569.
14 Ebisike K, Attah Daniel B E, Babatope B & Olusunle S O O, *Int J Eng Sci*, 2 (2013) 99.
15 Ramadevi P, Dhanalakshmi S, Srinivasa C V & Bennehalli B, *Bioresources*, 7 (2012) 3515.
16 Kiruthika V & Veluraja K, *Fiber Polym*, 10(2009) 193.
17 Sinon F G, Kohler R, Cotter M & Muller J, *J Biobased Mater Bio,* 5 (2011) 433.