Assessment Of Performance Characteristics Of Blended Nonwovens From Areca Catechu Husk Fibers And Polypropylene Fibers

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Abstract
Husk fibers of Areca Catechu which have limited applications have been selected and prepared to various stages to improve its absorbency and find its suitability as a nonwoven material. Box-Behnken experimental design has been used to optimize the delignification process in which 3 parameters have been considered to improve the absorbency of the fibers. The cooking time and alkali % of Alkali pretreatment of the fibers in an autoclave, and the time taken for delignification using laccase enzymes are the 3 parameters used. These processed fibers have been blended with Polypropylene fibers to render the required thermo plasticity for the fusing technique. The delignified fibers are converted to webs and later with fusing method converted to nonwovens. The prepared nonwovens have been tested for various properties and are compared with the properties of the commercially available polypropylene nonwovens.

Keywords—**Areca catechu husk fibers, Arecanut husk fibers, Lacasse enzyme, Delignification, Box-behnken, nonwovens**

INTRODUCTION
Apparel industry is showing higher commitments towards environmental sustainability and focus on them as part of their brand strategies both. This emphasize on eco-friendliness is done as a part of business practices and also to induce financial profits [1]. Sustainability is defined as the balance between three elements: economy, environment and social equity [2] & [3]. A product is said to be green based on the intrinsic characteristics of the product, using recycled raw materials [4]. While finished products are safe to human beings during their application, its manufacturing process should also be cautiously done so it does not harm the environment [5]. This cautiousness becomes mandatory in the textile and clothing industry, which is a composite industry encompassing a diverse range of activities from the transformation of raw materials to the final consumable product [6]. As one of the reflections of the cautiousness, a lot of initiatives are being taken in the areas of converting bio-lignocellulosics to highly valued products and that too in environmentally friendly ways. This sudden gain in importance is majorly due to the renewable nature of the lignocellulosic bio-mass [7]. These lignocellulosics are found richly in the agricultural biomass, which are polymers of lignin, cellulose and hemicelluloses [8].

Areca Catechu is one such type of a husk fiber of beetle nut which belongs to the Arecaceae family. These fibers are popular and cheap as they are a perennial crop and are available abundantly [9]. These husk fibers are a good alternative to natural fiber and could also be used to replace the synthetic fibers owing to environmental concerns [10]. Alkali treatment improves strength and the matrix adhesion of these natural fibers, which extends its application to a wider scope. By doing so its applications could be extended to textile field too [11]. The average filament length is around 4 cms. which is too short compared to other bio-fibers. Mainly 2 types of fibers are present – one very coarse and the other very fine. The coarser ones are ten times coarser than jute [9].

The coarser fibers called hard fibers, which are found in the inner layer of the arecanut are composed of irregularly lignified group of cells. The portions of the middle layer below the outermost layer are soft fibers, which are very similar to the jute fibers [24]. The arecanut husk fibers have varying compositions of very little traces of cellulose, hemicelluloses (35-64.8%), lignin (13-24.6%), ash (4.4%), water content (8-25%), pectin and proto-pectin. Their mechanical properties are found to be similar to that of coir and kenaf [12, 13]

The cell walls of plant cells consist of cellulose fibrils embedded within a matrix of lignin and hemicellulosic polysaccharides [14], Lignin is the main constituent of arecanut fiber, responsible for hardening of plant cell wall and the reason for the fiber stiffness. [9]. Removal of lignin called delignification is used to improve the absorbency of the fibers, further improving its applications [22]. This lignin removal can be done through various enzymatic methods which are commonly known as enzymatic delignification. This bio-delignification with the help of enzymes like lacasse replaces the mechanical treatment done with acid, alkali, and steam explosion [23].

The main objective of this study is to develop a nonwoven material from delignified arecanut husk fibers blended with polypropylene fibers. This fiber has been selected and prepared to various stages to improve its absorbency. This blending of natural and synthetic fibers will help in reducing

the content of synthetic fibers in nonwovens thereby also reduces the land fill considerably. New applications of agro-waste fibers like arecanut husk fibers will focus on recycling the agriwaste and facilitate sustainability.

MATERIALS AND METHODS
MATERIALS:
Areca Catechu (AC): Arecanut fibers are extracted from its matured and dried fruit. It is composed of both thick fibers and fine fibers. The finer fibers were extracted from the nut and used for further processing.

Polypropylene fibers (PP): Polypropylene fibers have been selected to blend with the prepared areca nut fibers to render the required thermo plasticity for the fusing technique of preparing nonwovens

Sapindus Mukorossi: Areca catechu fibers were cleaned with Sapindus Mukorossi, an organic cleaning agent, in order to remove natural wax content and impurities.

Chemicals: Sodium hydroxide has been used for pretreating the Areca Catechu fibers. Later these fibers were delignified using Bactosal LAC powder (a combination of laccase powder)

METHODS
Cleaning of Areca Catechu fibers with Sapindus Mukorossi (puchakai)
For cleaning organic cleaning agents called **Sapindus mukorossi** (puchakai) were used. Since ancient times, Sapindus mukorossi has been used as a detergent for shawls, silks and as a hair shampoo due to its rich natural lather owing to its rich saponin content[15] 10 % concentration of **Sapindus mukorossi** solution was prepared to treat Areca Catechu fibers with a M:L ratio of 1:10 till the boiling point.

Pre-treatment of Areca Catechu fibers by alkali cooking:
Alkali treatment of natural fibers is used to produce high-quality fibers by transferring crystallinity from cellulose I into cellulose II. Sodium hydroxide (NaOH) aids the greatest in degradation of hemicelluloses and lignin to a greater extent and the areca catechu fibers have been treated with alkali for this purpose [16, 17, 18, and 19]. Alkali pre-treatments at lower temperatures give coarser fibers but higher temperatures can give finer fibers. So in this study higher temperature was selected [20]
Areca Catechu fibers were cooked with varying concentrations of NaOH Alkali such as 5%, 7.5%, and 10% at the pH of 12 with varying cooking time of 30, 35 or 40 whistles in a autoclave. The temperature and pressure maintained in a autoclave is 120° C and 13 PSI pressure. [21]

Delignification
After the pretreatment with alkali cooking the Areca Catechu fibers are processed with the lacasse enzyme. The bath is set with M:L ratio of 1:10, pH 5-5.5, and temperature maintained at 65ºC and treated with 6% concentrations of Bactosol (lacasse enzyme) with varying duration of 4 hrs, 5 hrs or 6 hrs at 70ºC. Later fibers are rinsed in plain water and dried in the oven at 100ºC temperature for half an hour.

Box-Behnken Experimental design software
A Box-Behnken statistical design with 3 factors, 3 levels, and 15 runs was selected for the optimizing the alkali pre-treatment and the delignification of Areca Catechu fibers with laccase enzyme. The 3 Independent variables along with each of their 3 levels of values as given in the Table 1 below were defined. The independent variables selected were, alkali concentrations for the alkali cooking pretreatment in percentage, cooking time in the form of number of whistles allowed for cooking in the pressure cooker and finally the time in hours for treating the fibers in enzyme. A total of 15 runs with triplicate centre points as given in Figure 1 were generated. As per the fifteen batches of runs generated, the samples were alkali pre-treated and delignified. The absorbency of these delignified fiber samples were determined as per the standard Absorbency test for fibers INDA IST 10.1, 10.2. The optimization was done using the response factor analysis by giving the required absorbency target to be achieved. This will be generated depending on the maximum and minimum absorbency values (response values) which are obtained from the 15 runs.

Table 1: Variables Used For Optimizing The Fiber Processing

Independent Variables	Name	Units	Low Level	Medium Level	High Level
A	Alkali	Concentration in percentage	5%	7.5%	10%
B	Cooking time	Number of whistles on a pressure cooker	30	35	40
C	Time	Delignification Time in Hours	4	5	6

Preparation of blended nonwoven samples with the prepared Areca Catechu husk fibers and polypropylene fibers:
As per the optimized parameters for delignification Areca Catechu fibers were alkali pre-treated and delignified. These fibers were blended in various proportions with polypropylene fibers as given in the Table 2. The blended fibers were prepared into webs in a mini carder machine. These webs were used to prepare nonwoven sheets using the fusing technique. To avoid the sticking of fibers onto the fusing belt, food grade aluminum foil is placed on the top and bottom of the web and this arrangement is placed between sheets of news papers and then passed between the rollers for fusing. The parameters for preparing the fused nonwoven sample were maintained as:
Temperature -100° C, Pressure - 3 bars and Speed of 10 mts/mins.

Table 2: Areca Catechu / Pp Blended And Fused Nonwoven Samples

S.No	Sample name	Description of the fiber composition
1	PP-F	PP – 100%
2	PP-SB	PP – 100%
3	AC-PP -50	Raw Areca Catechu : PP -50/50
4	DAC-PP-25	Delignified Areca Catechu : PP -25/75
5	DAC-PP -50	Delignified Areca Catechu : PP -50/50
6	DAC-PP -75	Delignified Areca Catechu : PP -75/25

Dimensional and comfort properties of prepare fused nonwovens:

The prepared varieties of fused nonwovens were tested for their dimensional and performance properties using the standard test methods as given below in the Table 3. Triplicates were taken for each of the samples for all the properties and their mean were recorded.

Table 3: Tests Done On Prepared Nonwoven Samples

S.No	Experiment	Standard
1	GSM	ASTM D 2646
2	Air permeability	Airtronic permetester, ASTM D6476
3	Water vapour permeability	Water vapour permetester, ASTM E9680
4	Tearing strength	Instron , ASTM D2261
5.	Wicking	Vertical wicking test, ASTM D1777
6.	Thickness	Thickness tester, ISO 3616

RESULTS AND DISCUSSION

Optimization of Enzymatic Delignification of Fibers with Box- Behnken Experimental Design

Based on runs generated as in the Figure 1, the 15 delignified samples were processed. The various prepared delignified samples of areca nut were tested for absorbency in the fiber stage and the values are tabulated in the Table 4, given below. The samples were analyzed using the BOX-BEHNKEN experimental design, with fiber absorbency as the dependant variable

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Figure 1: Box-Behnken Optimized 15 Runs
Table 4: Absorbency Values Of Delignified Samples

Alkali %	Cooking time(Whistles)	Time in Hours	Absorbency
5.0	40	5	550
10.0	30	5	400
7.5	35	5	368
7.5	35	5	368
5.0	35	6	580
10.0	35	6	420
7.5	30	4	320
5.0	30	5	450
7.5	40	6	410
10.0	35	4	380
7.5	35	5	365
7.5	30	6	380
5.0	35	4	400
10.0	40	5	350
7.5	40	4	345

The delignified fibers have recorded fiber absorbency ranging from 320 -580. Majority of fibers have recorded fiber absorbency from 345 to 450. The regression coefficients for the absorbency % were estimated and later the delignification process was optimized using the response optimizer tool. The R^2 value achieved was about 97.62% and the estimated Regression Coefficients for absorbency is given below as:

Equation for Absorbency = 367.00-53.75A+13.125B+43.125C+75.87A^2 -5.375B^2 -37.500AB-35.000AC+1.25BC
Where: A is = Alkali%, B is = Cooking time (In whistles) and C is = Time in hours

Table 5: Parameters Input To Find The Global Solution In Box-Benkhen Experimental Design

Goal	Lower	Target	Upper	Weight	Import
absorbency-Target	320	550	580	1	1

With the above inputs the Global Solution for achieving a target of 550 fiber absorbency, is alkali % = 5.71578, cooking time = 40, delignification time = 6, with Predicted Responses as Absorbency % = 550.000, desirability = 1.000000, Composite Desirability = 1.000000**.** This is pictorially represented in the figure 2 given below.
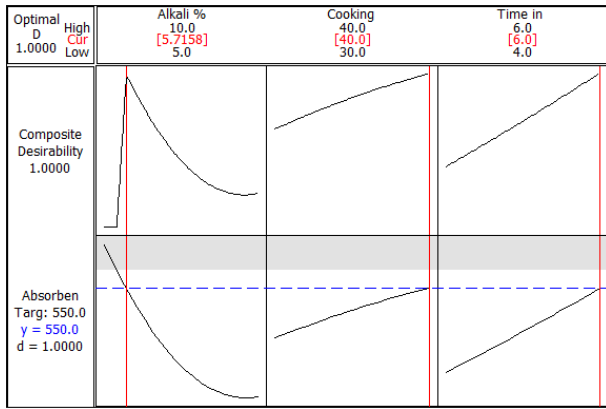
Figure 2: Response Optimization For Absorbency Target 550

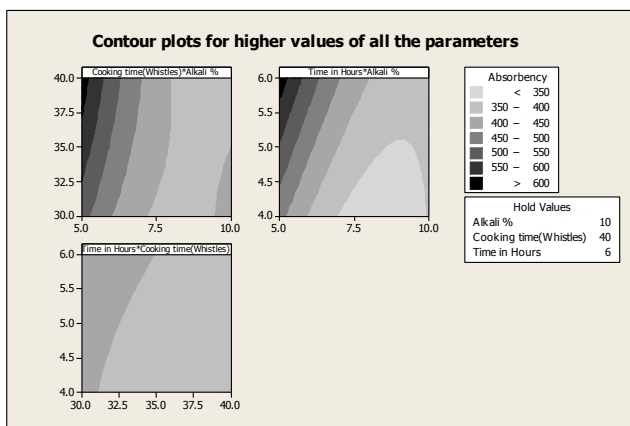## CONTOUR PLOTS TO ANALYZE THE DELIGNIFICATION OF ARECA CATECHU FIBERS:

Figure 3: Analysis Of Contour Plots For Higher Values Of All The 3 Parameters.

The figure 3 shows the contour plots with higher values of all the 3 parameters. When the time for delignification has been maintained high, maximum values of cooking time in combination with lower value of alkali have prepared the areca catechu fibers with maximum absorbency % more than 600. Fibers prepared with medium values of cooking time and alkali values have shown the least absorbency % lower than 350. Similar behavior has been observed when the alkali cooking time has been maintained high. A higher value of alkali has not showed good fiber absorbency.

When analyzing the contour plots for middle values of all the 3 parameters given in the figure 4, higher fiber absorbency values are observed in the contour plots with cooking time held at medium values and with higher values of delignification time and lower alkali %. The other
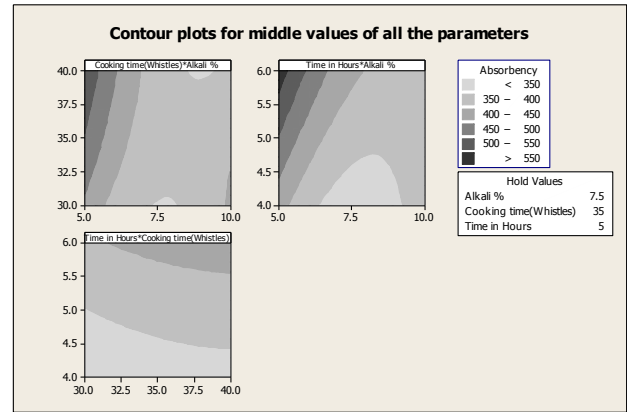

Figure 4: Analysis Of Contour Plots For Middle Values Of All The 3 Parameters.
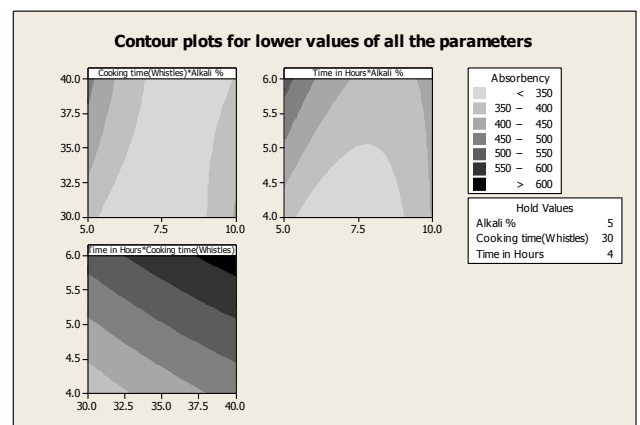

Figure 5: Analysis Of Contour Plots For Lower Values Of All The 3 Parameters.

parameters namely delignification time and alkali maintained at middle values have given fibers with medium and low fiber absorbency %

Figure 5 presents the contour plots with all the 3 parameters held in lower values. Lower values of alkali %, when used in combination with higher values of alkali cooking time and time for delignification give fibers with higher absorbency above 600. The other 2 contour graphs presenting the lower values for alkali cooking time and time for delignification have recorded medium fiber absorbency only. In order to achieve good absorbency of delignified areca catechu fibers, lower values of alkali % and higher values of alkali cooking time are required for alkali pretreatment of fibers and require higher values time required for enzyme delignification. The highest range of fiber absorbency will be around 600.

Dimensional and comfort properties of prepared fused nonwoven samples:

The Table 6 presents dimensional and comfort properties of prepared fused nonwoven samples. PP-SB and PP-F are 100% polypropylene (PP) and both are least in GSM when compared to the blended fibers. When comparing between PS-SB sample and PP-F, they differ in the manufacturing

method, one being spun bond and the other being fused. Still the difference in the GSM of both samples is negligible.

Table 6: Dimensional And Comfort Properties Of Prepared Fused Nonwoven Samples

Samples	GSM (g/sq.m)	Thickness (mm)	WVP (gm/m2/day)	Air Permeability (l/min)	Strength (MPa)	Wicking (sec)
PP-SB	21.14	0.39	1261.08	64.1	68.37	135
PP-F	20.69	0.12	1289.32	45	103.33	150
AC-PP-50	29.65	0.31	1314.14	58.3	101.34	193
DAC-PP-25	26.45	0.22	1350.12	65.3	105.43	180
DAC-PP-50	28.25	0.25	1342.46	56.5	102.56	185
DAC-PP-75	30.26	0.36	1335.36	45.7	98.37	200

The GSM of the blended samples ranges from 26.45 grams/square meter to 30.26 grams/square meter. More the presence of the Areca Catechu fiber more is the GSM. The GSM of AC-PP-50 is greater than DAC-PP -50. This is due to the fact that the alkali pre treatment and delignification process has removed more quantity of hemicellulose and lignin in the case of DAC-PP -50.

When comparing the thickness of the prepared nonwoven samples, PP-SB is 0.4 mm which is higher than that of PP-F sample which is only 0.12mm. PP-F has recorded the least thickness among all the samples. This is due to the difference in the manufacturing method of both the samples. Spun-bond method of nonwoven preparation, binding points which integrate the fibers, are present only at patterned intervals where the melting happens. Due to this the PP-SB have more freedom and therefore have more air space. Due to this reason the PP-F sample is lesser in thickness.

All the blended samples have recorded thickness in the range of 0.22mm-0.36mm which are much thicker than the PP-F samples but are very much similar to the thickness of PP-SB nonwovens which is almost 0.4 mm thickness. This is due to the thickness of the Areca catechu natural fibers. As the natural fibers are not thermoplastic, they do not melt and will help to maintain the air spaces as in spun bond method of nonwoven manufacturing

The results for water vapor permeability (WVP) of the fused nonwovens are higher when compared to the spun bonded sample. This is due to the reason that fused samples have more gaps in them and due to the absorbency of natural fibers. The highest WVP has been recorded by DAC-PP-25 sample with 1350.12 gm/m^2/day. The least WVP has been recorded by PP-SB sample with 1261.08 gm/m^2/day.

The highest Air permeability has been recorded by DAC-PP-25 sample with 65.3 L/Min. The least Air permeability has been recorded by PP-F sample with 45 L/Min. PP-SB have more thickness but have bonding points at regular intervals which gives more freedom to the fibers and allow more air to pass through but PP-F does not allow so much air to pass. In the case of blended samples, more the

presence of natural fibers, there is less air permeability. This is due to the fact that PP fibers are finer than the Areca Catechu fibers.

When analyzing the tearing strength of the prepared samples, all the fused samples have shown higher strength ranging between 103.33 MPa to 98.37 MPa, when compared to the PP-SB with 68.37 MPa, which is a spun bonded nonwoven. This is due to the reason that spun bond has less number of binding points, where fibers are melted. But fused samples have more points where fibers are melted, which enhance the tearing strength of the fused samples. When comparing between the blended fibers, more the presence of the natural fibers, there is a decrease in the tearing strength of the fused samples.

When comparing the wicking of all the samples, PP-SB has the least time of 135 seconds followed by PP-F with 150 seconds. All the fused samples have recorded higher time for wicking than both the 100% PP fibers. The wicking time has increased with increase in the Areca Catechu fiber content ranging from 193 to 200 seconds. Though the absorbency of the Areca catechu has improved due to the delignification process, this has not helped in improving the wicking of the natural fibers equivalent to perform like the PP fibers. This is viewed as a limitation of the fused samples. But DAC-PP -50 has recorded much lesser than the AC-PP-50 sample.

CONCLUSION

Estimated Global Solution as per BOX-BEHNKEN experimental design for delignification of Areca Catechu fibers with laccasse enzyme to get 550 as the Target for fiber absorbency is 40 as the cooking time and 6% NAOH concentration for alkali pre-treatment in an auto clave and 6 hours as the time of delignification with Lacasse enzyme at 6% concentration. These 3 variables will give a processed fiber with 550 as the fiber absorbency. When analyzing the contour plots of the 15 runs as per BOX-BEHNKEN experimental design, we understand the relationship between the 3 parameters. In order to achieve good absorbency of delignified areca catechu fibers, lower values of alkali % and higher values of alkali cooking time are required for alkali pretreatment of fibers and require higher values time required for enzyme delignification. The highest range of fiber absorbency will be around 600

During the fusing process there is an even application of temperature and pressure on the web due to which more fibers are subjected to melting. Due to this reason the PP-F sample is lesser in thickness. All the blended samples have recorded thickness very much similar to the thickness of PP-SB nonwovens. This is due to the thickness of the Areca catechu natural fibers. So the application of Areca Catechu fibers with PP fibers and fusing them to form a nonwoven sheet has helped in preparing a nonwoven which is similar to the thickness of PP-SB commercially available polypropylene non-woven. As the natural fibers are not thermoplastic, they do not melt and will help to maintain the air spaces as in spun bond method of nonwoven manufacturing.

PP-SB have more thickness but have binding points at regular intervals which gives more freedom to the fibers and

allow more air to pass through but PP-F does not allow so much air to pass. In the case of blended samples, more the presence of natural fibers, there is less air permeability. This is due to the fact that PP fibers are finer than the Areca Catechu fibers.

Delignification has improved the water vapour permeability, Air permeability, strength and wicking of the delignified Areca catechu/PP blended nonwovens samples when compared to the raw Areca catechu/PP blended nonwovens samples. This study on preparing fused nonwoven samples from PP and Areca Catechu blended fibers has introduced a new functionality to the agri-waste Areca Catechu husk fibers. This study has paved way to sustainability as the waste has been recycled and has been processed in eco-friendly methods. Combining bio-based materials with PP fibers to develop an effective product having many applications is a better way to follow sustainability [25]

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