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Impact of Storage Time on Chemical Constituents, Pulp and Papermaking Properties of Bamboo

Abstract: *Significant changes take place in cellulosic raw materials during storage over a period of time. Present study was carried out to quantify the impact of bamboo storage on chemical constituents of bamboo, response during pulping, bleaching, refining, morphological and physical strength properties of bleached bamboo pulp. Freshly harvested bamboo (Melocanna baccifera) collected from North-East India was stacked for a period of one year in open atmosphere to simulate mill conditions. Samples were collected after every two months interval for detailed analysis. After one year storage of bamboo, loss in biomass was 8.8% and decrease in hemicelluloses and extractives was from 21.1% to 18.9% and 7.7% to 4.8%, respectively. One percent sodium hydroxide solubility was increased from 19.9% to 27.3% whereas unbleached pulp yield was reduced from 51.6% to 46.3% due to decay in holocellulose with marginal reduction in fiber length. Tensile, tear and burst indices of final bleached pulp were reduced substantially with the storage by 18%, 25% and 15%, respectively. The results of the study revealed that storage of bamboo is not advisable beyond 6 months period due to the significant reduction in biomass, pulp yield, brightness, viscosity and physical strength properties of bleached pulps.*

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Key Words: *Bamboo, Storage losses, Hemicelluloses, Extractives, Pulp yield, Coarseness.*

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Introduction

Bamboo is a fast growing and wide spread natural resource belonging to grass family Poaceae. The pulp made from bamboo is suitable for making almost all type of papers like writing and printing, packaging, tissue and duplex board (1). India is one of the largest bamboo growing country in the world having 136 species of 23 genera grown on 15.69 million ha area (2) producing about 4-6 million tonnes of bamboos annually, out of which 2.2 million tonnes is used in the paper industry (3). The bamboo species reported to be used for making pulp and paper are *Bambusa balcooa Roxb., Bambusa vulgaris, Bambusa stenostachya Hackle, Bambusa tulda, Bambusa bambos, Dendrocalamus strictus, Dendrocalamus giganteus, Dendrocalamus homiltonii, Melocanna baccifera and Neosinocalamus affinis* etc. (4). Due to the non- availability of bamboo in the monsoon period, mills are storing the bamboo for considerable period of time extended up to 12 months depending on the harvesting period and availability in the particular region. Storage of raw materials helps the mills by providing continuous supply of material for uninterrupted production and capability for uniform blending of different types of fibrous raw materials in appropriate mix for producing similar quality of end products.

During the storage period change in chemical constituents of fibrous raw material takes place due to auto-oxidation with heat and ultraviolet light in presence of oxygen and moisture. Like wood bamboo is susceptible to attack by similar microrganisms. Deterioration may also be caused by insects, microorganisms like mould, stain and rot fungi and some bacteria. During storage the problem is created by rot fungi more; as stain and mould fungi get inhibited when bamboo gets dried. Insects like beetle, termites utilize the cellulosic fibers as their food sources resulting into its degradation. These factors combined together cause adverse impact on the pulp and paper making properties to a large extent. Decay of bamboo/ wood during storage also leads to losses of biomass and occupies the same volume as sound materials. Therefore, lesser amount of degraded raw material in comparison to sound material is fed to the digester during pulping process leading to loss in production capacity. Earlier, different researchers have reported the behaviour of fibrous raw materials specifically wood based raw materials with respect to their storage period and its impact on the chemical constituents of that raw material and pulp produced from it (5-7).

Very little information is available on relationship of storage of bamboo with changes in chemical constitution and its impact on pulp and paper making properties. Moreover, the results of the investigations are also quite contradictory, one study indicates that there were no appreciable differences in unbleached and bleached pulp yield between fungal attacked and sound bamboos after storage (8) whereas another study concluded about 25% loss in pulp yield and 15-40% loss in pulp strength due to fungal attacked bamboo as compared to sound bamboo (9). Another researcher reported increased alkali demand during pulping for stored fungal attacked bamboo and higher bleaching chemical consumption in comparison to fresh raw material (10).

Storage of raw materials has some advantages as well as disadvantages for the pulp and papermaking. Advantages include decrease in extractives and foam generation during brown stock washing operation. Storage of wood for a period of 12 months was reported to reduce the extractives content by 60 to 80% as compared to that of the fresh material (11). It was also reported that the extractives content reduced rapidly in the initial period of storage specifically during first 60 days after harvesting (6). Reduction in extractives in the stored wood also decreased the pitch problem during papermaking process (5) and reduced the refining energy requirement (12). The disadvantages of wood storage include 4-15% loss of biomass, less pulp yield, poor delignification, lower brightness and physical strength properties (13-14). It was also reported that the severe loss occurred when wood or wood chips were stored for a long period (15).

This paper summarized the effect of storage on changes in chemical constituents, pulping behavior, bleaching response, refining behavior, morphological properties, and physical strength properties of bamboo. Results of this study may help the mill personnel to decide the length of storage for bamboo to avoid excessive decay.

Materials and Methods

Material

The study was carried out on bamboo species *Melocanna baccifera* procured from northeast part of India. Experimental stack of bamboo was prepared with seven rows of known weight. After every 2 months one row was dismantled for the evaluation of raw materials and quality of pulp. Moisture content and basic density of bamboo was determined as per TAPPI T 258 om-11. Bamboo samples were chipped in the drum chipper, mixed thoroughly and kept in a polythene bag for using it in further studies.

Analysis of chemical constituents

Bamboo chip samples were analyzed for proximate analysis after grinding in Wiley mill and screening on 40 mesh sieve. Powdered material was characterized for 1% NaOH solubility, extractives and lignin content as per TAPPI test methods T 212 om-12, T 204 cm-07, T 222 om-11, respectively. Cellulose and hemicelluloses in the bamboo stored for different periods were analysed using methods described by Updegroff (16) and Deschatelets and Errest (17), respectively.

Pulping

Kraft pulping of bamboo chips was performed in lab autoclave digester consisting of six bombs of 2.5 L capacity maintaining following pulping conditions: Active alkali as Na_2O – 15.5%, sulphidity – 23.4%, bath ratio – 1:3, cooking temperature – 160°C, cooking time – 60 min.

The cooked mass was disintegrated and screened on 0.15 mm slotted screen using Somerville type equipment as per TAPPI T 275 sp-12. The screened pulp was analysed for kappa number, brightness and viscosity as per TAPPI test methods T 236 om-06, T525 om-12 and T 230 om-08, respectively. The black liquor generated during pulping was analyzed for pH, Total Solid, Residual Active Alkali, Swelling Volume Ratio (SVR), Gross Calorific Value (GCV) following the methods IS:3025 (Part11), TAPPI T 650 om-09, Oye et al. (18), T 625 cm-85, T 684 om-11, respectively.

Bleaching

Unbleached bamboo pulp was bleached using $C_DE_{OP}D_1D_2$ sequence targeting to attain final pulp brightness of about 88% ISO, where, CD – Chlorine with partial (20%) substitution with chlorine dioxide, D_1 and D_2 - chlorine dioxide, E_{OP} - extraction stage using sodium hydroxide, oxygen and hydrogen peroxide. Details of bleaching conditions maintained are provided in the appropriate table.

Morphological properties

Morphological properties of the pulps were analyzed using L&W fiber tester. Average value obtained for different parameters were reported.

Physical strength properties

Pulps were refined as per TAPPI T 248 sp-08 to different freeness levels. Freeness of pulp was measured as per TAPPI T 227 om-09. Laboratory handsheets of 60 g/m^2 were prepared from refined and unrefined pulps as per TAPPI T 205 sp-06. Different physical strength properties were analysed as per TAPPI T 220 sp-10.

Results and Discussion

Moisture and weight loss during storage

Detailed results of moisture and weight loss during storage of bamboo are given in Table 1. Moisture loss in the bamboo or any fibrous biomass during storage depends on weather conditions. During this study period bamboo released the moisture quickly from 43.7% to 16.8% after first two months of storage period. The moisture loss became stagnant after 8 months of storage period to about 11% till the end of 12 months period. Sudden rise in moisture level at the end of 6th month was due to rainfall and moisture absorption by the bamboo. The extent of biomass loss during first six months storage period was lower compared to loss of biomass after the 6th month onwards. After 12 months of storage period 8.8% biomass was lost/ deteriorated on oven dried mass basis. This may be attributed due to the volatilization, auto-oxidation, microbial decay individually and their collective impact on the fibrous material are responsible for deterioration/ weight loss of material (19-20).

Table 1: Moisture and weight loss in bamboo at different storage period

Particulars	Storage period (month)					
	2nd	4th	6th	8th	10th	12th
Average moisture at the time of stacking (%)	43.7					
Average moisture at the time of de-stacking (%)	16.8	11.5	23.3	12.8	11.3	11.0
AD quantity loss in particular row (%)	33.3	37.4	28.3	38.0	40.1	42.5
BD quantity loss in particular row (%)	1.2	1.7	2.1	3.7	5.3	8.8

Change in chemical constituents

Basic density and bulk density of bamboo was gradually reduced from 510 kg/m^3 to 490 kg/m^3 and 208 kg/m^3 to 195 kg/m^3, respectively after storage period of 12 month (Figure 1). Degradation results in loss of weight of material through conversion of organic matters into carbon dioxide and water leading to decreased density of material (21). Based on previous literature bamboo is susceptible to attack by white rot Basidiomycetes and soft-rot Ascomycetes. Kim et al (22) claim that for the decay main culprits were T. versicolor and A. arundinis species of Basidiomycetes and Ascomycetes, respectively. Decay had little effect on the volume of wood but the extent of weight loss resulted in decreased pulp yield.

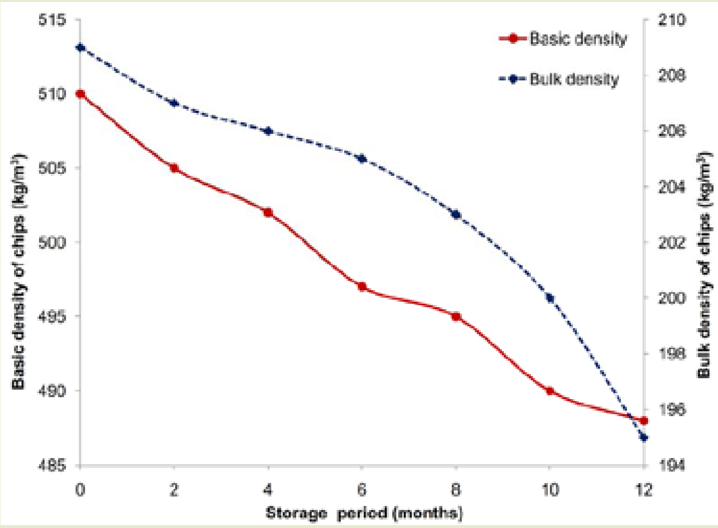

Figure 1: Effect of storage period on basic and bulk densities of the bamboo chips

One percent NaOH solubility, which indicates the extent of deterioration of raw material was increased from 19.5 to 27% after 12 months of storage of bamboo. Previous studies by many researchers on hardwood reported the same trend. On storage of eucalyptus for twelve months one percent NaOH solubility was increased from 14.65 to 17.20 % (7). Different other hardwoods also showed the same tendency, one percent NaOH solubility was increased after nine months storage period (23). The total solvent-extractable substances in bamboo, which mainly consist of fatty acids, resin acids, sterols, waxes, fats, non-volatile hydrocarbons and polyphenols were decreased gradually over the storage period possibly through volatilization, auto-oxidation and free radical formation (Figure 2). Many studies revealed that that with the storage period the amount of extractive content was reduced for other raw materials (6, 24). It was hypothesized that the insects, fungi and bacteria may degrade the extractive content in the material when exposed to ordinary environment for long period (25). Loss in the extractive content is advantageous for the pulping process as lowering of extractive content reduced generation of foam during pulp washing and pitch formation.

Effect of storage period on hemicelluloses, cellulose and lignin content in bamboo is shown in Figure 3. Hemicelluloses and cellulose contents were decreased whereas lignin content was increased with the storage of bamboo. Increase in lignin content may be due to the excessive decay in carbohydrates and extractives content as compared to lignin. Loss of cellulose and hemicelluloses may be due to the conversion of some poly-oligosaccharides into mono or due to the free radical formation by UV, atmospheric oxygen and degradation by bacteria, fungi etc. Many fungi use the hollocellulose as carbon source for their growth and modifying the lignin component. Hollocelulose is degraded by brown rot fungi with oxidative mechanism (26). Hemicelluloses have the amorphous structure and can be more easily hydrolyzed compared to cellulose, this may be the reason that hemicelluloses degraded more compared to cellulose. The values of different chemical constituents are obtained in the study found to be in agreement with the earlier reported values for different bamboo species (27-28).

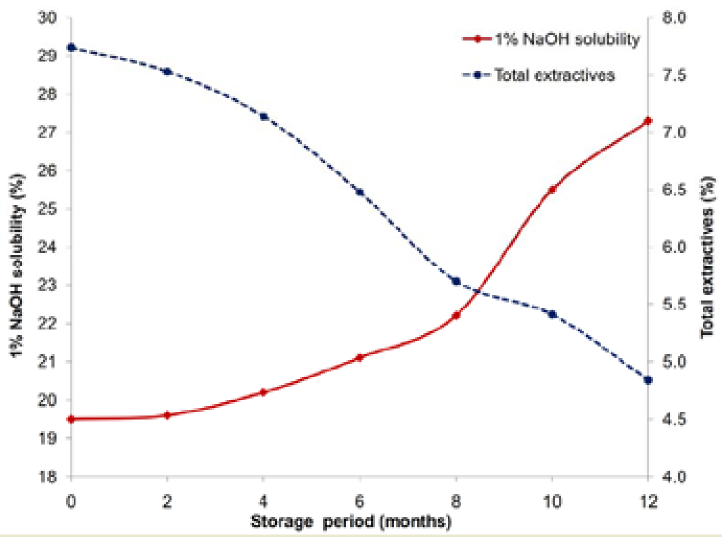

Figure 2: Effect of storage period on 1% NaOH solubility and extractive contents in bamboo

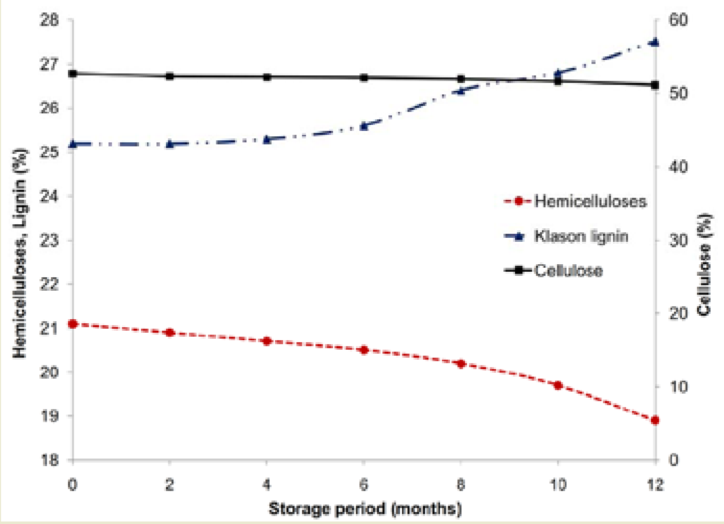

Figure 3: Effect of storage period on cellulose, hemicelluloses and lignin content in bamboo

Pulping of bamboo

Detailed pulping results of bamboo stored for different period are given in Table 2. While using similar pulping conditions kappa number of pulp was increased marginally from 19.6 to 21.6 after 12 months of storage period. Increase in kappa number while using similar active alkali for stored bamboo indicates that requirement of active alkali to produce similar kappa pulp will be higher as compared to that of fresh bamboo. Similar trends were also reported earlier by other researchers (24, 29). While using similar pulping conditions, unscreened and screened pulp yields were reduced substantially from 51.6% to 46.3% and from 51.5 to 45.7%, respectively (Figure 4). Intrinsic change in different chemical constituents during the storage period may result in lower pulp yield and viscosity. Decrease in viscosity may occured due to depolymerisation of cellulose by cellulase producing microorganism (29). Degradation of cellulose increased the fine content which got loosed during screening of pulp and resulted in lower pulp yield (24). Brightness of unbleached pulp showed the negative trend as the kappa no of pulp was increased. Total solids of black liquor were increased with increase in storage time. This might be due to the degradation of low molecular weight hemicelluloses and cellulose that gets solubilised during pulping process. Increased swelling volume ratio (SVR) values indicating that organic content had increased in black liquor with storage time.

Low amount of free alkali in the black liquor also indicated the presence of acidic compounds formed during the fungal decay of cellulosic/lignin components which may consume a part of the active alkali. The black liquor generated with stored bamboo have low GCV as compared to fresh bamboo indicating that more hemicelluloses, which have relatively lower calorific value than lignin, were dissolved in black liquor.

Table 2: Pulping results of bamboo at different storage period

Particulars	Storage period (month)						
	0	2nd	4th	6th	8th	10th	12th
Properties of the pulp							
Unscreened pulp yield (%)	51.6	51.1	51.0	50.1	49.6	48.6	46.3
Rejects (%)	0.1	0.2	0.3	0.4	0.4	0.6	0.6
Screened pulp yield (%)	51.5	50.9	50.7	49.7	49.2	48.0	45.7
Kappa no.	19.6	19.3	19.5	19.9	20.2	20.5	21.6
Viscosity (cP)	27.2	27.0	25.9	25.3	25.2	25.9	23.3
Brightness (% ISO)	25.9	25.1	24.8	24.0	23.5	23.3	22.6
Properties of black liquor							
pH	12.2	12.2	12.1	12.0	12.1	12.0	12.1
Total solids (%)	19.6	19.7	19.7	20	20.2	20.6	21.4
Free alkali (g/l) at 20% solids	7.95	7.53	7.2	6.5	6.3	5.9	5.6
SVR (ml/g)	60	65	69	75	76	76	75
GCV (kcal/kg)	3270	3100	3000	2950	2880	2850	2790

Bleaching of pulp

Detailed bleaching results along with properties of final bleached pulps are given in the Table 3. The bleachability of pulp was adversely affected with increasing storage period of bamboo. Bleaching chemical requirement was increased along with increasing storage period due to the increase in kappa number of pulp.

The brightness and viscosity of final bleached pulp at the time of stacking was 90.1% ISO and 20.0 cP, respectively, which reduced gradually with the increasing storage period (Figure 5). Earlier, researchers also achieved the bamboo pulp brightness of 83.7% ISO using ECF sequence ODED (30) and up to a brightness of 88.6% ISO by using chlorine along with chlorine dioxide in first stage of bleaching sequence OC/DED (31).

With the use of chlorine and chlorine dioxide in CD stage of bleaching at 0.22 kappa factor and similar bleaching chemicals is E_{OP}, D_1 and D2 stages of bleaching, the brightness of pulp was reduced steadily with the increasing storage period, from an initial 90.1% ISO to 87.5% ISO after 12 months of storage. The reason behind brightness drop was proposed due to the generation of chromophoric substance and condensed aromatic components during storage due to weather and enzymatic reactions of fungi with carbohydrates (32-33). Viscosity of pulp was reduced significantly, 19.5% reduction in viscosity was observed after 12 months storage period. Brightness stability of pulp was also got reduced.

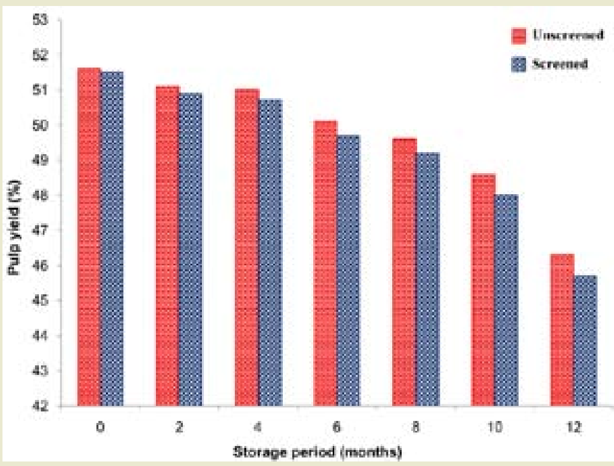

Figure 4: Effect of storage period on unscreened and screened unbleached pulp yields

Table 3: Bleaching results of bamboo pulps after different storage period

Particulars	Storage period (month)						
	0	2nd	4th	6th	8th	10th	12th
Kappa Number	19.6	19.3	19.5	19.9	20.2	20.5	21.6
C_D stage *(Consistency – 3%, time – 45 min, temperature 35°C)*							
Cl_2 added (%)	3.88	3.82	3.86	3.94	4.00	4.06	4.28
ClO_2 added (%)	0.16	0.16	0.16	0.17	0.17	0.17	0.18
End pH	1.8	1.8	1.8	1.8	1.9	1.9	1.9
E_{OP} stage *(Consistency – 10%, time – 120 min, temperature 80°C, H_2O_2 –0.5%)*							
NaOH added (%)	2.4	2.3	2.4	2.4	2.4	2.5	2.6
End pH	11.4	11.4	11.3	10.9	10.8	10.7	10.8
D_1 stage *(Consistency – 10%, time – 180 min, temperature 75°C)*							
ClO_2 added (%)	0.9	0.9	0.9	0.9	0.9	0.9	0.9
End pH	3.5	3.4	3.4	3.2	3.4	3.4	3.5
D_2 stage *(Consistency – 10%, time – 180 min, temperature 75°C)*							
ClO_2 added (%)	0.3	0.3	0.3	0.3	0.3	0.3	0.3
Final pH	3.5	3.6	3.5	3.5	3.45	3.8	3.45
Properties of final bleached pulp							
Brightness (%ISO)	90.1	90.1	89.6	89	88.6	88.2	87.5
P.C. no.	0.51	0.58	0.63	0.61	0.6	0.56	0.66
Viscosity (cP)	20	20.1	19.4	18.9	18.2	17.6	16.1

Morphological properties

Effects of storage period on different fiber morphological properties like average fiber length, fiber width, vessels, coarseness, fines content and kink etc. are given in Table 4. At the time of stacking the average fiber length of bleached bamboo pulp was 1.72 mm, width 17.1 µm; which is similar to the earlier reported values for bleached bamboo pulps (34-35). With the storage average fiber length was marginally reduced from 1.72 mm to 1.68 mm and average fiber width remained unaffected. Coarseness of the bamboo fibers was reduced substantially with the storage period (Figure 6) may be due to the depletion of cellulose chain length as evident from loss in pulp viscosity.

Table 4: Morphological properties of bleached bamboo pulp at different storage period

Particulars	Storage period (month)						
	0	2nd	4th	6th	8th	10th	12th
Average fiber length (mm)	1.72	1.72	1.71	1.71	1.68	1.68	1.68
Average fiber width (µm)	17.1	16.9	17.1	17.0	17.2	17.1	17.1
Vessels per lacs fiber (no.)	146	136	143	100	121	104	155
Coarseness (µg/m)	103	101	100	97	92	89	87
Fines, mass basis (%)	3.9	4.1	4.1	4.1	4.1	4.2	4.3
Mean kink angle	56.9	58.5	56.3	56.5	56.8	60.8	60.3
Number of kinks per fiber	0.756	0.822	0.799	0.768	0.762	0.924	0.854
Mean kink index	1.556	1.837	1.538	1.545	1.551	1.903	1.858

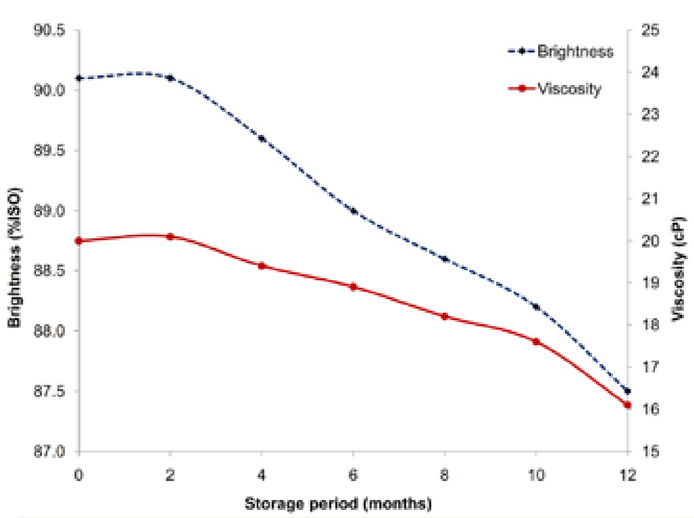

Figure 5: Effect of bamboo storage period on bleached pulp brightness and viscosity

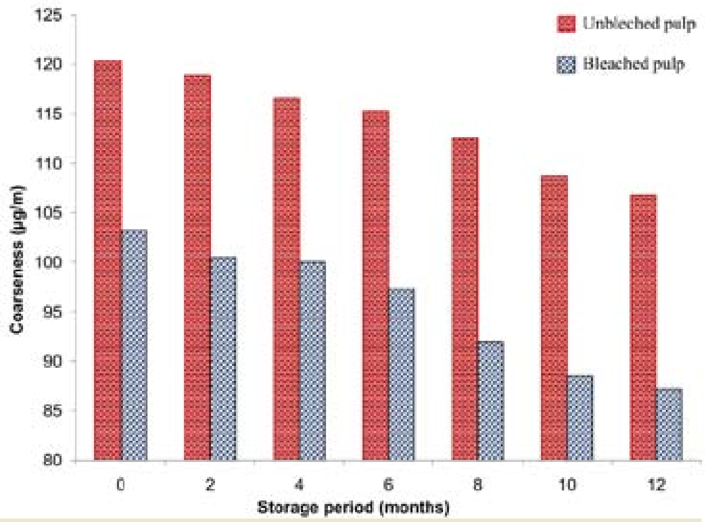

Figure 6: Effect of storage period on coarseness of unbleached and bleached pulps

Refining of pulp

Bleached bamboo unrefined pulp has Canadian standard freeness of about 710 ml CSF. The pulps were refined in PFI mill at different freeness levels by applying 1800, 3100 and 4100 PFI revolutions to get refined pulps of about 600, 500 and 400 ml CSF (Figure 7). It was observed that pulps produced from stored bamboo had lower freeness at the same refining levels as compared to that with fresh bamboo. This may be due to the deterioration of cellulosic fibers (evident from lower pulp viscosity), reduced cellulosic material in the fiber cell wall (evident from lower coarseness of fiber), smaller fiber length, more fines in the pulp (evident from morphological properties).

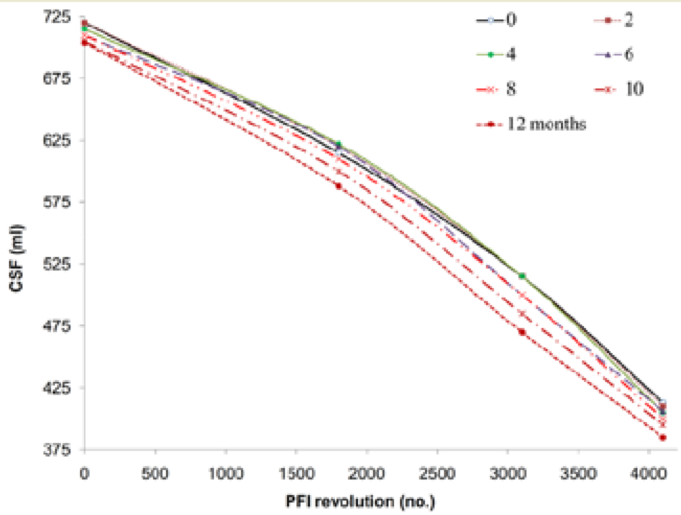

Figure 7: Effect of storage period on refining behavior of bleached bamboo pulp

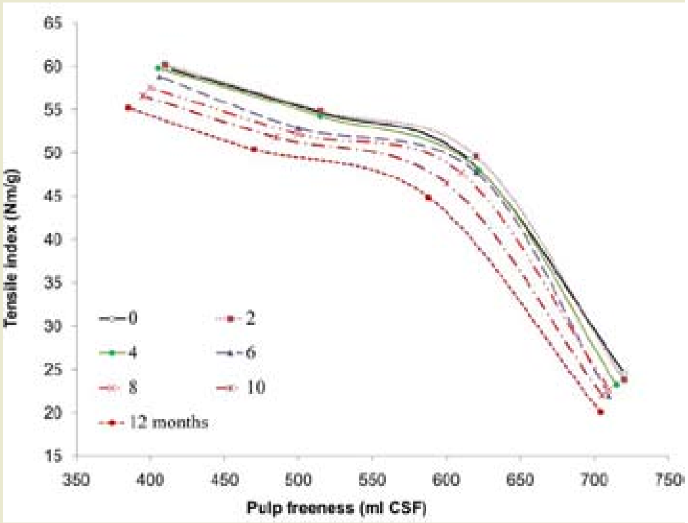

Figure 8: Effect of storage period on tensile index of bleached bamboo pulp

Tear index of pulp was initially improved by refining the pulps to about 600 CSF afterwards it got reduced at freeness levels of about 500 and about 400 ml CSF. Tear index of pulp was also comparable up to a storage period of 6 months; afterwards it was reduced significantly up to 25% by the 12th month of storage (Figure 9).

Burst index of pulp was improved with the refining (reduction in freeness values of pulp) for different pulps obtained from pulping of bamboo stored for different periods. Burst index of pulp was also comparable up to a storage period of 6 months; afterwards it was reduced up to 15% by the 12th month of storage (Figure 10).

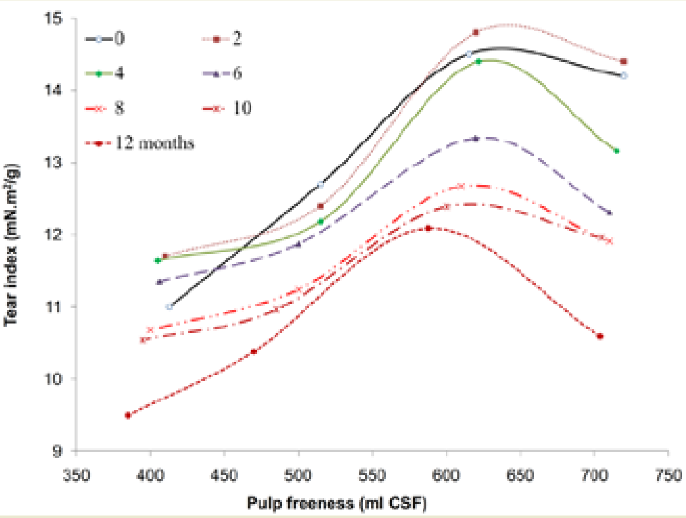

Figure 9: Effect of storage period on tear index of bleached bamboo pulp

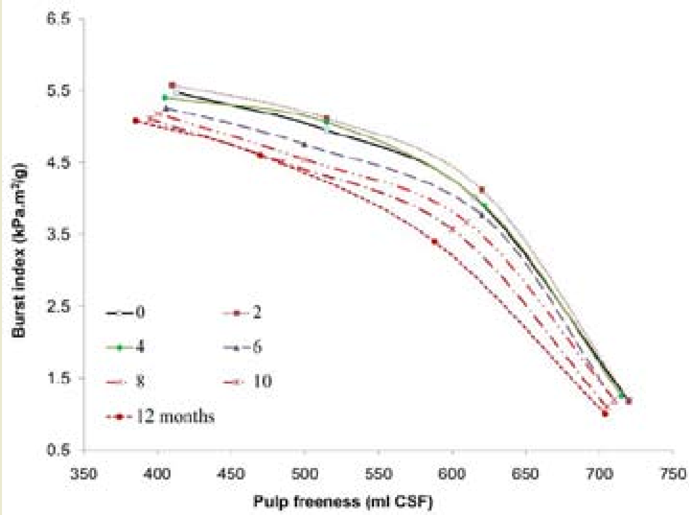

Figure 10: Effect of storage period on burst index of bleached bamboo pulp

After storing the raw material for a period of one year the losses in physical strength properties of bamboo pulps (18%, 25% and 15% for tensile, tear and burst indices, respectively) are comparatively higher than the reported values of losses in physical strength properties of debarked eucalyptus pulp i.e. 12%, 14% and 12% for tensile, tear and burst indices, respectively (7).

Conclusions

The results presented from the study revealed that bamboo stack stored for 12 month in open atmosphere got degraded significantly after a period of time. Bamboo did not loose significant intrinsic fiber properties and the biomass when stored up to a period of 6 months, afterwards, the decay of raw material became severe. The loss of biomass was increased substantially to 8.8% on oven dried mass basis after 12 months of storage period. Extractives content and hemicelluloses content in bamboo were reduced significantly along with the storage period. After six month storage chemical constituents like cellulose and hemicelluloses were also decreased significantly. Due to degradation of cellulose, pulp was produced with inferior physical strength properties. Therefore beyond 6 months of storage period it is not advisable to store for bamboo due to the significant loss in biomass, unbleached pulp yield, bleached pulp brightness, viscosity and physical strength properties etc.

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Conflict of interest

None

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