ISSN: 0379-5462
IPPTA: Quarterly Journal of Indian Pulp and Paper Technical Association
Vol. 35, E2, 2023, p. 92-96

EXPLORING THE POTENTIAL OF BACTERIAL CELLULOSE AS A REINFORCING ADDITIVE FOR ENHANCED STRENGTH IN FOOD PACKAGING PAPERS




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Abstract:

Bacterial cellulose (BC) is emerging as an important biodegradable material that can be used in a wide range of applications. Due to its high purity, fine fiber structure, high porosity, and high surface area, BC has emerged as an attractive area to study its application in day-to-day use. One such area where bacterial cellulose can be helpful is paper or paper-based products. Paper made from Wood and Agro-based raw materials generally has fewer barrier properties, which limits its variety of uses in packaging food or fluid materials. To enhance the barrier properties of paper for use in food packaging, BC can be applied as an additive to these papers. In the current study, microbes have been used to synthesize cellulose under laboratory conditions, and the synthesized BC has been characterized by XRD, FTIR, and SEM. The BC synthesized in the laboratory was used in the wet-end of papermaking in two different ways using recycled pulp. Firstly, the BC was used alone in the wet-end of papermaking, and secondly, the BC was blended with cationic starch (CS) and then used in the wet-end of papermaking. The results showed that the use of BC, along with cationic starch at a very low dose of 0.8% (with respect to CS), increased the efficiency of CS as a dry strength additive in the wet-end of papermaking by enhancing the breaking length, tear index and burst index by 33.0 %, 2.53% and 63%, respectively in comparison to the strength properties of the paper made using only pulp. The study has shown the potential of the use of BC as an additive for enhancing strength properties in food packaging paper.

Keywords: Bacterial Cellulose, Paper and Pulp, Strength Property, Wet-End of Papermaking, Cationic Starch

1. INTRODUCTION:

Cellulose is a fibrous material which is tough in nature and water-insoluble polymer. Cellulose plays an essential role in maintaining the structure of plant cell walls. Moreover, cellulose is a biodegradable, biocompatible, and renewable natural polymer and hence it is considered an alternate to nondegradable fossil fuel-based polymers. The possibilities of technological use of cellulose are enormous, however, obtaining pure cellulose, i.e., separating it from the encrusting components requires a chemical treatment which is non-environment friendly, non-cost effective and requires lot of energy. Plant is the major contributor of cellulose but nowadays, alternative source of cellulose is bacteria due to increasing demand on derivatives of plant cellulose had increased wood consumption as raw material, also causing deforestation and global environmental issue.

In addition to plants, certain microorganisms have the ability to produce cellulose. Bacterial cellulose was discovered by Brown in the year 1886. He identified the growth of unbranched pellicle with chemically equivalent structure as plant cellulose. Bacterial cellulose is an organic compound produced by certain types of bacteria. Firstly, bacterial cellulose was produced from Acetobacter xylinum and also produced by some Gram-negative bacteria's such as Acetobacter, Rhizobium, Agrobacterium, Azotobacter Pseudomonas, Salmonella as well as Gram-positive bacterial species such as Sarcina ventriculi (Nguyen et al. 2008). The molecular formula of BC is $(C_6H_{10}O_5)$ n, it have a β-1,4 linkage between two glucose molecules, which is same as of plant cellulose, but differ to physical and chemical features such as in the highly order structure of these celluloses. Unlike the plant cellulose, BC is free of lignin, hemicelluloses and extractives and is characterized by high purity. BC can be produced by culturing a strain of Acetobacter xylinum, reclassified as the genus Gluconacetobacter, which is typically found on fruits, decaying fruits, vegetables, vinegar, fruit juices, and alcoholic beverages (Nguyen et al. 2008). BC has been used in multiple field such as. Due to its biocompatibility, BC has found several applications in biomedical industry, paper restoration, wound dressing, blood vessel regeneration, artificial skin, and food industries (Shah and Brown 2005; Klemm et al. 2006; Hong and Qiu 2008; Kurosumi et al. 2009; Zeng et al. 2011; Janbade et al. 2022). In addition, its nano structure and presence of large number of free hydroxyl groups make BC an excellent biomaterial for use in the Pulp & Paper Industry (Jonas and Farah 1998; El-Saied et al. 2004; Chawla et al. 2009; Cheng et al. 2011, Janbade et el. 2022).

Studies conducted elsewhere have demonstrated the use of BC as strength reinforcing additive for production of high strength papers, improving strength properties of secondary fibre and agro residue papers and also for improving barrier properties of food packaging paper products. BC is also known to improve the performance of AKD sizing (Yamanake and Watanabe 1995). In view of the above, current study focuses on BC for use as strength reinforcing additive for enhancing the strength property of food packaging papers.

2. MATERIALS AND METHODS

2.1 Materials

Fruits, Tea and Sugar was bought from local market. Sodium Hydroxide was obtained from Sigma-Aldrich. The recycled pulp used in the study for wet-end of papermaking was prepared in laboratory by recycling using old corrugated containers. The retention aid, a medium to high molecular weight cationic polyacrylamide (CPAM) was donated by a chemical supplier of Delhi, India. Wet-end chemicals used were: Alkyl ketene dimer (AKD) collected from Sood Resins and Polymers, Himachal Pradesh; All chemicals used were of high purity and analytical grade. Cationic starch (CS) of 0.020-0.025 degree of substitution procured from Bharat starch Industries Limited, Yamuna Nagar.

2.2 Production of Bacterial cellulose

Fruits (banana, orange, tomato) taken in a tray and crushed. Added distilled water and sugar in the ratio of 1:2:0.3 respectively and incubated at 30˚C for 7 to 14 days to see mat like structure as top layer. Further, one litre of distilled water in a beaker was taken and boiled. Then mixed with three tablespoon tea and 100g sugar with continuous stirring. Allowed it to cool for 30 min. Filtered the tea with the help of muslin cloth and transferred into separate sterile beakers. In beakers, BC pellicles from fruit fermentation were mixed and covered with sterile muslin cloth and further incubated them at 30^{0}C for 10 to 15 days to see mat like thick BC layer on top.

2.3 Cleaning process

BC pellicle harvested from tea fermentation contains lots of microbes and media components. Thus, pellicles must be cleaned by first boiling the pellicle in water to kill active microbes, and then treated with alkali medium. The gel like pellicle were boiled and cut into regular pieces. And then treated four times in 0.1M aqueous NaOH at 90oC for 20 min. After alkali treatment, pellicle increased their transparency and becomes white in color, finally the BC pellicles were neutralized with DI water overnight. They were shaken regularly, and the water was refreshed every 6 hours until the pH reached a neutral value. After alkali treatment and washing in distilled water BC can be dried. After the cleaning, the pellicle changed colour from brownish yellow to milky white, indicating the most of the medium and microbes were removed.

Further yield (g/l) of BC production was calculated by dividing Dry BC membrane weight(g) to Volume of culture medium used (l).

2.4 Characterisation

2.4.1 SEM - The surface morphology of dried BC layer was done using scanning electron microscope (TESCAN, MODEL-MIRA 3 LMH, USA). The scanning electron micrographs of BC were obtained at 1000X magnifications to analyse the surface morphology. For performing the SEM of BC, the samples of size 1 × 1 cm^2 were cut and coated with 5 nm thick gold film before scanning under the SEM microscope.

2.4.2 ATR-FTIR - ATR (Attenuated total reflectance) - FTIR spectra of BC sample was obtained by means of Frontier MIR LiTa/KBr/AI spectrometer (PerkinElmer, UK).

2.4.3 XRD - XRD analyser (RIGAKU, MODEL-Ultima IV, Japan) was used to record XRD pattern. X-ray diffraction analysis of the materials produced in the diffraction angle range 2θ = 0–80°.

2.5 Wet End Application of BC in Increasing Strength Properties of Paper

2.5.1 Preparation of CS alone and with BC for wet-end application

To cook the slurry of 1% (w/v) CS in distilled water, it was placed in a water bath at 50ºC with a gradual increase in temperature and continuous mild stirring. The slurry was then kept at 90ºC for about 30 minutes and cooled at ambient temperature prior to use as a strength aid. Similarly, the cooking was done for the blend of BC and CS. To prepare 0.1% (w/v) of CPAM, it was dissolved in distilled water at 40ºC and kept for 30 minutes at 300 rpm.

2.5.2 Characterization of hand-sheets prepared

Paper hand-sheets were prepared on a laboratory hand-sheet former as per TAPPI T 205 sp-02. The grammage (g/m^2) of paper hand-sheets was determined by taking weight of 200 cm^2 circular disc of paper multiplied by 50. Different paper properties were analysed according to ISO/IS standard methods. The handsheets were conditioned at 27±2°Cand 65±5% relative humidity for at least 24 h as per ISO 187: 1990. Different properties of the paper handsheets such as burst index (L & W Bursting Strength Tester, Code: 180, Type: 973156, No. 324), Breaking length, and tear index (L&W tearing tester, Code: SE009, Type: 970435, No. 5776) were determined as per standards IS: 1060 (Part 1). The experiments were done thrice and an average was taken to report the resulted value.

3. RESULTS AND DISCUSSION

3.1 Production of Bacterial cellulose

After 14 days of incubation, mat like structures were formed on the solid surface of fruit mixture and gel like layer were formed at the air-liquid interface in beaker as shown below images (Fig. 1-2). This top layer along with tea as media was stored at room temperature in glass bottles until further use as an inoculum. To calculate the yield the BC layer was dried at 60^{0}C for overnight (Fig. 3). The yield of BC produced in 2L beakers was found to be 2.7gm/L.

Figure 1: Sample of fruit fermentation

Figure 2: Bacterial cellulose layer been formed on Liquid-air interface on production media

Figure 3: Purified washed and dried BC

3.2 Characterisation –

3.2.1 SEM - The analysis using a Field Emission Scanning Electron Microscope showed the microstructure of the BC, characterized by a closely packed network of intertwined bundles of nanofibrils with no specific arrangement (Fig. 4). The results shown were in-line with previously published work by Orlando et al. (2020). To calculate the fiber diameter statistics 200 BNC nanofibrils have been selected from SEM images at a magnification of 15,000 X. The diameter of the nanofibers ranged from 20 to 80 nm, with an average diameter of 50 nm.

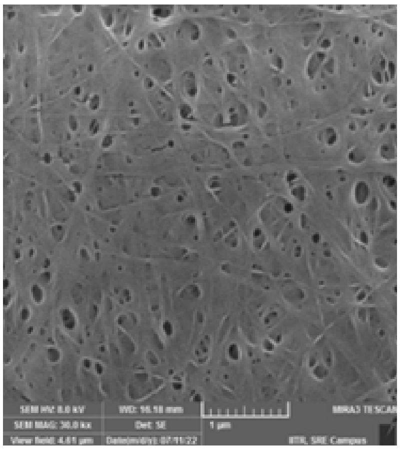

Figure 4:SEM image of purified BC

3.2.2 XRD - The X-ray diffraction (XRD) pattern of pellicles displayed the three characteristic peaks at 2θ values of 14.4°, 16.6°, and 22.4°, which indicate the crystallographic planes (100), (010), and (110) of the cellulose lattice structure, respectively(Fig.5). These peaks are representative of the distinctive cellulose I form, as indicated by the Miller indices. A similar XRD pattern was previously reported for BNC produced under static conditions (Tsouko et al. 2015). The XRD profile did not show the typical points for cellulose II at 2θ values of 12.1° and 20.8°.

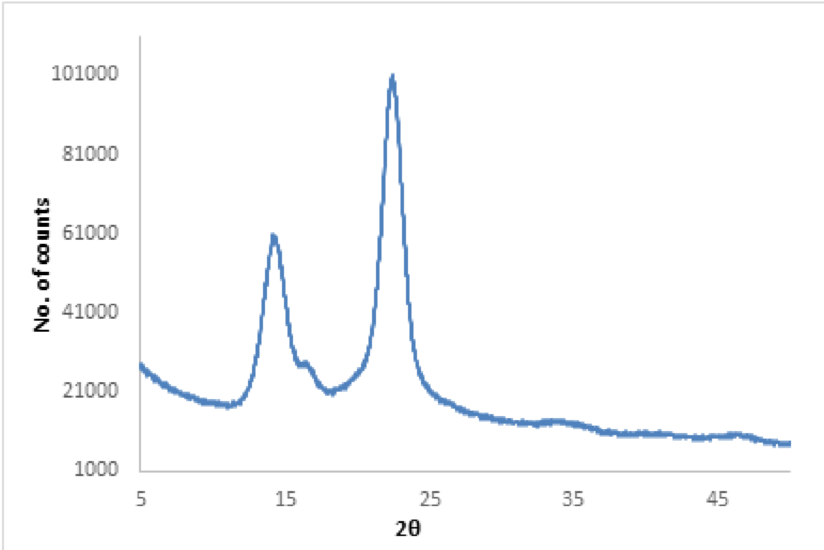

Figure 5: XRD pattern of purified BC

3.2.3 ATR-FTIR - This spectroscopy is a crucial tool for analyzing functional groups and chemical bonds in a sample. The ATR-FTIR analysis of the material produced in BC revealed a broad absorption band of hydroxyl bonds at around 3600–3000 cm-1, due to OH-stretching vibrations (Fig. 6). This result is consistent with the findings from the ATR-FTIR analysis (Daltro et al. 2016). An absorption band at 2858 cm-1 was attributed to C-H stretching vibrations, confirming the presence of amorphous cellulose. The band at 1057 cm-1 indicated the presence of cellulose. The stretching vibrations of CH and CH2 in the range of 3000−2800 cm−1, C–O and C–C stretching motion in the range of 1200−900 cm− 1 are the finger print of crystalline cellulose.

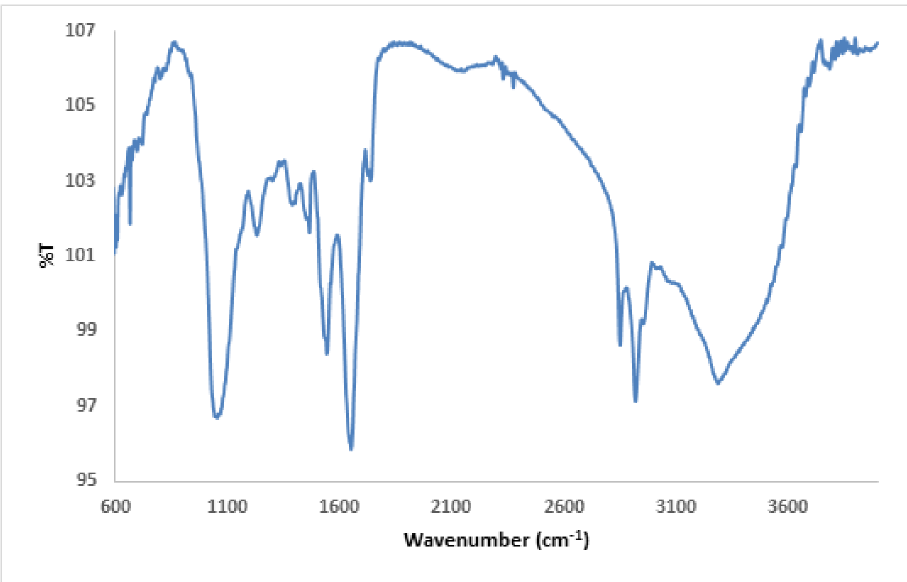

Figure 6: FTIR pattern of purified BC

3.3 Application of Bacterial cellulose for improving paper properties (Wet-End based)

Six different sets of papermaking were made to evaluate the effect of BC in wet-end of papermaking along with recycled pulp. The impact of addition of CS alone, bacterial cellulose alone and the addition of blend of CS and bacterial cellulose in wet-end of papermaking on different strength of paper handsheets are shown in table 1.

Table 1: Properties of unbleached hardwood kraft pulps

Parameter	Control Set	Set 1	Set 2	Set 3	Set 4
Dose of BC in CS, %	-	-	0.8	0.8	-
Dose of CS, Kg/t	-	5	-	-	-
Dose of mixed BC and CS, Kg/t			2.5	5	
Only BC, Kg/t	-	-	0.02	0.04	2.66
Dose of Retention Aid, g/t	250				
Grammage, g/m^2	60±3				
Breaking length, m	2414	3036	3025	3230	2670
Burst index, kN/g	1.38	2.03	2.01	2.25	1.39
Tear index, mNm^2/g	7.50	7.68	7.63	7.69	7.53

Addition of BC in CS improved the efficiency of CS for wet-end application. The application of CS @ 5 kg/t in wet-end of papermaking improved the breaking length, tear index and burst index by 25.8%, 2.40% and 47.1%, respectively in comparison to the strength properties of the paper made using only pulp. This result is found to be in close proximity to the results of an earlier study by Bhardwaj et al., 2017 & 2019 in which CS was used for improving the strength of different pulp furnishes. The increase in strength properties of the paper may be attributed to the fact that CS, form non-covalent linkage with the cellulose and increase the hydrogen bonding due to their chemical structure. The addition of BC 0.8% (with respect to CS) in CS and adding the blended solution further @ 2.5 kg/t in wet-end of papermaking showed similar effect as that shown by the addition of only CS @ 5 kg/t in wet-end of papermaking. The addition of BC 0.8% (with respect to CS) in CS and adding the blended solution further @ 5 kg/t in wet-end of papermaking enhanced the breaking length, tear index and burst index by 33.0 %, 2.53% and 63%, respectively in comparison to the strength properties of the paper made using only pulp. By the addition of BC as such in wet-end of papermaking maximum at a dose of 2.66 kg/t have shown slight improvement in breaking length whereas the burst and tear indices were not improved. This may be due to lower dose of BC added in the wet-end of papermaking in the current study because by the addition of BC at a dose 2.5% to 15% in wet-end of papermaking along with pine pulp improved the strength properties of paper as reported earlier by Danielewiczet al., 2015.

4. CONCLUSION

Scientists worldwide are exploring the potential of BC as a sustainable material for manufacturing various products. While BC has already found applications in several industries, recent studies have focused on its use in the paper sector. BC offers numerous advantages over plant cellulose, such as not requiring the cutting of trees for production, its high purity, and the ability to blend it with plant cellulose. Several ongoing research studies have highlighted the immense potential of BC in the pulp and paper industry. In current study, the potential of BC as a reinforcing additive for enhancing strength properties in food packaging papers has been explored. Two different methods were employed to use laboratory-synthesized BC in the wet-end of papermaking, both of which involved recycled pulp. In the first method, BC was used alone in the wet-end, while in the second method, BC was blended with CS before being used. The results indicated that adding BC to CS at a very low dose of 0.8% increase the efficiency of CS as a dry strength additive by enhancing strength properties of paper i.e., breaking length by 33.0%, tear index by 2.53%, and burst index by 63%. The effect observed by adding a combination of BC at a concentration of 0.8% to CS, and then adding the resulting mixture at a dosage of 2.5 kg/t in the wet-end of papermaking, was similar to the effect shown by only CS when added at a dosage of 5 kg/t in the wet-end of papermaking. The findings of this study highlight the potential of BC as a valuable additive for enhancing the strength properties of food packaging papers. The addition of BC directly at the wet-end of papermaking was done at a very low dose. Therefore, future research should explore higher doses of BC in different pulp furnishes to further evaluate its efficacy and potential applications in food packaging.

Acknowledgements

We gratefully acknowledge Director, CPPRI Saharanpur for providing necessary research facilities, and Research Steering Committee of Development Council for Pulp, Paper and Allied Industries (RSC-DCPPAI) for funding this project.

References

1. Bhardwaj, S., Bhardwaj, N. K., & Negi, Y. S. (2017). Chitosan as wet-end additive for papermaking using mixed hardwood pulp. Appita: Technology, Innovation, Manufacturing, Environment, 70(2), 139-148.
2. Bhardwaj, S., Bhardwaj, N. K., & Negi, Y. S. (2019). Cleaner approach for improving the papermaking from agro and hardwood blended pulps using biopolymers. Journal of Cleaner Production, 213, 134-142.
3. Brown, A. J. (1886). XLIII.—On an acetic ferment which forms cellulose. Journal of the Chemical Society, Transactions, 49, 432-439.
4. Chawla, P. R., Bajaj, I. B., Survase, S. A., & Singhal, R. S. (2009). Microbial cellulose: fermentative production and applications. Food Technology and Biotechnology, 47(2), 107-124.
5. Cheng, K. C., Demirci, A., & Catchmark, J. M. (2011). Pullulan: biosynthesis, production, and applications. Applied microbiology and biotechnology, 92(1), 29-44.
6. Daltro, P., Daltro, G., De Oliveira, G., Basmaji, P., & Guastaldi, A. (2016). Hydrogel and bacterial cellulose mats behavior with calcium phosphate deposition. In Front Bioeng Biotechnol conference abstract: 10th World Biomaterials Congress, pp. 17-22)
7. Danielewicz D, Surma-Ślusarska B. Properties of Bleached Pulps from Low and High Kappa Number Old Corrugated Containers (OCC). FIBRES & TEXTILES in Eastern Europe 2015; 23, 5(113): 129-135.
8. El-Saied, H., Basta, A.H., Gobran, R.H., (2004). Research Progress in Friendly Environmental Technology for the Production of Cellulose products

(bacterial cellulose and its application)', Polym-Plast Technol Eng, 43, 797±820

9. Hong, F., & Qiu, K. (2008). An alternative carbon source from konjac powder for enhancing production of bacterial cellulose in static cultures by a model strain Acetobacter aceti subsp. xylinus ATCC 23770. Carbohydrate Polymers, 72(3), 545-549.

10. Janbade, A., Zaidi, S., Vats, M., Kumar, N., Dhiman, J., & Gupta, M. K. (2022). A Mini Review on Current Advancement in Application of Bacterial Cellulose in Pulp and Paper Industry. In Proceedings of International Conference on Innovative Technologies for Clean and Sustainable Development (ICITCSD–2021) (pp. 435-445). Cham: Springer International Publishing.

11. Jonas, R. and Farah, L.F. (1998). Production and application of microbial cellulose. Polymer Degradation and Stability 59 (1–3): 101–106

12. Klemm, D., Schumann, D., Kramer, F., Heßler, N., Hornung, M., Schmauder, H. P., & Marsch, S. (2006). Nanocelluloses as innovative polymers in research and application. Polysaccharides Ii, 49-96.

13. Kurosumi, A., Sasaki, C., Yamashita, Y., & Nakamura, Y. (2009). Utilization of various fruit juices as carbon source for production of bacterial cellulose by Acetobacter xylinum NBRC 13693. Carbohydrate Polymers, 76(2), 333-335.

14. Nguyen, V. T., Flanagan, B., Gidley, M. J., & Dykes, G. A. (2008). Characterization of cellulose production by a Gluconacetobacter xylinus strain from Kombucha. Current Microbiology, 57(5), 449-453.

15. Orlando I, Basnett P, Nigmatullin R, Wang W, Knowles JC and Roy I (2020) Chemical Modification of Bacterial Cellulose for the Development of an Antibacterial Wound Dressing. Front. Bioeng. Biotechnol. 8:557885. doi: 10.3389/fbioe.2020.557885).

16. Shah, J., & Brown, R. M. (2005). Towards electronic paper displays made from microbial cellulose. Applied microbiology and biotechnology, 66(4), 352-355.

17. Tsouko, E., Kourmentza, C., Ladakis, D., Kopsahelis, N., Mandala, I., etc. (2015). Bacterial cellulose production from industrial waste and by-product streams. International journal of molecular sciences, 16(7), 14832-14849

18. Yamanake, S., & Watanabe, K. (1995). Applications of bacterial cellulose in cellulosic polymers. Cellulosic Polymers, Blends and Composites, Hanser Inc., Cincinnati, OH, USA.

19. Zeng, X., Small, D. P., & Wan, W. (2011). Statistical optimization of culture conditions for bacterial cellulose production by Acetobacter xylinum BPR 2001 from maple syrup. Carbohydrate Polymers, 85(3), 506-513