EVALUATION OF RECYCLING POTENTIAL OF VIRGIN MIXED HARDWOOD PULP TREATED WITH CELLULASE AND CHEMICALS

NISHI K. BHARDWAJ,[*] PUNEET PATHAK,[**] CHAKARVATI SANGO,[**]
ANSHUL SABHARWAL,[**] RAJEEV K. KAPOOR[***] and PRATYOOSH SHUKLA[****]

[*]*Department of Paper and Packaging Technology, Indian Institute of Technology Roorkee, Saharanpur Campus, Saharanpur – 247001, Uttar Pradesh, India*
[**]*Avantha Centre for Industrial Research and Development (ACIRD), Thapar Technology Campus, Bhadson Road, Patiala – 147004, Punjab, India*
[***]*Enzyme Technology and Protein Bioinformatics Laboratory, Maharshi Dayanand University, Rohtak – 124001, Haryana, India*
[****]*School of Biotechnology, Institute of Science, Banaras Hindu University, Varanasi – 221005, Uttar Pradesh, India*
✉ *Corresponding author: N. K. Bhardwaj, nishi.bhardwaj@pt.iitr.ac.in*

Received June 30, 2022

The global demand for paper continues to rise, increasing pressure on wood resources. Recycling waste paper provides a sustainable way to reduce reliance on virgin fibre. This study evaluated virgin mixed hardwood kraft pulp (VMHP) through nine recycling loops using either an alkaline chemical system or a cellulase-based enzymatic treatment. After each cycle, handsheets were prepared and assessed for pulp wetness, bulk, formation, tensile strength (breaking length), tear factor, burst factor, and optical properties (brightness, whiteness, yellowness). Enzymatic treatment offered better strength preservation, reflected in higher breaking length and burst factor compared to chemically treated pulp, though tear strength declined more sharply. In contrast, chemical treatment delivered superior optical performance, maintaining higher brightness and whiteness with lower yellowness. Overall, cellulase-based recycling better retained fibre strength, while chemical treatment enhanced optical quality. These findings highlight the potential of enzyme-assisted recycling as an environmentally friendly method for sustaining fibre performance across multiple cycles.

Keywords: recycling, mixed hardwood pulp, cellulase, alkaline treatment, pulp properties, optical properties, strength properties

INTRODUCTION

The paper industry faces constant challenges due to the rising demand for printing, packaging and tissue grades alongside limited wood supply, high energy costs, and environmental concerns. This has made efficient use of virgin fibres and repeated recycling of secondary fibres essential. The waste paper utilization rate and the recycling rate are key measures used to assess how much recycled fibre is being used. In Europe, for example, recycling rates above 70% have been reported, reflecting a mature system of collection and reuse. Understanding how fibres behave over multiple recycling cycles is therefore crucial for maintaining paper quality and ensuring mill economics. Numerous investigations have focused on understanding how papermaking fibres perform during repeated recycling.[1-10]

Virgin mixed hardwood kraft pulp (VMHP), often rich in eucalyptus, is widely used for printing, tissue, and packaging due to its smooth surface, good bonding ability, and opacity. However, once dried and re-wetted, even these high-quality fibres undergo property loss. Laboratory recycling studies simulating repeated drying, rewetting, and repulping have shown that water retention value (WRV), tensile strength,

and burst strength decrease with each cycle, while drainability often improves after the first few recycles.[9,11] Refining may temporarily restore tensile strength, but it cannot reverse the underlying structural changes caused by hornification. Most studies agree that the largest changes occur within the first three to five recycling loops, after which properties tend to stabilize at lower levels.

Hornification is the central phenomenon explaining these changes. It refers to the irreversible stiffening of fibre walls and the reduction in swelling and flexibility after drying. This leads to fewer available bonding sites and weaker fibre–fibre interactions.[12] Kraft pulps, sulfite pulps, and mechanical pulps respond differently to recycling. While kraft fibres typically lose strength, lignin-rich mechanical fibres can sometimes show improved mechanical properties due to increased flexibility after repeated processing.[2,3,5,13,14] Other mechanisms contributing to strength loss include fibre shortening, collapse of the fibre lumen, and reduced inter-fibre bonded area.[14] Because these effects are interactive, repeated recycling does not yield a single, universal trend; rather, outcomes depend on the furnish, chemical conditions, and the chosen reconditioning method between cycles.

Optical properties are also affected by recycling.[15] Chemical alkaline/peroxide stages can enhance brightness and whiteness while reducing yellowness, owing to chromophore removal and bleaching effects.[16,17] Enzymatic treatments, on the other hand, generally have limited influence on optical properties unless combined with bleaching or deinking agents. Enzymes such as cellulases and xylanases are more often applied to improve drainage, reduce refining energy, or modify fibre surfaces.[18-20] However, overdosing enzymes can increase fines and fibre cutting, which reduces tear strength.[21-24]

Research also shows that fines produced during recycling play an important role in handsheet properties. The effect of fines depends on the type of pulp, freeness level, and amount of addition. Fines in unbeaten pulps influence sheet properties more significantly than in beaten pulps.[25] Thus, the behaviour of recycled fibres is determined by a combination of fibre chemistry, recycling history, and reconditioning method.

During recycling, cellulose fibres gradually lose quality: sheet strength drops and deterioration accumulates with each cycle, which lowers the usable value of the fibres for papermaking.[9,26,27] Therefore, it is essential to examine how recycled cellulose fibres behave across repeated cycles and to track the properties that govern their performance in paper. Given these complexities, modifying recycled fibres with chemical or enzymatic treatments has become a practical approach to offset property losses. Two types of reconditioning methods are especially relevant to the paper mills: (1) chemical alkaline/peroxide treatments (pH ≈ 11): swell fibre walls, increase WRV, open up the internal structure, solubilize or oxidize chromophores; the degree of swelling depends on NaOH concentration and the sequence of refining; too much alkali can cause darkening or strength losses, too little yields weak optical gains;[16,17] (2) enzymatic (cellulase/xylanase) treatments (pH ≈ 6): act effectively on both the surface and inner layers of cellulose/hemicelluloses and selectively modify fibre surfaces to enhance drainage and reduce refining energy.[28,29] Each method has advantages and limitations, and their effects accumulate across repeated cycles.

In this study, we examine the recycling behaviour of virgin mixed hardwood kraft pulp (VMHP) subjected to nine recycling loops. Two reconditioning strategies were compared: (1) chemical alkaline treatment and (2) cellulase-based enzymatic treatment. We followed a controlled laboratory protocol that repeated the steps of disintegration → sheet forming → drying → repulping in up to nine loops. For each cycle, pulp and paper properties were measured, including wetness/drainability, bulk, formation, tensile (breaking length), tear factor, burst factor, and optical properties (brightness, whiteness, and yellowness). The study parameters correspond to common papermill practices – alkaline charge and pH to regulate swelling, and enzyme dose/time to adjust fibre surface action. By using virgin hardwood pulp and controlled laboratory conditions, this work isolates recycling-induced changes from post-consumer contaminants. The comparative results provide insights into how chemical and enzymatic treatments affect fibre performance over multiple recycling cycles and highlight their potential for extending fibre usability in papermaking.

EXPERIMENTAL

Pulp, chemicals, and enzyme

Bleached mixed hardwood pulp was sourced from a wood-based paper mill in northern India. Before the recycling trials, the pulp furnish was refined in a PFI mill to 29±1 ºSR. The chemical reconditioning system comprised sodium hydroxide (NaOH), sodium silicate (Na_2SiO_3), and Tween-80 (non-ionic surfactant). For the enzymatic treatment route, a commercial cellulase formulation (Enzyme A, EA) was obtained from an Indian supplier. Activity profiling of EA showed an endoglucanase optimum at pH 6.0 and temperature of 50 °C. Under these conditions, the enzyme preparation remained reasonably stable over 120 min, retaining ~70% residual activity.

Recycling methodology

Virgin mixed hardwood pulp (VMHP) handsheets were prepared and evaluated after each recycling loop from cycle 0 (virgin control) through cycle 9 for strength and optical properties. For cycle 0, 600 g (oven dry, OD) of VMHP was hydrapulped for 30 min at an agitation rate of 300 rpm under two separate conditioning routes: chemical treatment (NaOH 1.5%, Na_2SiO_3 1.5%, Tween-80 0.5%, adjusted to pH 11.0 at a temperature of 50 °C) and enzymatic treatment (cellulase at pH 6.0, 50 °C with a total incubation time of 60 min, including the 30 min hydrapulping period). After hydrapulping, the pulp was thoroughly washed. For each treated batch, 60 g of OD pulp was withdrawn to make a handsheet (70 gsm; 30 g) and to measure the designated pulp and paper properties. The remaining 540 g OD pulp from cycle 0 was used to make handsheets of 200 gsm and dried to serve as the feed for cycle 1 repulping under the same treatment conditions, followed by washing and another 60 g OD sampling for handsheet preparation and testing, as mentioned previously. The process is termed repulping as it includes the re-dispersion of dry fibres in water. This repulping–washing–sampling sequence was repeated iteratively (*i.e.*, re-dispersing the fibres in water for each new loop), so that after each cycle, a 60 g OD aliquot was taken for handsheet formation and property measurements, and the balance was advanced to the next cycle. The procedure continued in this manner through cycle 9 (see Fig. 1 for a detailed workflow).

All analytical determinations were performed in triplicate unless otherwise noted (duplicate), and results are reported as mean ± standard deviation.

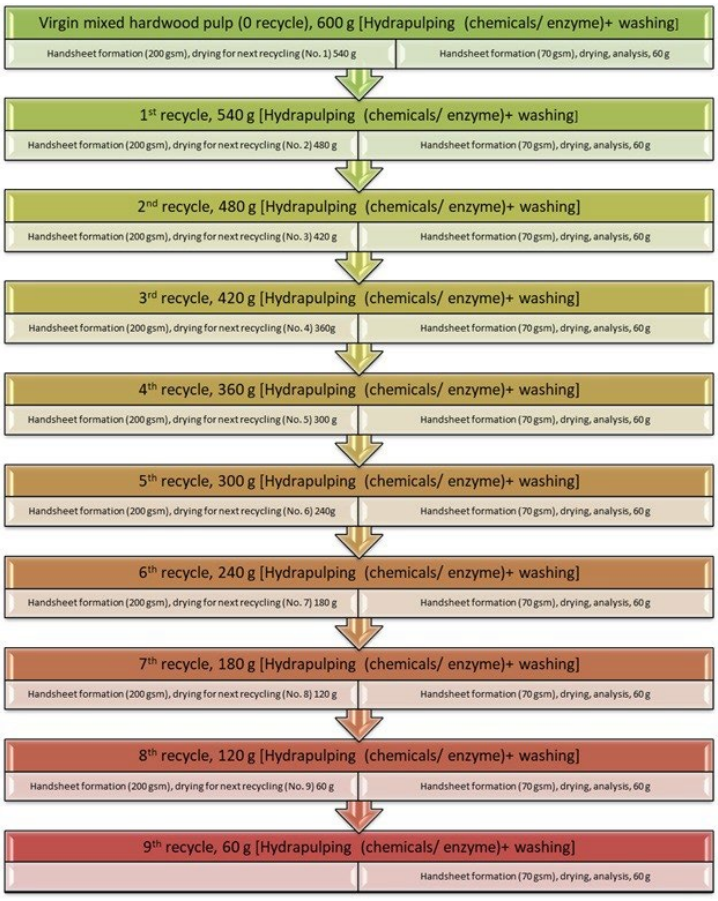

Figure 1: Methodology used in recycling experiments for chemical and enzyme treatments

Pulp and handsheet properties

The wetness of the pulp was determined using a Schopper-Riegler (ºSR) tester according to ISO 5267/1 (1999). The ºSR value reflects the pulp's drainage capacity, which depends on its hydrophilic nature, fibre fibrillation, and the amount of fine particles present. In all experiments, standard handsheets (70 g/m^2) were prepared using a British sheet-forming machine according to Tappi method T-205 cm-80. The handsheets were conditioned at 27±2 °C and 65±2% relative humidity as per ISO: 187, and then tested after each recycling stage for different physical strength properties, including breaking length (TAPPI T 404 cm-92), burst factor (TAPPI T 403 om-97), and tear factor (TAPPI T 411 om-98). The brightness was tested following TAPPI T 218 sp-97 using the Buchner funnel method. Optical properties were determined according to TAPPI T 452 om-92. The formation index of the paper handsheets was measured with a Paprican Microscanner using transmitted light.

RESULTS AND DISCUSSION

The mixed hardwood pulp was recycled from the 0th to 9th cycle, using chemical and enzyme treatments separately. After each cycle, the recycled pulp was tested and compared for both strength and optical properties. The results reflect the quality of the pulp used and its papermaking performance after each treatment, including wetness, sheet bulk, formation index, tensile (breaking length), tear factor, burst factor, and optical properties such as brightness, whiteness, and yellowness.

Effect on pulp wetness (°SR)

With chemical treatment, °SR values rose sharply to a peak around 33–35 by the 2nd–3rd cycle, then gradually declined to ~26 by cycles 6–9, where they stabilized. This early increase is linked to alkali swelling from NaOH, while the later decline reflects hornification due to repeated recycling that reduces the water retention value (WRV) and thus lowers °SR. In contrast, the cellulase enzyme treated pulp maintained a nearly constant °SR (29–30) across all cycles, with only minor fluctuations (±1 °SR). This indicates stable drainage during repeated recycling (Fig. 2). The action of cellulase is supposed to cause external fibrillation during the re-slushing process. Due to fibrillation, pulp wetness will be increased due to the bonding of water molecules with exposed hydroxyl groups of the fibrillated structure of recycled fibres. The °SR test variance of a few units is common, matching the error bars. These findings agree with earlier studies showing that repeated recycling generally improves drainability (*i.e*., tends to lower °SR over cycles), as seen in the late-cycle chemical treatment data.[30,31] In another study on hardwood pulp, upon recycling, the °SR decreased more proportionally to the increasing recycling number for up to 5 recycling rounds.[9] However, enzymatic treatment has also been reported to improve the drainage of pulp.[32]

Effect on bulk

Under chemical treatment, pulp bulk increased between cycles 3–6 to ~2.05–2.15 cm^3/g and then remained stable (~2.0–2.1 cm^3/g) up to cycle 9. In contrast, the cellulase treatment showed a small increase at cycle 1 (~2.0), then a steady decline to ~1.7 (cycles 3–4), followed by a sharp drop to ~1.45 by cycle 6 and stabilizing around 1.40–1.45 cm^3/g through cycles 7–9. Overall, chemical treatment helped maintain or increase bulk, whereas cellulase treatment led to densification (bulk loss) of the sheet. Chemical treatment deteriorated the fiber through hornification, affecting the bulk density of the paper. Under chemical treatment, the bulk increased after each recycling, from 1.81 cm^3/g (cycle 0) to 2.06 cm^3/g (9th cycle) (Fig. 3). Under enzymatic treatment, the bulk was reduced from 1.81 cm^3/g (cycle 0) to 1.40 cm^3/g (9th cycle). This lower value of the bulk indicated tighter fibre packing, the densification or consolidation of the fibres present in the sheets.

The action of cellulase is supposed to make fibre less stiff than the fibre obtained after chemical treatment. Alkaline swelling expands fibre walls and can increase handsheet bulk, consistent with the sustained higher bulk in the chemical route.[16,33,34] Previous work also showed that repeated recycling altered flexibility (hornification), but sheet bulk trends depended on fines and bonding, higher fines content raised apparent density (lowered bulk).[35] Cellulase-rich treatments can increase fines and WRV and shorten fibres, often leading to denser sheets – aligning with bulk loss in the enzyme treated pulp.[21] In contrast, some mild enzymatic pretreatments reported little change in apparent density, showing formulation/dose sensitivity.[36]

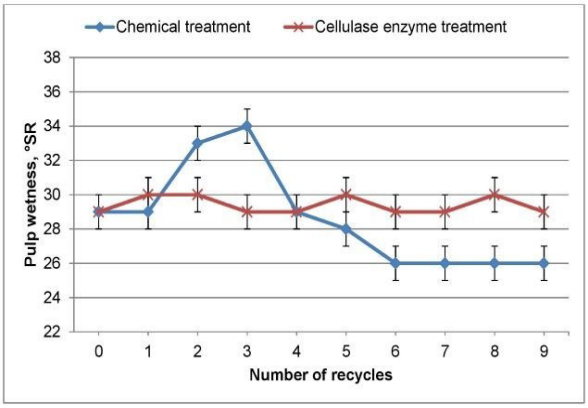

Figure 2: Effect of chemical and enzymatic treatments on pulp wetness after 9 recycles of mixed hardwood pulp

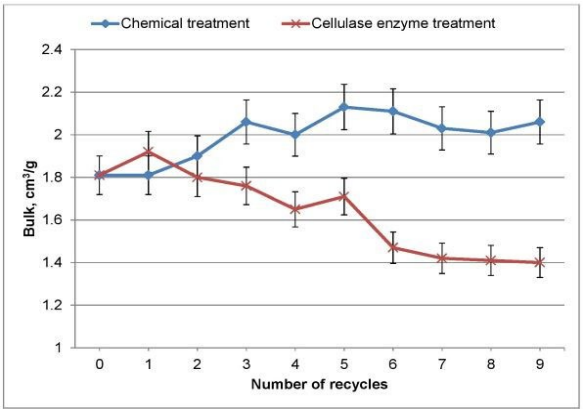

Figure 3: Effect of chemical and enzymatic treatments on bulk after 9 recycles of mixed hardwood pulp

Effect on formation index

The formation reflects how evenly fibres are distributed in a paper sheet and is influenced by pulp consistency, drainage, fines, fibre flexibility and retention chemistry. The effect of both chemicals and enzymatic treatments on paper formation during the recycling process was investigated. In chemical treatment, the paper formation index peaked at ~220 in cycle 1, then dropped to ~190–195 by cycles 3–4, bottomed near ~185 by cycles 6–7, and then recovered to ~200–205 by cycle 9. The cellulase treatment route dipped to ~198 at cycle 1, stayed slightly higher and relatively stable (~195–205) compared to the chemical route through mid-cycles, and ended highest (~213) at cycle 9. Overall, sheets made after enzyme treatment showed more stable (and late-cycle better) formation, while chemical treatment showed a U-shape trend – early improvement, mid-cycle deterioration, late recovery. The pattern showed that the formation index of cellulase enzyme-treated handsheets was about 5% higher when compared to that of chemically treated pulp. After the 9th recycling, the formation index was 213 and 203 for the cellulase enzyme and chemically treated handsheets, respectively (Fig. 4). The softening action of cellulase is expected to provide a better formation during handsheet making due to less stiff fibres. Enzymatic pretreatments often reduce refining energy and floc size, helping maintain or slightly improve formation. Alkaline swelling can raise WRV and disturb drainage, explaining the mid-cycle dip here (then recovery as recycling/hornification lowers swelling). Treating fibres with cellulase enzyme was also reported to provide several benefits for papermaking – the most notable being improved sheet formation due to fibre shortening.[37]

Effect on strength properties

Understanding the strength properties of paper during recycling is important for improving the utilization of recycled fibres. In this study, mixed hardwood pulp was recycled throughout 9 cycles, using chemical and enzymatic treatments, and the changes in strength properties were compared. Strength properties were mostly affected up to the 9th cycle. The most noticeable effects occurred during the first few cycles, with both treatments showing a sharp loss of tensile strength in the initial two loops.

Effect on tensile strength (breaking length)

During beating, fibre fibrillation enlarges the active surface area.[38-40] This effect induces an increase in fibre bonding, paper strength, and finally the increase in breaking length. However, recycling caused a rapid loss. After the first cycle, the breaking length of chemically treated pulp dropped by ~15% (from 4365 m to 3065 m), while that of enzyme-treated pulp declined more moderately – from 4365 m to 3618 m (Fig. 5a). By the second cycle, the breaking length in enzyme-treated pulp dropped further to ~2354 m. It was also reported that the tensile indexes of the handsheets made from hardwood pulps dramatically decreased during the early rounds of recycling, especially in the first and the second cycles.[9]

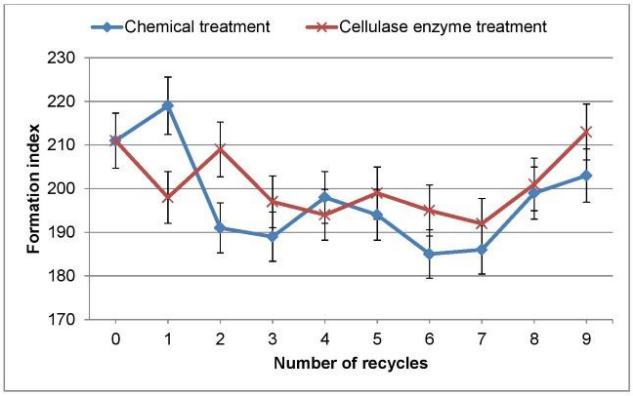

Figure 4: Effect of chemical and enzymatic treatments on formation index after 9 recycles of mixed hardwood pulp

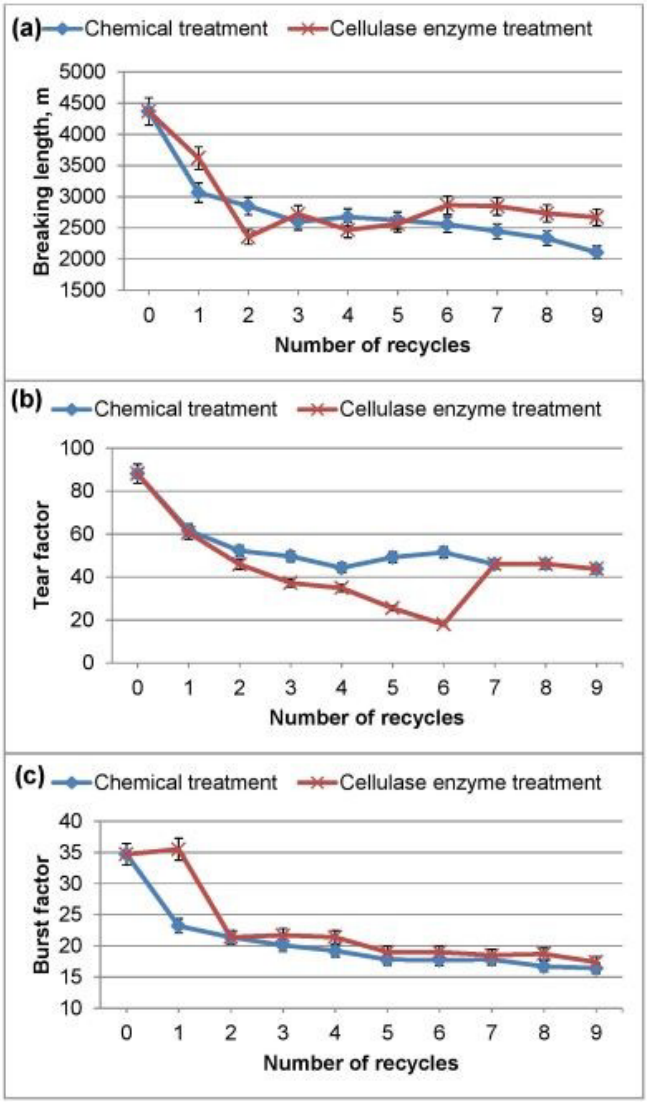

Figure 5: Effect of chemical and enzymatic treatments on strength properties after 9 recycles of mixed hardwood pulp, (a) breaking length, (b) tear factor, and (c) burst factor

Meanwhile, from the 3rd up to 5th cycles, both chemical and enzyme-treatments showed an almost similar trend, but from the 4th cycle onward, enzyme-treated pulp consistently showed higher breaking length (about 11% to 21% higher) compared to chemically recycled pulp.

This improvement in the strength properties may be due to higher surface fibrillation due to cellulase action. After nine cycles, breaking length was 2670 m for the cellulase treated pulp, compared to 2107 m for the chemically treated one (Fig. 5a). Overall, the biggest damage happened early, enzymes helped maintain a modest tensile advantage mid-cycle, but neither path prevented the longer-term decline. The fibres are simultaneously shortened during beating/disintegration, which causes deterioration of mechanical properties. Moreover, a decrease in the thickness of fibre walls during pulp fibre recycling causes a loss of tensile strength.[41] This declining trend in breaking length was due to a decrease in fibre wall thickness, as well as tensile strength. Repeated recycling (hornification) reduces swelling and bonding, so tensile/breaking-length typically falls – matching the strong early drop and low late-cycle plateau. Reviews and studies on chemical and recycled hardwood pulps report a decline in tensile strength throughout recycles, sometimes with the largest loss in the first 2–3 loops.[9,42]

Effect on tear factor

Both treatments showed at first a tear factor of ~90. The tear factor then dropped for both routes, chemical and enzymatic. The chemically treated sample exhibited a drop to ~45 by the 4th cycle and then showed minor fluctuations before declining again towards the 9th cycle. In contrast, the enzyme-treated pulp showed a sharper decline, reaching a minimum of ~18 at cycle 6, then partially recovered to ~46 by cycles 7–9, ending close to the result of chemically treated pulp. The continuous drying and fibrillation of fibres during recycling increase fibre surface area, but make fibres more brittle, which reduces tearing strength. The action of the enzyme also produces fines and fibre shortening, leading to greater mid-cycle losses. The late recovery may be due to stabilization of fibre characteristics and inter-fibre hydrogen bonding effects.[43,44] Overall, tear is the most enzyme-sensitive of the three properties in Figure 5 and showed a characteristic "V" shape for the enzyme route.

Effect on burst factor

In the first cycle, enzyme-treated pulp showed a higher burst factor (~35) compared to the chemically treated one (~24). Thereafter, both pulps declined quickly to ~21 by cycle 2 and then remained low and similar through later cycles up to cycle 9 (Fig. 5c). The burst factor of chemically and enzyme-treated pulps showed an almost similar decreasing trend after 1st recycling with a modest improvement (5-10% higher) in enzyme treatment, compared to chemical treatment. This may be attributed to the fiber-to-fiber bonding improvement due to the action of cellulase on the fibre surface. Still, repeated drying and rewetting caused permanent loss of swelling and fibre bonding capacity, leading to an overall decline. This decline in bursting strength after the chemical or enzyme treatment was due to the breakdown of secondary fiber. Any early enzyme-related bonding advantage was transient.

Paper generally gets its strength from the strength of individual fibres and the number of H-bonds formed among fibres during the papermaking processes.[44] It is expected that more flexible fibres have better conformability, resulting in higher bonding among fibres. The bonding ability of the fibres is generally degraded due to drying and recycling processes.[44] During drying, fibre walls collapse and form irreversible internal hydrogen bonds that prevent complete re-swelling in later cycles. This stiffens the fibres and reduces their conformability, producing weaker sheets during the recycling process.[1] However, a tendency of flattening due to collapsing of fibre lumen after recycling resulted in denser and stronger paper than in the case of the first cycle of papermaking for mechanical pulps.[13] Due to repeated mechanical action and drying, fibrillation in the S1 layer leads to peeling away of the outer fibre layers, revealing the inner S2 layer and reducing tear strength.[45] The use of cellulases, particularly endoglucanases, has been shown to increase dewatering, tensile strength, and sheet smoothness.[22] It is also suggested that endoglucanases target already weakened zones in fibres, since these regions have a more amorphous structure.[46] Another study examined how *Paenibacillus* cellulase affects fibre surfaces.

Results showed that cellulase treatment caused fibres to swell even without mechanical refining, and refining after cellulase treatment led to better fibrillation. Cellulase enzymes can break down parts of the cell wall, loosening the structure. This breakdown increases the exposed surface area, allowing stronger interactions with water molecules and making fibres easier to fibrillate externally.[47] Fibre oxidation, micro-cracks, and loss of polymerization during repeated cycles further weaken the fibres. Acidic conditions worsen degradation, while neutral or alkaline conditions reduce it. Enzyme-treated pulp showed relatively better strength retention, compared to alkaline treated pulp, as cellulase improved surface fibrillation and bonding, despite overall fibre deterioration.

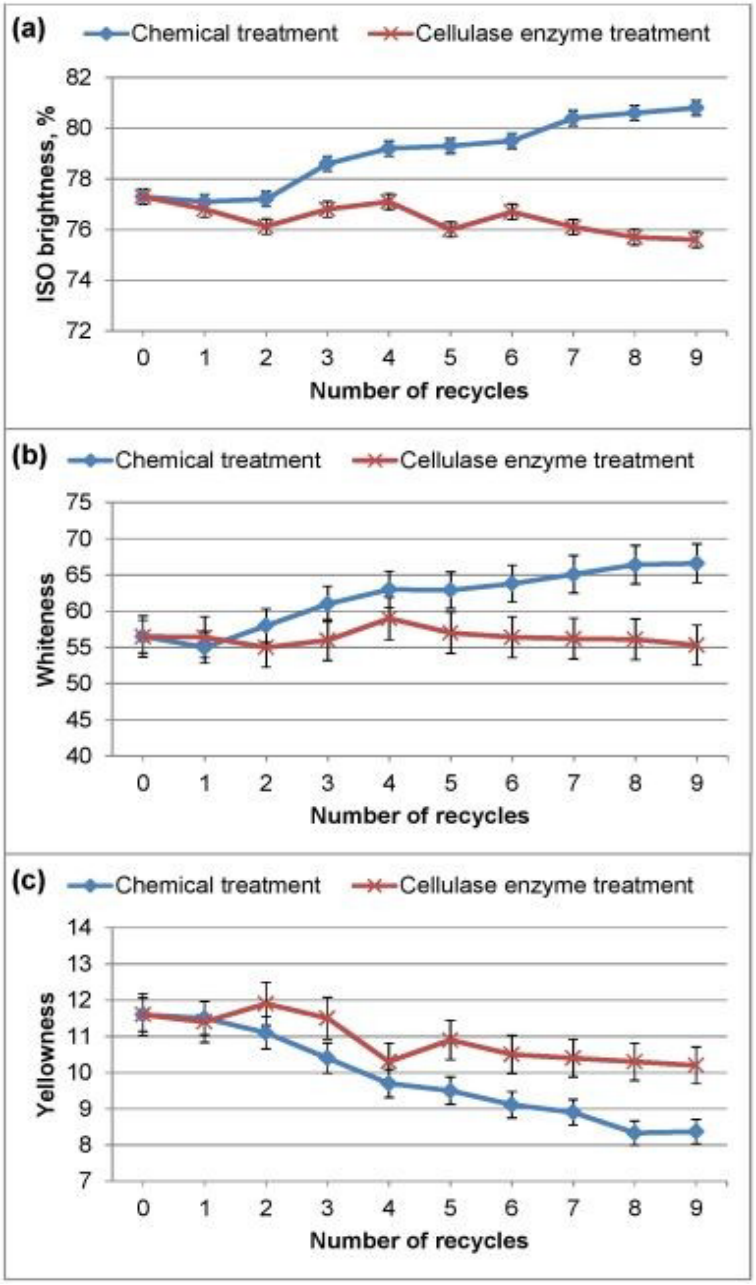

Figure 6: Effect of chemical and enzymatic treatments on optical properties after 9 recycles of mixed hardwood pulp, (a) brightness, (b) whiteness, and (c) yellowness

Effect on optical properties

Effect on brightness

The initial pulp brightness was 77.3% ISO. After chemical treatment, brightness increased steadily after the second recycle and reached ~80.8% by the 9th cycle. In contrast, enzyme-treated pulp showed a gradual decline, ending at ~75.6% by cycle 9. Thus, repeated recycling improved brightness in chemically treated pulp, while the enzymatic treatment caused a slight decrease (Fig. 6a). This improvement may be attributed to more effective removal of chromophores, enhanced delignification during recycling and dilution during papermaking. In comparison, enzyme-treated pulp showed irregular (zig-zag) brightness changes across cycles, but overall remained 3–5 points lower than that of the chemically treated one (Fig. 6a). This reduction may result from enzyme-induced hydrolysis of fines or fibre surface changes that expose lignin-rich sites or generate new chromophoric groups, reducing brightness over successive recycles

Effect on whiteness

Whiteness and yellowness of paper showed opposite trends. Both treatments started with a CIE whiteness of 56.5. Chemically treated pulp showed continuous improvement, reaching ~66.6 by the 9th cycle. Enzyme-treated pulp, however, showed only a slight increase to ~59 at cycle 4 before dropping to ~55.3 by cycle 9. Hence, whiteness across recycles increased for chemically (alkaline) treated pulp, whereas cellulase treatment led to a net decline after 9 cycles. Since whiteness measures the full visible spectrum (unlike ISO brightness, which uses only the blue band), the consistent increase in the sample subjected to the chemical route indicates broader-spectrum optical improvement due to chemical action and chromophores reduction. After the 9th recycle, the whiteness of chemically treated pulp was higher (up to 20%), compared to the enzyme treated mixed hardwood pulp (Fig. 6b). By contrast, cellulase alone can expose lignin-rich sites or generate fines that scatter differently, yielding lower spectral reflectance.

Effect on yellowness

The yellowness index was initially around ~11.6. Chemical treatment caused a steady decline, reaching ~8.3 by the 9th cycle, while enzyme-treated pulp stayed nearly constant between ~10.5 and 11.6, with only a slight reduction to ~10.2 at cycle 9. Since whiteness improved more in the chemically treated sample, yellowness remained significantly higher (up to ~21%) in enzyme-treated pulp after 9 cycles (Fig. 6c). The increase in yellowness is related to oxidative reactions of cellulose and hemicelluloses during recycling, where carboxyl groups formed from hydroxyl groups act as chromophores, contributing to yellowing.[48,49]

In alkaline/peroxide bleaching around pH 11, the best results for high brightness and low yellowness were obtained when the alkali-to-peroxide ratio was adjusted properly. The highest brightness was achieved at a $NaOH/H_2O_2$ ratio of 0.75.[17] Starting the bleaching process at pH 11 also gave the best brightness outcome. It was reported that hydrogen peroxide bleaching at this initial pH produced the greatest whiteness and brightness values.[50] Alkaline stages help reduce brightness reversion, and properly controlled peroxide stages at about pH 11 can decrease yellowness.[17] Neutral enzymes do not directly bleach pulp; any improvement comes from cleaning fibre surfaces or aiding deinking.[51] Cellulase and xylanase were reportedly applied to eucalyptus pulp to obtain biobleached fibres. Xylanase pretreatment clearly enhanced the bleaching effect, while cellulase showed little improvement. Still, both enzymes, particularly xylanase, were effective in removing HexA from the fibres.[52] Earlier work showed that biotreatment with *Fusarium concolor* X4 increased the brightness of poplar CTMP and reduced light-induced yellowing.[53] However, in the present study, with bleached hardwood pulp, enzyme-treated samples showed higher yellowness.

CONCLUSION

This study demonstrated that repeated recycling of virgin mixed hardwood pulp results in progressive losses of both strength and optical properties, though the magnitude and trajectory of decline depend on the treatment applied.

Enzymatic treatment with cellulase helped maintain higher breaking length and burst factor compared to chemical treatment. The improved fibre–fibre bonding from cellulase-mediated fibrillation contributed to this strength advantage. However, tear factor declined more sharply with

the enzyme treatment, reflecting fines generation and fibre shortening. Chemically treated pulps consistently outperformed enzyme-treated samples in brightness, whiteness, and yellowness reduction over nine cycles. This was attributed to the oxidative effects of alkaline chemistry on chromophore removal, whereas cellulase action did not provide bleaching benefits.

Enzyme treatment represents a more sustainable and environmentally friendly route for enhancing strength retention during fibre recycling, whereas chemical treatment is preferable when optical quality is the primary requirement. A hybrid strategy, combining enzymatic modification with controlled chemical treatment, may therefore offer the best balance for prolonging fibre usability in industrial papermaking.

ACKNOWLEDGMENT: The laboratory work was conducted when the first four authors (NKB, PP, CS and AS) were affiliated with the Avantha Centre for Industrial Research and Development (ACIRD), Yamuna Nagar, India. The study was performed as part of a collaborative project with Maharshi Dayanand University, Rohtak, Haryana. Financial assistance from the Department of Biotechnology, Government of India (Grant no. BT/PR27437/BCE/8/1433/2018) is gratefully acknowledged.

REFERENCES

[1] R. C. Howard and W. Bichard, *J. Pulp Pap. Sci.*, **18**, 151 (1992)
[2] L. N. Law, J. L. Valade and J. Quan, *TAPPI J.*, **79**, 167 (1996)
[3] L. N. Law, J. L. Valade and Z. Li, *TAPPI J.*, **79**, 181 (1996)
[4] I. Eriksson, P. Lunabba, A. Pettersson and G. Carlsson, *TAPPI J.*, **80**, 151 (1997)
[5] C. Sharma, N. J. Rao and S. P. Singh, *IPPTA J. Convention Issue*, Dec., 69 (1998)
[6] M. Garg and S. P. Singh, *TAPPI J.*, **3**, 25 (2004)
[7] M. A. Hubbe, *BioResources*, **2**, 739 (2007), https://doi.org/10.15376/biores.2.4.739-788
[8] T. Yamauchi and M. Yamamoto, *APPITA J.*, **61**, 396 (2008)
[9] Y. Chen, J. Wan, Q. Wu, Y. Ma and M. Huang, *Cellulose Chem. Technol.*, **50**, 1061 (2016), https://www.cellulosechemtechnol.ro/pdf/CCT9-10(2016)/p.1061-1067.pdf
[10] N. Jirarotepinyo, J. Nguyen, A. Cross, H. Jameel and R. A. Venditti, *J. Mater. Cycles Waste Manage.*, **27**, 1901 (2025), https://doi.org/10.1007/s10163-025-02227-2
[11] Y. Chen, J. Wan, X. Zhang, Y. Ma and Y. Wang, *Carbohyd. Polym.*, **87**, 730 (2012), https://doi.org/10.1016/j.carbpol.2011.08.051
[12] G. V. Laivins and A. M. Scallan, in *Procs of the 10th Fundamental Research Symposium*, PITA (Paper Industry Technical Association), Lancashire, U.K. Sept. 19-24, 1993, pp. 1235 1260
[13] M. M. Nazhad and L. Paszner, *TAPPI J.*, **77**, 171 (1994)
[14] M. Garg and S.P. Singh, *APPITA J.*, **59**, 274 (2006)
[15] M. Souček, *Pap. Cellulose*, **64**, 164 (2009)
[16] K. H. Choi, A. R. Kim and B. U. Cho, *BioResources*, **11**, 3769 (2016)
[17] F. Zeinaly, M. Karimi, J. Shakhes and H. Mohammadi, *Cellulose Chem. Technol.*, **50**, 285 (2016), https://www.cellulosechemtechnol.ro/pdf/CCT2(2016)/p.285-292.pdf
[18] R. Singh and N. K. Bhardwaj, *IPPTA J.*, **23**, 121 (2011)
[19] R. Singh and N. K. Bhardwaj, *IPPTA J.*, **22**, 109 (2010)
[20] N. K. Bhardwaj, P. Bajpai and P. K. Bajpai, *J. Biotechnol.*, **51**, 21 (1996)
[21] K. P. Buzała, P. Przybysz, H. Kalinowska and M. Derkowska, *PLOS One*, **11**, e0161575 (2016), https://doi.org/10.1371/journal.pone.0161575
[22] P. K. Verma, N. K. Bhardwaj and S. P. Singh, *Can. J. Chem. Eng.*, **94**, 430 (2016), https://doi.org/10.1002/cjce.22410
[23] N. K. Bhardwaj, P. Bajpai and P. K. Bajpai, *APPITA J.*, **48**, 378 (1995)
[24] R. Singh, N. K. Bhardwaj and B. Choudhury, *APPITA J.*, **67**, 226 (2014)
[25] S. K. Gülsoy and S. Uysal, *Drvna Ind.*, **71**, 327 (2020), https://doi.org/10.5552/drvind.2020.1904
[26] Y. Chen, J. Wan and Y. Ma, *APPITA J.*, **62**, 290 (2009)
[27] M. S. Jahan, *TAPPI J.*, **2**, 9 (2003)
[28] E. M. Cadena, A. I. Chiriac, F. I. J. Pastor, P. Díaz, T. Vidal *et al.*, *Biotechnol. Progress*, **26**, 960 (2010), https://doi.org/10.1002/btpr.411
[29] A. I. Chiriac, E. M. Cadena, T. Vidal, A. L. Torres, P. Díaz and F. I. J. Pastor, *Appl. Microbiol. Biotechnol.*, **86**, 1125 (2010), https://doi.org/10.1007/s00253-009-2350-8
[30] M. A. Zanuttini, V. A. Marzocchi and P. Mocchiutti, *Cellulose Chem. Technol.*, **43**, 65 (2009), https://www.cellulosechemtechnol.ro/pdf/CCT1-3-2009/p.65-69.pdf
[31] B. Sjöstrand, C. A. Karlsson, C. Barbier and G. Henriksson, *BioResources*, **18**, 3856 (2023), https://doi.org/10.15376/biores.18.2.3856-3869
[32] Z. Efrati, M. Talaeipour, A. Khakifirouz and B. Bazyar, *Cellulose Chem. Technol.*, **47**, 547 (2013),

https://www.cellulosechemtechnol.ro/pdf/CCT7-8(2013)/p.547-551.pdf
[33] K. H. Choi, A. R. Kim and B. U. Cho, *Nord. Pulp Pap. Res. J.*, **33**, 503 (2018), https://doi.org/10.1515/npprj-2018-3059
[34] Y. Ji, Y. Peng, A. Strand, S. Fu, A. Sundberg *et al.*, *BioResources*, **13**, 7310 (2018), https://doi.org/10.15376/biores.13.4.7310-7324
[35] H. Lee, W. S. Nam, S. D. Sohn and K. H. Paik, *J. Ind. Eng. Chem.*, **17**, 100 (2011), https://doi.org/10.1016/j.jiec.2010.12.004
[36] M. Kmiotek, K. Dybka-Stępień and A. Karmazyn, *BioResources*, **16**, 4221 (2021), https://ojs.bioresources.com/index.php/BRJ/article/view/23333
[37] U. B. Mohlin and B. Pettersson, *Progress Biotechnol.*, **21**, 291 (2002), https://doi.org/10.1016/S0921-0423(02)80032-6
[38] H. N. Banavath, N. K. Bhardwaj and A. K. Ray, *IPPTA J.*, **21**, 141 (2009)
[39] N. K. Bhardwaj, V. Hoang and K. L. Nguyen, *Bioresour. Technol.*, **98**, 1647 (2007), https://doi.org/10.1016/j.biortech.2006.05.040
[40] H. N. Banavath, N. K. Bhardwaj and A. K. Ray, *Bioresour. Technol.*, **102**, 4544 (2011)
[41] T. Okayama, *Japan TAPPI J.*, **56**, 986 (2001), https://doi.org/10.2524/jtappij.56.986
[42] M. Li, K. Hu and S. Shao, *Cogent Eng.*, **10**, 2116828 (2022), https://doi.org/10.1080/23311916.2022.2116828
[43] J. L. Klofta and M. L. Miller, *Pulp Pap. Canada*, **95**, 41 (2004)
[44] M. A. Hubbe, *BioResources*, **1**, 281 (2006), https://doi.org/10.15376/biores.1.2.281-318
[45] E. Afra, H. Yousefi, M. M. Hadilam and T. Nishino, *Carbohyd. Polym.*, **97**, 725 (2013), https://doi.org/10.1016/j.carbpol.2013.05.032
[46] N. Gurnagul, D. H. Page and M. G. Paice, *Nord. Pulp Pap. Res. J.*, **7**, 152 (1992), https://doi.org/10.3183/npprj-1992-07-03-p152-154
[47] C. H. Ko, C. Y. Yang, F. C. Chang and L. D. Lin, *J. Environ. Manage.*, **241**, 1 (2019), https://doi.org/10.1016/j.jenvman.2019.03.133
[48] Margutti, G. Conio, P. Calvini and E. Pedemonte, *Restaurator*, **22**, 67 (2001), https://doi.org/10.1515/rest.2001.67
[49] R. Solár, *Wood Res.*, **54**, 1 (2009), https://www.woodresearch.sk/wr/200904/01.pdf
[50] L. Li, S. Lee, H. L. Lee and H. Youn, *BioResources*, **6**, 721 (2011), https://doi.org/10.15376/biores.6.1.721-736
[51] T. W. Jeffries, *Mat. Res. Soc. Symp. Proc.*, **266**, 277 (1992)
[52] C. Valls, E. M. Cadena and M. B. Roncero, *Carbohyd. Polym.*, **92**, 276 (2013), https://doi.org/10.1016/j.carbpol.2012.08.083
[53] D. Zhang, X. Li and J. Zhao, *Biomed. Res. Int.*, **1**, 9497215 (2020), https://doi.org/10.1155/2020/9497215