Amphiphilic Cyclodextrin-Modified Recycled Fibers for Green Hygiene Products with Enhanced Strength and Flushability

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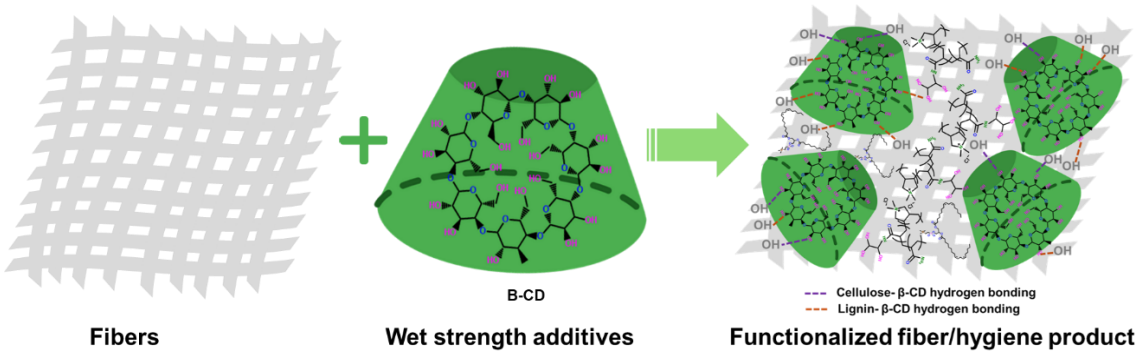
GRAPHICAL ABSTRACT



Amphiphilic Cyclodextrin-Modified Recycled Fibers for Green Hygiene Products with Enhanced Strength and Flushability

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People living in densely populated settlements face inadequate sanitation and rising incidences of acute bacterial infections, underscoring the urgent need for effective and sustainable hygienic products. The amphiphilic molecular architecture of β-cyclodextrin (β-CD), characterized by a hydrophobic internal cavity and a hydrophilic external surface, offers a unique platform for designing sustainable additives for hygiene products. This study employs β-CD to improve the mechanical performance and end-of-life management of old corrugated container (OCC)-derived recycled fibers for tissue and towel applications. β-CD operates through dual interactions: its hydrophobic cavity associates with lignin and other hydrophobic moieties *via* van der Waals interactions, while its hydrophilic exterior forms hydrogen bonds with cellulose, enhancing fiber–water interactions and hydrogen bonding. These coupled molecular interactions enhance dry strength while promoting fiber swelling, water penetration, and rapid disintegration, which is essential for flushability and recyclability. In dual-stage systems combining β-CD with low dosage of glyoxalated polyacrylamide (g-PAM), pretreated OCC (POCC) fibers achieved an immediate wet tensile comparable to g-PAM-only at a higher dosage while exhibiting substantially reduced disintegration time. Overall, this work demonstrates a viable pathway for producing recyclable and flushable recycled-fiber hygiene products that maintain high dry and immediate wet tensile strength during use, yet disintegrate rapidly during flushing or repulping.

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Keywords: Recycled fibers; Cyclodextrin; Green chemical additives; Temporary wet strength; Flushability; Sustainable hygiene products

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INTRODUCTION

Functional wet-end additives, including dry- and wet-strength aids, sizing agents, and retention aids, are widely used in hygiene and packaging paper production (Marton and Marton 1976; Roberts *et al.* 1986; Malton *et al.* 1998). A substantial fraction of these additives remains associated with fibers during recycling, and highly charged wet-end polymers can continue to influence fiber charge characteristics even after drying and repulping (Grau *et al.* 1996; Sjöström and Ödberg 1997). Many paper grades are engineered to deliver both dry and wet strength (Spence 1999), often through synthetic wet-strength additives (WSAs) that bind to cellulose either permanently or temporarily.

WSAs limit fiber–fiber loosening and preserve sheet structure upon water exposure by maintaining fiber–fiber linkages and resisting dimensional change, thereby sustaining functionality in wet conditions during use (Lindström *et al.* 2005; Yuan and Hu 2012; Francolini *et al.* 2023; Seelinger and Biesalski 2023).

Common synthetic WSAs derived from non-renewable resources include urea–formaldehyde (UF) (Husić and Botonjić 2023), melamine–formaldehyde (MF) (Francolini *et al.* 2023), polyethylenimine (PEI) (Sun *et al.* 2015), polyamideamine-epichlorohydrin (PAE) (Sun *et al.* 2015), and polyvinylamine (PVAm) (DiFlavio *et al.* 2005). Among these, PAE is widely used under neutral-to-alkaline conditions (Obokata and Isogai 2007; Zhang *et al.* 2018) and provides permanent wet strength *via* covalent bonding through homo-crosslinking (resin–resin) and co-crosslinking (resin–fiber), typically mediated by reactive azetidinium groups (Obokata and Isogai 2007). These linkages persist even when water disrupts hydrogen bonding, thereby increasing wet tensile strength (Huang *et al.* 2017; Zhang *et al.* 2018; Korpela *et al.* 2022). However, PAE synthesis can generate toxic by-products such as chloropropanediol (CPD) and dichloropropanol (DCP), raising health and environmental concerns (Seelinger *et al.* 2021).

Glyoxalated polyacrylamides (g-PAMs) are another class of synthetic polymers, that are widely considered benchmarks for enhancing dry strength (Chan 1994; Daulplaise and Guerro 1998). Importantly, g-PAM chemistry can be tailored to control both product efficiency and the decay rate of wet strength (Proverb and Pawlowska 2011; Rosencrance *et al.* 2012). A slower decay rate is desirable for towel grades, whereas bath tissue requires rapid disintegration after use to meet flushability expectations. While g-PAMs are widely used in tissue and towel manufacturing and exhibit low polymer toxicity, concerns remain regarding residual acrylamide, aldehyde functionality, and their persistence in recycled fiber systems, motivating the development of greener, alternative chemistries to balance strength performance with recyclability and flushability in hygiene products.

Tissue and towel strength can also be enhanced through the addition of dry-strength agents to fiber suspensions, with additive effectiveness governed by parameters such as charge density, molecular weight (MW), and molecular architecture (Matsushita *et al.* 2004; Rice *et al.* 2018; Bildik Dal *et al.* 2020). Cross-linked polymers represent an important class of strengthening agents formed by interconnecting polymer chains (Allcock *et al.* 1999). Their extended network structures have made them a subject of significant research interest (Hagiopol *et al.* 2005). When applied in papermaking systems, cross-linked polymer chains can interpenetrate fiber–fiber contact regions, forming a more robust paper network that restricts fiber mobility and enhances mechanical strength (Yang *et al.* 2002).

Mechanical treatments can also enhance the dry strength of paper; however, this is often achieved at the expense of bulk and softness (Salem *et al.* 2022, 2023). In contrast, mild mechanical treatments such as high-shear homogenization can preserve bulk while improving strength (Debnath *et al.* 2021). Nevertheless, these approaches generally do not provide adequate wet strength, highlighting the need for complementary chemical strategies.

Beyond the dry and wet strength, faster disintegration is critical for the safe disposal and functionality of hygiene products. Rapid disintegration reduces the risk of sewer clogging and supports efficient passage through wastewater systems. Flushable consumer products can mitigate sanitary sewer overflows (SSOs), which remain a major infrastructure challenge; the United States experiences an annual average of ~50,000 SSOs and ~400,000 basement backups, largely due to pipe blockages (EPA 2001, 2004). Such

events contribute to substantial economic and public health burdens, including outbreaks of waterborne diseases such as cholera, giardiasis, cryptosporidiosis, and hepatitis (Curriero *et al.* 2001).

Previous approaches to improve softness and dispersibility include cationic surfactants (fatty-alkyl quaternary ammonium compounds) used as softeners/debonders, which can promote breakdown through hydrolysis (Poffenberger *et al.* 2000). Remoistened wet wipes treated with PAE or PEI have also been reported to retain functional wet strength while remaining dispersible upon flushing (Maug *et al.* 1997). In addition, soy lecithin (SLP) has been shown to increase bulk and tensile strength by virtue of its amphiphilic nature, which supports interactions with cellulose and promotes a more open sheet structure (Naithani *et al.* 2016). In this study, SLP was included as an amphiphilic interfacial modifier to improve β-CD dispersion and to promote more uniform interactions among β-CD, lignin-rich fiber surfaces, and the aqueous phase. The SLP-containing formulations were used to test whether a bio-based amphiphile could improve the strength-bulk-disintegration balance of β-CD-treated recycled fiber sheets.

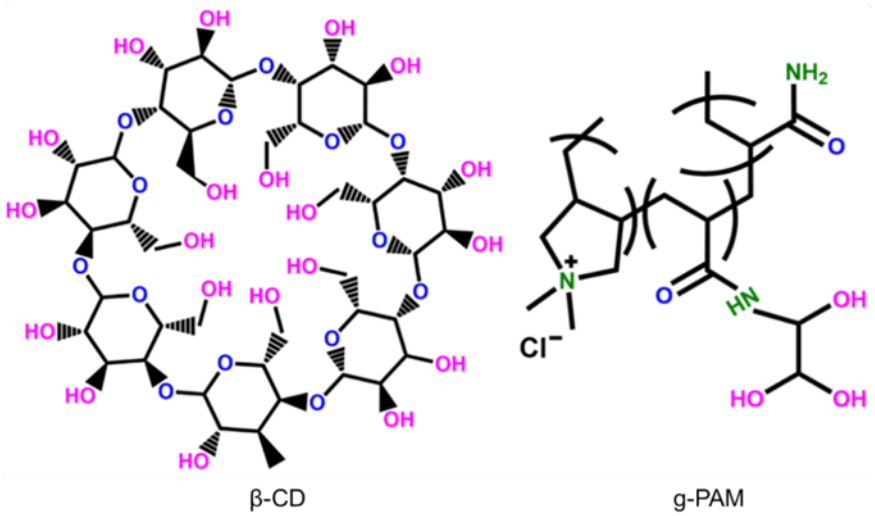

Fig. 1. Chemical structures of glyoxalated polyacrylamide (g-PAM) and β-cyclodextrin (β-CD). g-PAM is a widely used paper strength resin that imparts dry and wet strength through covalent and hydrogen-bond interactions, while β-CD is a cyclic oligosaccharide whose amphiphilic architecture enables simultaneous interactions with hydrophobic lignin domains and hydrophilic cellulose surfaces.

Cyclodextrins are cyclic oligosaccharides produced during the bacterial digestion of amylose. β-CD, composed of seven glucopyranoside units, can associate with both hydrophobic and cationic species in papermaking systems (Dong *et al.* 2014). Owing to its molecular structure, β-CD has been hypothesized to function as a dry and temporary wet-strength agent, potentially forming reversible interactions with cellulose during dehydration. The structures of g-PAM and β-CD are shown in Fig. 1. The toroidal β-CD molecule contains a hydrophobic internal cavity and a hydrophilic exterior, enabling interactions with lignin-containing recycled fibers while maintaining strong affinity for cellulose. Prior studies indicate that cyclodextrins can encapsulate hydrophobic groups and modify the behavior of cationic polyacrylamides, thereby enabling higher-bulk sheet structures while improving dispersibility and processability (Cova *et al.* 2018; Wang and Banerjee 2009; Zhang *et al.* 2019).

Although cyclodextrins are widely used in food, pharmaceutical, and topical formulations, their safety profile must be considered for hygiene products intended for direct skin contact. Cyclodextrins have been extensively investigated as excipients because of their cyclic oligosaccharide structure, inclusion-complex-forming ability, and generally low toxicity when used at appropriate concentrations (Irie and Uekama 1997; Loftsson and Brewster 2010; Musuc 2024). The safety profile of cyclodextrins depends on the route of exposure, applied concentration, and specific cyclodextrin type used. Dermal exposure is generally considered lower risk due to the limited penetration of cyclodextrins through intact skin (Irie and Uekama 1997). In dermal studies, β-CD did not induce irritation or allergic contact dermatitis in repeated insult occlusive patch testing in human volunteers; nevertheless, high concentrations of cyclodextrins may extract skin lipids and cause irritation (Irie and Uekama 1997). In the present study, β-CD was applied at relatively low wet-end dosages of 3 to 9 lb/ton, corresponding to approximately 0.15 to 0.45 wt.% based on dry fiber. Therefore, the β-CD levels used in this work are expected to present low dermal safety risk, although final product-level irritation, sensitization, and extractable-residue testing would be required before use in consumer hygiene products, particularly for sensitive populations such as infants.

Accordingly, this study evaluated β-CD as a more sustainable, bio-based additive for enhancing dry strength, temporary wet strength, and disintegration in OCC-derived recycled fibers for hygiene tissue and towel applications. The performance of β-CD was benchmarked against g-PAM and soy lecithin (SLP) under both single- and dual-stage treatment strategies (Pal *et al.* 2022).

EXPERIMENTAL

Materials

Glyoxalated polyacrylamide (cationic g-PAM; Fennorez® 110, 11.5% solids) was commercially sourced and used as a wet-strength resin. Commercial old corrugated container (OCC) pulp was obtained from a tissue mill in North Carolina (USA). Per mill specification, OCC pulp was refined to 170 mL Canadian Standard Freeness (CSF) using a Valley beater. β-cyclodextrin (β-CD; Cavamax W7, molecular weight: 1134.98; Wacker Chemicals, USA) was obtained from Sigma-Aldrich (USA). Soy lecithin (SLP; Performix® E) was supplied by Archer Daniels Midland (ADM).

Methods

For the refining of OCC pulp, the TAPPI T 200 standard method (Valley beater) was used, while the pretreated OCC (POCC) pulp was refined using the TAPPI T 248 method (PFI mill). Paper towel sheets (target basis weight: 40 g m^{-2}) were prepared following TAPPI T 205 sp-02 (2006), with modifications: couching was performed using a lightweight foam roller (~0.15 kg), and pressing and ambient drying steps were omitted. After couching, paper towel sheets were drum-dried twice at 105 °C and approximately 10 to 12 ft/min (3.0 to 3.7 m/min). Paper towel sheets were conditioned at 23 °C and 50% relative humidity (RH) prior to testing. Chemical dosages (3, 6, and 9 lb ton^{-1}) were mixed with fibers prior to sheet formation. Pulp and sheet properties were measured using TAPPI standards: freeness (T 227), basis weight (T 410), thickness (T 411), dry tensile strength (T 494), and wet tensile strength (T 456).

Single-stage treatment

OCC pulp was refined using a lab-scale Hollander-style beater (Valley Machinery, Inc.). Pulp was dispersed for 30 min under no load and then refined to 170 mL CSF (control OCC freeness: 460 mL). Pulp pH was adjusted to 7 using 10% HCl or NaOH. Additives were mixed into 4% consistency stock for 5 min using a laboratory blender and then diluted to 0.3% consistency for paper towel sheets preparation (Fig. 2a).

Dual-stage treatment

OCC pulp was autohydrolyzed at 160 °C for 30 min in a finger reactor. After autohydrolysis, the pulp was cooled to room temperature, washed with water, and then PFI-refined at ambient laboratory temperature (23 ± 3 °C) to 180-200 mL CSF. This pulp is referred to as pretreated OCC (POCC). Additives were mixed for 5 min using a laboratory blender and diluted to 0.3% consistency for paper towel sheets preparation (Fig. 2b).

The β-CD is water soluble, its wet-end retention is expected to depend strongly on furnish composition, addition point, mixing, drainage rate, and the presence of retention aids. Unlike cationic polyelectrolytes, unmodified β-CD does not rely primarily on electrostatic fixation; instead, partial retention is expected through hydrogen bonding with cellulose and inclusion interactions with lignin-rich or hydrophobic domains present in OCC fibers.

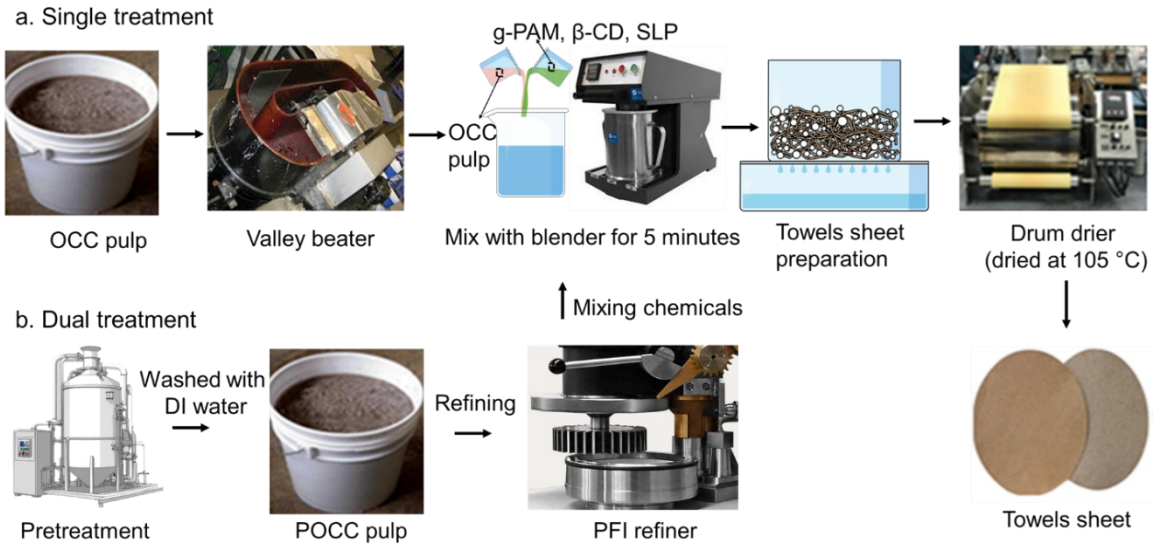

Fig. 2. Schematic illustration of single- and dual-stage treatment strategies for paper towel sheets preparation from recycled fibers. (a) Single-stage treatment involves refining OCC pulp followed by addition of g-PAM, β-CD, or soy lecithin (SLP), paper towel sheets formation, and drum drying. (b) Dual-stage treatment incorporates autohydrolysis pretreatment to produce POCC pulp, followed by PFI refining, chemical addition, paper towel sheets preparation, and drying.

Fiber Quality Analysis

Fiber properties were measured using a HiRes Fiber Quality Analyzer (FQA; OpTest Equipment Inc., Hawkesbury, Ontario, Canada). Fiber length (L_w), fines content, and related physical characteristics were recorded following instrument calibration per manufacturer guidance. All pulp samples (refined and unrefined OCC and POCC) were disintegrated prior to measurement.

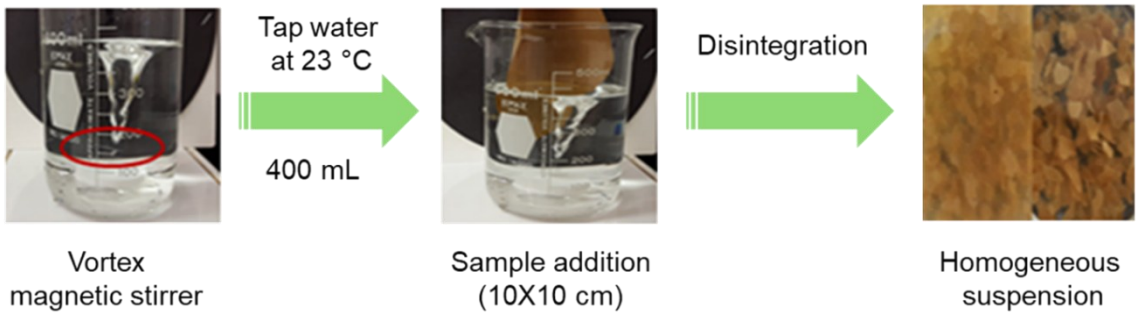

Fig. 3. Disintegration test procedure used to evaluate flushability. A 10 × 10 cm paper towel sheets sample was introduced into a vortex formed in 400 mL of tap water at 23 ± 3 °C under magnetic stirring, and the time required to reach a homogeneous suspension was recorded.

Disintegration Test

A 500 mL beaker containing a magnetic stir bar was filled with 400 mL tap water maintained at 23 ± 3 °C. The beaker was placed on a magnetic stir plate and stirred until a vortex formed to a depth corresponding to the 150 mL mark. At least ten 10 cm × 10 cm samples were prepared from paper towel sheets and introduced (unfolded) into the center of the vortex (Fig. 3). Disintegration time was recorded using a stopwatch. Disintegration was considered complete when the suspension appeared homogeneous and all fragments were smaller than ¼ inch, with no large visible pieces remaining.

RESULTS AND DISCUSSION

Fiber morphology

Fiber morphology was evaluated for OCC and POCC pulps, with and without refining. The results are summarized in Table 1.

Table 1. Fiber Properties of Unrefined and Refined OCC and POCC Pulp Fibers (OCC: Old Corrugated Container Pulp; POCC: Pretreated OCC; R: Refined)

Properties/ Sample IDs	OCC	OCC-R	POCC	POCC-R
Pretreatment (P)	No	No	Yes	Yes
Refining (R)	No	Yes	No	Yes
Freeness, CSF (mL)	460	170	597	208
Fiber Length L_w (mm)	1.54	1.51	1.4	1.15
Fines (%)	42.6	47.2	41.8	44.2
Curl Index (L_w)	0.07	0.06	0.16	0.08
Mean width (μm)	22.5	22.95	21.8	22.3
Kink Index (1/mm)	1.19	1.08	2.25	1.44

POCC exhibited the highest freeness (597 mL) relative to OCC (460 mL). This increase in freeness is more likely to be attributed to the loss of fines and removal of soluble or loosely bound components during autohydrolysis pretreatment, which reduced the water-retaining fraction of the pulp and enhanced drainage. In contrast, refining substantially decreased freeness, with OCC-R showing the lowest value (170 mL), consistent with increased external fibrillation, fines generation, and improved fiber hydration.

Fiber length decreased with pretreatment and refining (from 1.54 mm in OCC to 1.15 mm in POCC-R), while fines increased, reaching 44.2% in POCC-R. Pretreatment increased curl and kink indices, while fiber width decreased slightly. Morphological parameters such as fiber length, fines content, curl, kink, and width are strongly influenced by feedstock and pulping history (Salem *et al.* 2021).

Bulk and Dry Tensile Strength

Bulk and dry tensile properties (Table 2; Fig. 4) were strongly dependent on additive type and dosage. g-PAM-treated sheets exhibited a consistent increase in dry tensile index with medium dosage, which can be attributed to enhanced fiber–fiber bonding through hydrogen bonding and covalent interactions. Specifically, aldehyde groups present in g-PAM may react with cellulose hydroxyl groups to form hemi-acetal linkages, contributing to network reinforcement and initial strength development. Because these hemi-acetal linkages are water-sensitive and reversible upon rewetting, g-PAM is generally associated with temporary wet-strength development (Allcock *et al.* 1999). In agreement with this mechanism, g-PAM mainly promoted network densification and therefore produced slight increases in bulk at low dosage.

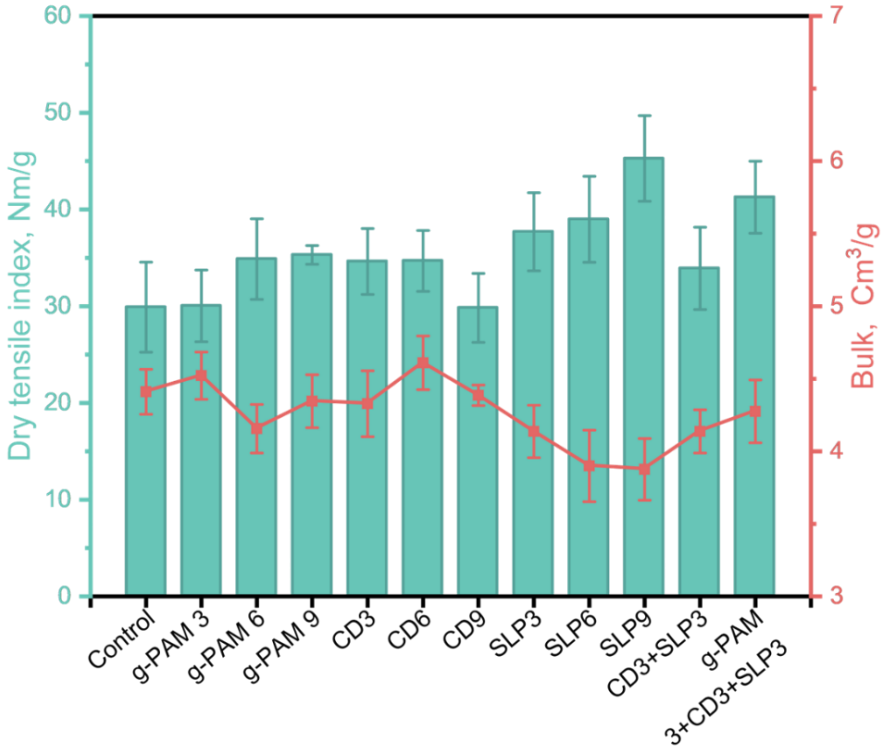

Fig. 4. Effect of chemical additives on bulk and dry tensile index of OCC paper towel sheets. Treatments include varying dosages of g-PAM, β-CD, SLP, and combined formulations. Control samples represent untreated OCC sheets.

When β-CD was added to refined OCC fiber suspensions during paper towel sheet formation, the resulting sheets exhibited a distinct bulk-strength balance compared with g-PAM-treated sheets. At 3 to 6 lb per ton, β-CD increased bulk relative to g-PAM while maintaining comparable dry tensile performance (Fig. 4). This behavior is consistent with β-CD's dual-interaction mechanism: the higher bulk is not attributed to the molecular size

of β-CD itself, but rather to its indirect influence on fiber-network consolidation during drying. By associating with lignin-rich or hydrophobic domains and presenting a hydrophilic exterior, β-CD may improve water access and reduce hydrophobe-driven fiber collapse, thereby preserving a more open sheet structure (Wang and Banerjee 2009; Wang *et al.* 2014; Naithani *et al.* 2017). At 9 lb per ton, β-CD reduced dry tensile index, which is consistent with excessive disruption of fiber–fiber bonding and reduced effective contact area (Fig. 5) (Naithani *et al.* 2017). The addition of soy lecithin improved β-CD performance at higher dosage, likely by promoting dispersion and more uniform adsorption; amphiphilic interactions can mitigate the reduction in tensile associated with high-bulk additives (Yessine and Leroux 2004).

Table 2. Physical Properties of OCC Fibers-based Paper Towel Sheets Treated with Different Chemical Additives (g-PAM, β-CD, SLP, and Combinations)

Sample ID	Dosage (lbs/ton)	Basis Weight (g/m^2)		Caliper (µm)		Bulk (cm^3/g)	
		Avg.	Std.	Avg.	Std.	Avg.	Std.
OCC (control)	0	40.86	0.02	180.84	3.04	4.41	0.15
g-PAM	3	39.99	0.02	178.81	5.33	4.52	0.16
	6	39.22	0.02	169.92	5.58	4.15	0.16
	9	39.35	0.02	172.97	6.60	4.34	0.18
β-CD	3	40.07	0.03	181.86	2.28	4.32	0.28
	6	37.92	0.01	179.57	5.84	4.60	0.18
	9	39.22	0.01	172.21	2.54	4.38	0.07
SLP	3	41.38	0.01	173.22	4.31	4.13	0.18
	6	41.86	0.01	166.87	7.11	3.89	0.36
	9	43.82	0.02	173.99	7.62	3.87	0.21
β-CD+SLP	3+3	42.40	0.02	171.45	6.60	4.13	0.14
β-CD+SLP+ g-PAM	3+3 +3	41.40	0.01	176.53	9.14	4.27	0.21

The increased bulk observed for β-CD-containing sheets may also be relevant to hygiene-product absorbency. Recycled OCC fibers are relatively stiff because of prior hornification and residual lignin; therefore, additives that increase water access and preserve an open network are desirable. The hydrophilic exterior of β-CD can promote fiber hydration and water penetration, whereas the hydrophobic cavity can associate with lignin-rich or other hydrophobic domains. SLP can act as an amphiphilic interfacial modifier, improving β-CD dispersion and potentially reducing localized fiber collapse during drying.

Immediate Wet Tensile Index (IWTI)

Wet strength development in paper commonly requires improving inter-fiber adhesion by adding polymers that adsorb onto fiber surfaces and reinforce fiber–fiber contact points (Gimåker *et al.* 2011). Figure 6 compares immediate wet tensile index (IWTI) responses across additives and combinations. The immediate wet tensile index of g-PAM-treated sheets did not increase substantially with increasing g-PAM dosage from 3 to 9 lb/ton, suggesting that wet-strength development reached a plateau within this dosage range. This behavior may be attributed to saturation of available anionic sites on the fiber surfaces and possible overcharging of the system at higher g-PAM dosages. Once the fiber surface is sufficiently covered with cationic g-PAM, additional polymer may not contribute effectively to fiber-fiber bonding and may instead promote electrostatic repulsion or less efficient polymer distribution. Therefore, the immediate wet tensile response is likely

governed not only by the amount of g-PAM added, but also by its retention, distribution, and effective interaction with cellulose through electrostatic attraction, hydrogen bonding, and hemi-acetal linkage formation (Allcock *et al.* 1999). Notably, β-CD achieved comparable IWTI at medium to high dosages (6 to 9 lb per ton), indicating that β-CD adsorption and its coupled hydrophobic/hydrophilic interactions can stabilize fiber bonding during initial wet exposure. This behavior is consistent with β-CD's affinity for lignin-containing domains and its capacity to create a balanced network of hydrophobic association and hydrophilic bonding, thereby supporting wet integrity during use (Wang and Banerjee 2009) (Fig. 6). SLP further improved β-CD performance, consistent with its surfactant-like amphiphilicity and ability to promote more uniform β-CD dispersion and adsorption on fiber surfaces, thereby enhancing wet strength without sacrificing disintegration tendencies (Yessine and Leroux 2004).

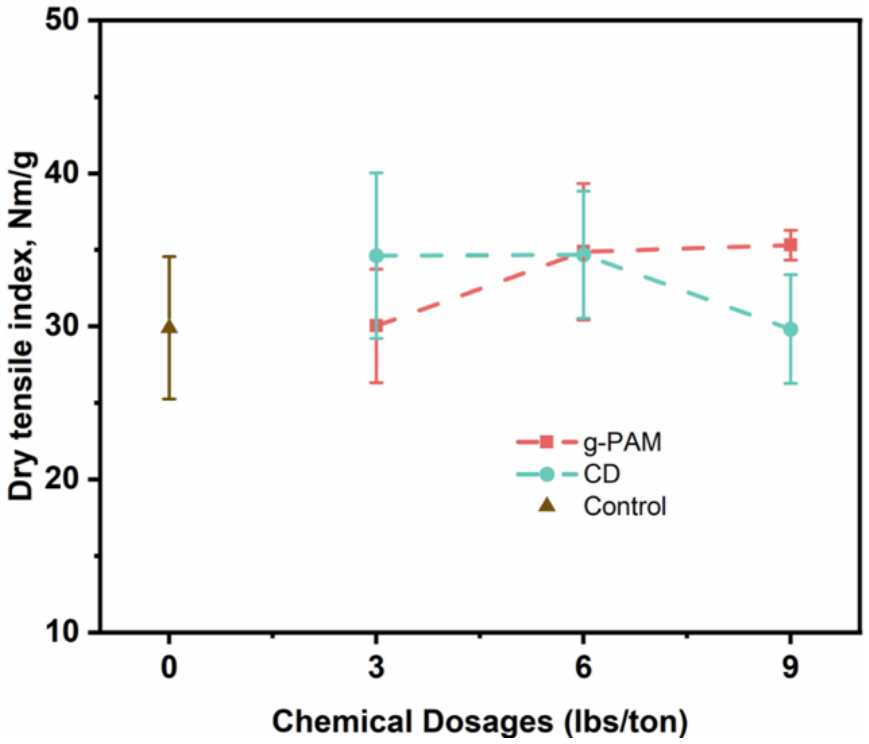

Fig. 5. Influence of β-CD and g-PAM dosage on the dry tensile index of recycled-fiber paper towel sheets. g-PAM exhibits an initial increase in tensile strength followed by plateauing at higher dosages, whereas β-CD improves strength at low-to-medium dosages but shows a decline at higher levels due to reduced effective fiber–fiber bonding.

Dual-stage Strategy (POCC): Balancing Dry and Wet Strength

To address strength limitations associated with recycled fibers while maintaining rapid disintegration, a dual-stage strategy was evaluated: OCC was first autohydrolyzed (160 °C, 30 min) to produce POCC, then refined to ~180 to 200 mL CSF and finally treated with β-CD and/or g-PAM prior to sheet formation. The autohydrolysis pretreatment increased freeness relative to untreated OCC, consistent with improved fiber hydration and partial removal of hydrophobic contaminants, followed by controlled fibrillation during PFI refining.

Figures 7 through 9 summarize the performance of dual-stage systems. Bulk remained within a relatively narrow range across POCC-based samples, indicating that the

treatment sequence preserved a porous sheet structure suitable for towel-grade applications. POCC exhibited moderate dry tensile index, which decreased slightly upon β-CD addition (POCC+CD), consistent with partial surface coverage and reduced fiber–fiber contact area. However, introducing a small amount of g-PAM after β-CD treatment (POCC+CD+g-PAM) resulted in a pronounced increase in dry tensile index, consistent with polymer bridging and enhanced stress transfer facilitated by a β-CD-conditioned interphase (Wang and Banerjee 2009; Naithani *et al.* 2017).

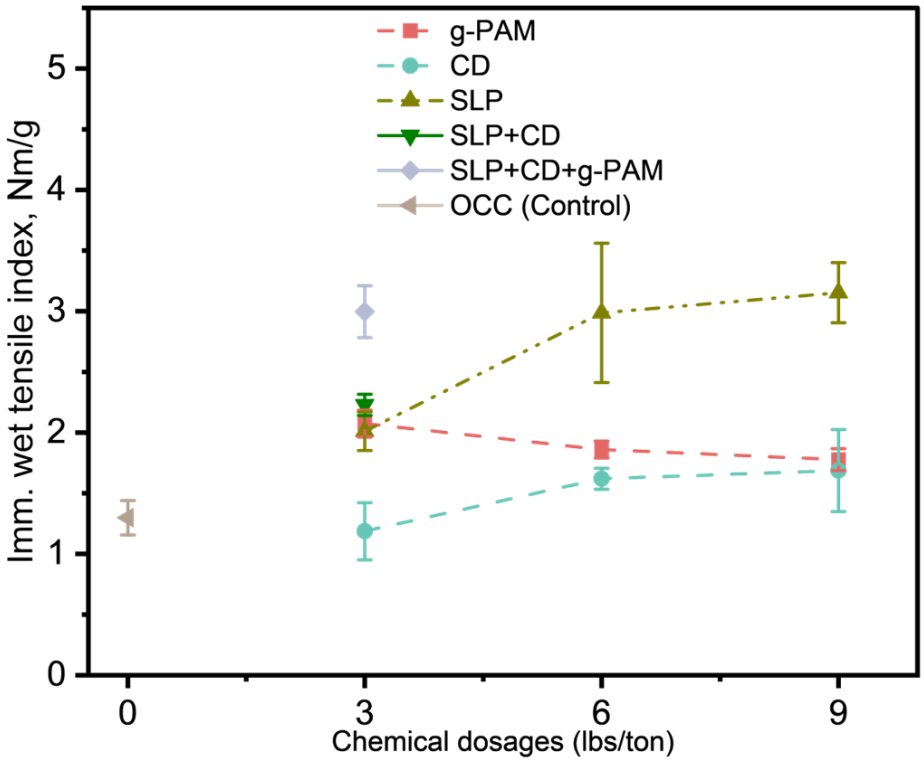

Fig. 6. Immediate wet tensile index (IWTI) of paper towel sheets treated with g-PAM, β-CD, SLP, and combined systems. β-CD achieved wet tensile performance comparable to g-PAM at optimized dosages, while SLP enhanced β-CD effectiveness through improved dispersion.

IWTI results further highlight the value of dual-stage treatment. As shown in Table 3 and Fig. 8, a formulation containing 9 lb per ton β-CD + 1 lb per ton g-PAM achieved IWTI comparable to higher-dosage g-PAM-only systems, indicating a synergistic effect in which β-CD contributes to wet integrity while limiting excessive crosslinking that would otherwise impede disintegration. In contrast, g-PAM-only systems provide robust wet strength but generally do not meet the disintegration requirements associated with flushable products, particularly when wet strength persists beyond the intended usage window (Ferguson 1992; Allcock *et al.* 1999).

These results validate the effectiveness of the dual-treatment strategy in addressing limitations identified in earlier studies. Ferguson (1992) reported significant strength loss in recycled fibers, while Allcock *et al.* (1999) noted the inability of g-PAM systems to achieve rapid hydrolysis and disintegration (Allcock *et al.* 1999; Ferguson 1992). By leveraging the encapsulation properties of β-CD (Wang and Banerjee 2009) in combination with fiber pretreatment effects (Horn 1975), this study demonstrates a scalable and sustainable approach for developing high-performance, flushable hygiene products.

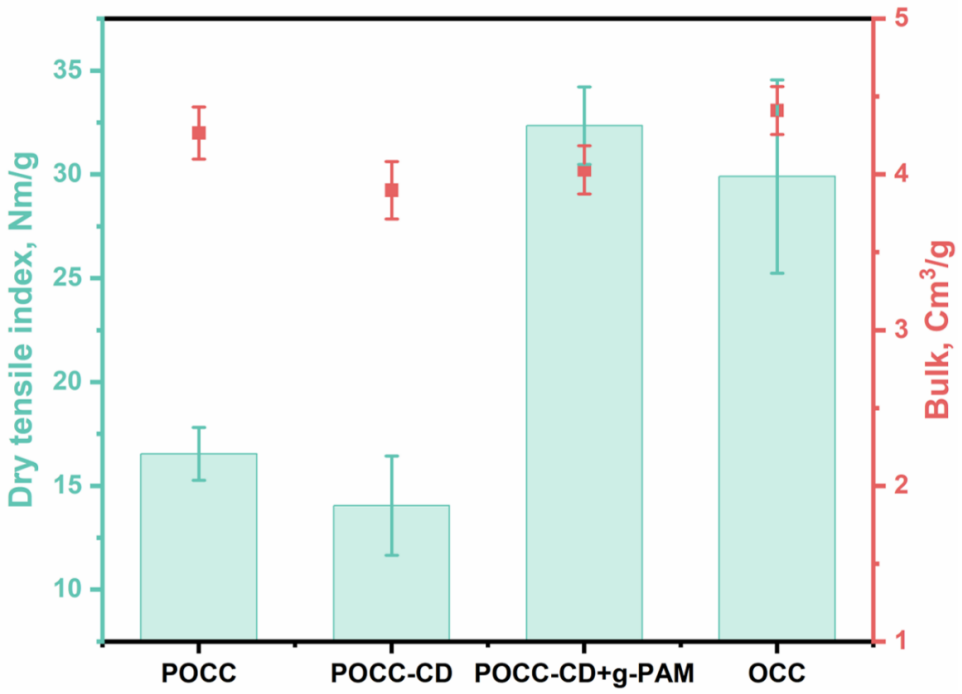

Fig. 7. Effect of β-CD and g-PAM on bulk and dry tensile index of POCC-based paper towel sheets. The combined β-CD + g-PAM system exhibited a favorable balance between bulk and significant tensile strength enhancement compared to β-CD-only treatment.

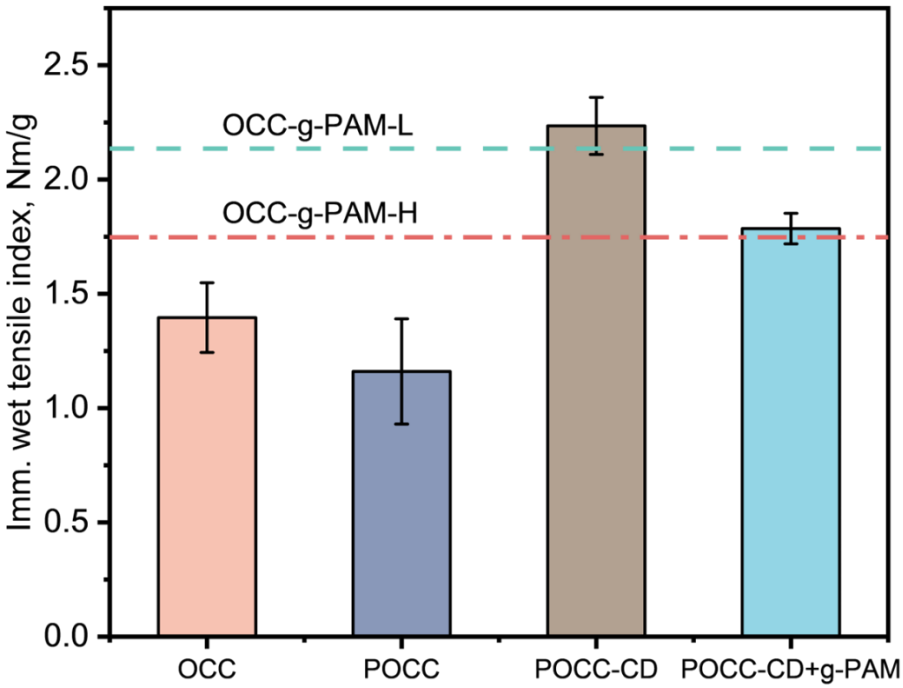

Fig. 8. Comparison of immediate wet tensile index for dual-stage β-CD + g-PAM systems relative to g-PAM-only controls. A formulation containing 9 lb per ton β-CD and 1 lb per ton g-PAM achieved IWTI comparable to higher-dosage g-PAM systems.

Table 3. Immediate Wet Tensile Index of OCC and POCC Systems under Selected β-CD and g-PAM Formulations

Sample ID	Dosage (lb/ton)	Immediate Wet Tensile Index (Nm/g)	
		Avg.	Std.
OCC	0	1.39	0.15
OCC + g-PAM -Low	3	2.07	0.11
OCC + g-PAM -High	9	1.80	0.08
POCC	0	1.16	0.23
POCC + CD	9	2.23	0.12
POCC + CD + g-PAM	9 + 1	1.78	0.06

Disintegration Mechanisms and Interphase Design: Protection *vs.* Reinforcement

Figure 9 illustrates the wet tensile index of POCC pulp at 2400 seconds in comparison with OCC pulp, highlighting the influence of β-CD and g-PAM, both individually and in combination. The OCC reference exhibits a moderate wet tensile index, serving as a benchmark for evaluating the performance of modified POCC systems. Disintegration behavior (Fig. 9) reinforces the dual-function benefit of β-CD.

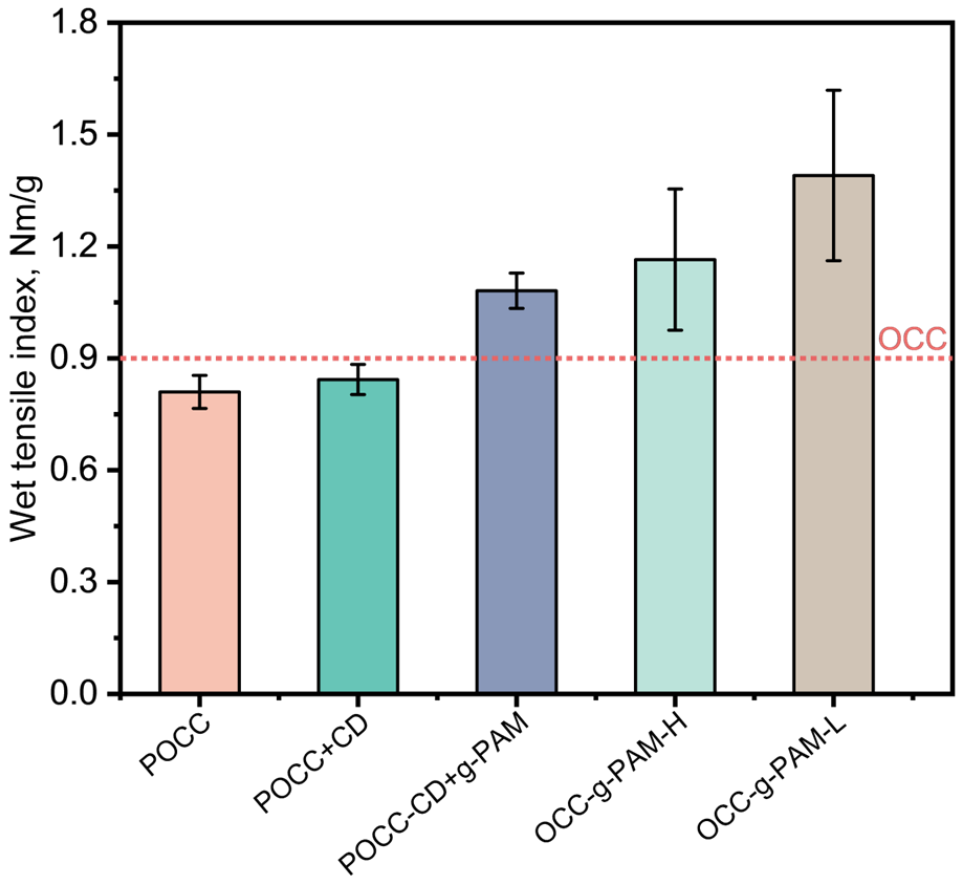

Fig. 9. Effect of β-CD and g-PAM on the wet tensile index of paper towel sheets at 2400 seconds. When combined with g-PAM, β-CD exhibits a synergistic effect that improves wet strength performance. This behavior is attributed to the hydrophilic exterior of β-CD, which promotes rapid water penetration into the fiber network, thereby reducing disintegration time. Enhanced water uptake weakens excessively strong fiber–fiber bonding, resulting in a balanced network structure that facilitates improved re-dispersibility.

The reduction in disintegration time is consistent with β-CD's hydrophilic exterior promoting water penetration and swelling, thereby weakening excessive fiber–fiber bonding and accelerating fiber separation during repulping or flushing. Furthermore, the increased water uptake induced by β-CD accelerates fiber rewetting and reduces disintegration time, which is critical for towel-grade products requiring both wet strength and re-dispersibility. Additionally, β-CD may partially interact with g-PAM, thereby moderating the strengthening efficiency of g-PAM. Since β-CD contains multiple exterior hydroxyl groups, some glyoxal/aldehyde groups of g-PAM may form hydrogen-bonded or reversible hemi-acetal/acetal-type interactions with β-CD instead of reacting exclusively with cellulose hydroxyl groups. This may reduce the amount of g-PAM available for direct fiber–fiber reinforcement (Francolini *et al.* 2023). However, this effect is beneficial for the present application because g-PAM is intended to provide temporary wet strength, and partial β-CD/g-PAM interaction may help maintain sufficient wet integrity while allowing rapid disintegration during flushing.

Figure 10 compares the disintegration behavior of selected systems. The rapid disintegration observed for POCC-based sheets within ~60 s indicates that the proposed approach can maintain re-dispersibility while still providing wet integrity during use. Two canonical mechanisms are typically invoked to explain wet-strength development in paper systems. In the protection mechanism, the additive self-crosslinks (or forms an insoluble network) around fibers and contact regions, limiting fiber swelling and separation upon rewetting and thereby preserving a fraction of dry strength. In the reinforcement mechanism, covalent linkages form between the additive and cellulose/hemicellulose, supplementing hydrogen bonding and creating water-resistant bonding at fiber–fiber contacts. Because covalent linkages are not disrupted by water, reinforcement can provide substantial wet strength. However, wet-strength systems can also reduce fiber–fiber bonding during recycling, and strength additives are generally most effective when they enhance fiber–fiber contact rather than inhibit it (Kamel *et al.* 2004).

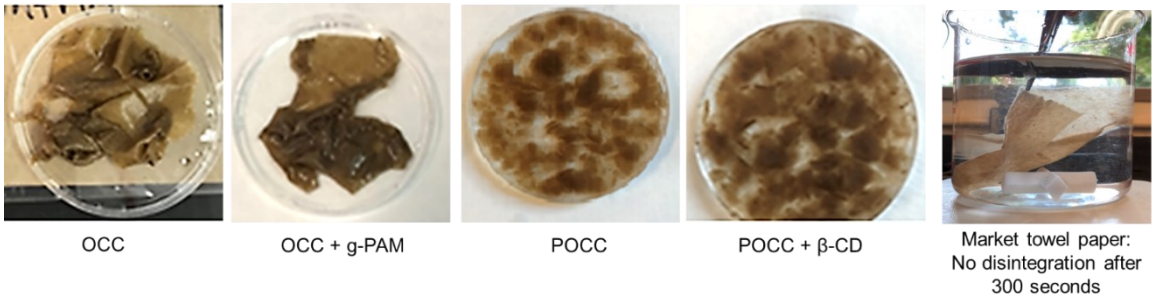

Fig. 10. Comparison of disintegration behavior for OCC, POCC, OCC + g-PAM, and POCC + β-CD and POCC and POCC + β-CD paper towel sheets after 60 seconds. The POCC + β-CD system demonstrates rapid disintegration despite improved wet strength, indicating suitability for flushable towel applications. As a reference, commercial market towel paper doesn't disintegrate even after 300 seconds.

From a practical perspective, commercial bath tissue is engineered to lose wet strength rapidly and disperse during flushing; however, it typically has a lower basis weight and substantially lower wet integrity than towel products. In contrast, the present POCC/ β-CD sheets were prepared at towel-relevant basis weight and retained measurable immediate wet tensile strength, while still reaching a homogeneous suspension within approximately 60 s. Thus, the main advantage of the proposed system is the combination

of towel-like wet functionality with disintegration behavior that approaches the dispersibility expected for flushable bath-tissue products. Under the same disintegration test, commercial towel paper did not fully disintegrate even after 300 s, whereas commercial bath tissue completely disintegrated within 120 s.

Proposed Mechanism: β-CD–Lignin Inclusion, Cellulose Hydrogen Bonding, and Controlled Reinforcement with g-PAM

The performance of β-CD in these recycled-fiber systems is consistent with coupled interfacial interactions among β-CD, lignin-containing domains, cellulose, SLP, and g-PAM. The hydrophobic cavity of β-CD may associate with relatively small lignin-derived aromatic or hydrophobic moieties through van der Waals and hydrophobic interactions, whereas hydroxyl groups present in lignin-related structures may interact with the outer hydroxyl surface of β-CD through hydrogen bonding. Because of the limited cavity size of β-CD, complete inclusion is expected mainly for low-molecular weight lignin-derived compounds rather than polymeric lignin or larger lignin oligomers. These interactions may reduce hydrophobe-driven interference with water uptake and facilitate fiber swelling. (Wang and Banerjee 2009; Wang *et al.* 2014). Concurrently, β-CD's hydrophilic exterior forms hydrogen bonds with cellulose, promoting water diffusion into the sheet network and weakening excessively strong fiber–fiber interactions during immersion, thereby accelerating disintegration.

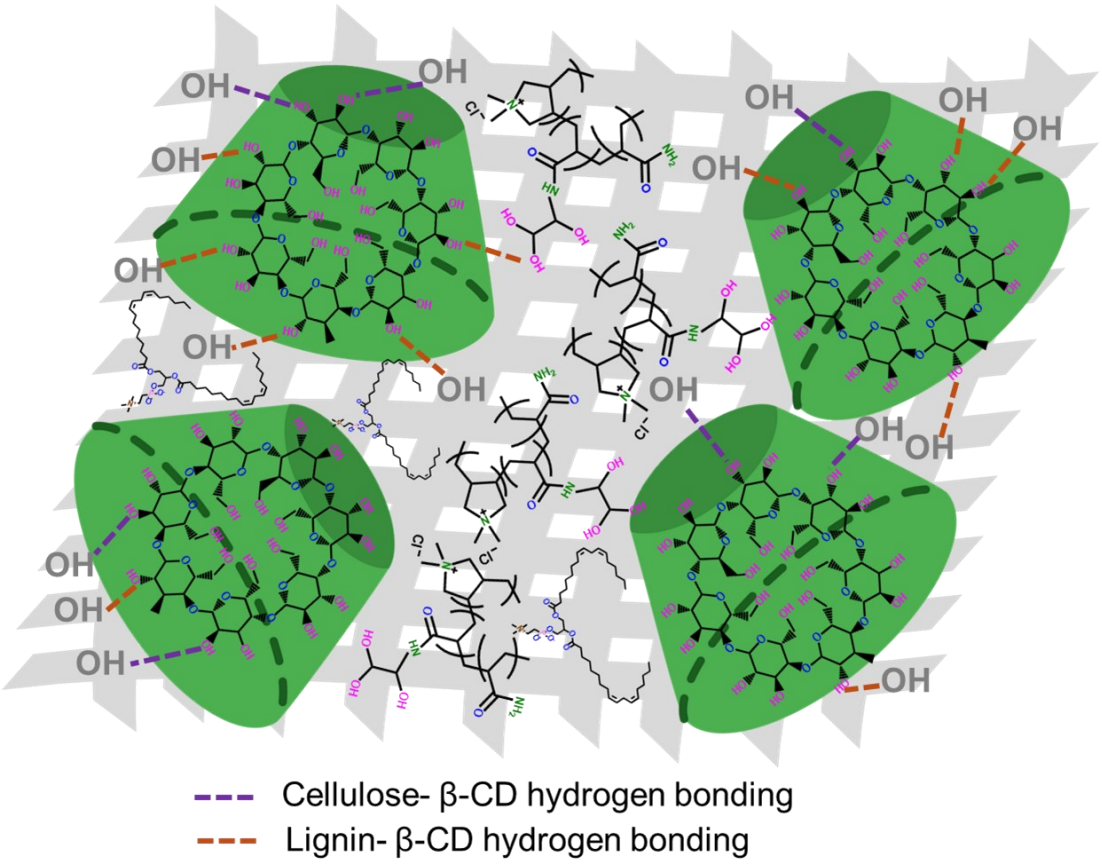

Fig. 11. Proposed mechanism illustrating synergistic interactions among β-CD, g-PAM, SLP, and recycled fibers. β-CD encapsulates hydrophobic lignin domains while forming hydrogen bonds with cellulose, promoting fiber swelling and rapid disintegration. g-PAM provides controlled wet-strength reinforcement, and SLP enhances dispersion and interfacial interactions.

Soy lecithin enhances this mechanism by acting as an amphiphilic dispersant: hydrophobic tails can associate with the β-CD cavity, while hydrophilic headgroups promote favorable interactions at the fiber–water interface. This synergistic amphiphilic behavior is consistent with membrane- and interface-active effects reported for related amphiphilic polymer and lipid systems (Yessine and Leroux 2004; Mecke *et al.* 2005).

In dual-stage systems, g-PAM provides wet-strength reinforcement through aldehyde-mediated bonding with cellulose (Allcock *et al.* 1999). Importantly, β-CD adsorption at fiber–fiber contact points appears to moderate excessive g-PAM crosslinking, thereby preserving wet strength during use while enabling faster fiber separation during disintegration. This supports β-CD's dual functionality as both a bulk/structure modifier and a disintegration promoter (Fig. 11).

CONCLUSIONS

1. This work demonstrates a cyclodextrin-enabled approach to simultaneously improve dry strength, provide temporary wet strength, and enhance disintegration/flushability in recycled-fiber towel systems.
2. Two application strategies were evaluated: a single-stage approach applied to refined old corrugated container (OCC) pulp and a dual-stage strategy combining autohydrolysis pretreatment (POCC) with additive treatments. In the single-stage OCC systems, beta-cyclodextrin (β-CD) at medium dosage (≈6 lb per ton) produced immediate wet tensile index (IWTI) values of approximately 1.4 to 1.5 N m/g, comparable to benchmark g-PAM) systems, while supporting faster disintegration. β-CD also improved bulk and maintained dry tensile performance at 3–6 lb/ton, consistent with its dual interaction mechanism. At higher dosage (≈9 lb/ton), dry tensile index decreased due to reduced effective fiber–fiber bonding, which was mitigated by incorporating soy lecithin, which likely improved β-CD dispersion and adsorption uniformity.
3. In the dual-stage POCC systems, combining β-CD with a small g-PAM dose enabled wet-strength performance comparable to g-PAM-only systems while substantially improving disintegration. A formulation containing 9 lb per ton β-CD + 1 lb/ton g-PAM achieved IWTI comparable to higher-dosage g-PAM-only controls, while disintegration time was reduced to approximately 60 seconds, substantially faster than typical commercial towel grades. The accelerated disintegration is attributed to β-CD's hydrophilic exterior promoting water penetration and fiber swelling, weakening excessive bonding and enabling rapid fiber separation, while g-PAM provides controlled reinforcement during use.
4. Overall, the results support β-CD as a promising, more sustainable additive strategy for developing recyclable and flushable recycled-fiber hygiene products that maintain dry and immediate wet strength during use yet disintegrate rapidly during flushing or repulping.
5. Achieving optimal disintegration remains sensitive to furnish properties and mixing uniformity, including the specific surface area of the fiber network and the balance of hydrophilic/hydrophobic interactions associated with lignin content and additive dispersion.

6. From a scale-up perspective, the autohydrolysis step in dual-stage treatment adds thermal demand relative to direct wet-end chemical addition; therefore, heat integration and residence-time optimization will be important for industrial implementation. In a mill setting, the process could potentially be integrated with existing steam or hot-water loops and heat-recovery systems. The additional energy requirement may be offset by using low-cost recycled OCC furnish, improving fiber hydration and drainage behavior, reducing basis weight, and lowering the required dosage of synthetic wet-strength resin. A detailed techno-economic assessment should be conducted to develop a viable commercial pathway.

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