

Preliminary Study on Feasibility of Manufacturing Injection-molded Composite Using Cellulosic Materials

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Wastepaper-derived fibers, natural starch binders, and micro-fibrillated cellulose (MFC) additives were preliminarily studied as key components in injection-moldable bio-composite formulations. Commercially available paper cups coated with polyethylene, corn starch, and MFC prepared in a laboratory were selected as raw materials. Since the injection molding machine is yet to be developed, handsheets were prepared as a substitute for the future injection-molded composites, and their physical properties were evaluated to assess the promising formulation candidates of recycled fibers, binder, and reinforcing agent. The optimal paper cup powder–starch ratio was 60:40, which balanced the tensile strength, elongation at break, and burst strength of the material while maintaining good processing characteristics and avoiding problems related to dewatering and adhesion to equipment during drying. The incorporation of up to 5% (by weight) MFC considerably improved the mechanical properties of the sheets by enhancing their fiber–fiber bonding. However, increasing the MFC content above 5% prolonged the dewatering time, compromising process efficiency, and making handsheet preparation impossible. These findings suggested that used paper cups, when combined with starch and MFC, could be effectively recycled into injection-molded composite materials, thereby contributing to environmental conservation and the advancement of resource circulation in manufacturing.

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Keywords: Injection-molded composite; Recycled paper; Paper cup; Starch; Micro-fibrillated cellulose (MFC)

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INTRODUCTION

Driven by the increasing global awareness of environmental issues, industries worldwide are transitioning from conventional petroleum-based plastics to more sustainable and biodegradable alternatives (Nanda *et al.* 2022; Zhang *et al.* 2022). This transformation has been spearheaded by the packaging industry, a major source of plastic waste generation (Reddy *et al.* 2013; Shafqat *et al.* 2020). Among the most promising alternatives to traditional plastic packaging are molded pulp products derived from renewable, cellulose-based raw materials, which are fully biodegradable and recyclable (Didone *et al.* 2017). These products have gained popularity on account of their minimal environmental footprint, cost-effectiveness, and alignment with circular economic principles (Didone *et al.* 2017; Su *et al.* 2018; Nanda *et al.* 2022).

Packaging items such as trays, clamshells, and protective inserts have been successfully produced by conventional pulp molding technologies such as wet-press molding, thermoforming, and transfer molding (Semple *et al.* 2022). Despite their commercial success, these methods are unsuitable for high-performance applications (Didone *et al.* 2017; Semple *et al.* 2022). Owing to inconsistent dimensional stability, long cycle times, low mold durability, and limited design flexibility, pulp-molded items cannot compete with precision-formed plastic components (Didone *et al.* 2017; Danielewicz, 2025). Moreover, the existing pulp molding technologies must evolve alongside the market demand, which is shifting toward customized, durable, and structurally complex bioproducts (Semple *et al.* 2022).

To overcome these limitations, researchers and industries are exploring the potential of injection-molding technology for lignocellulosic-based composites (Pacheco *et al.* 2023). Injection molding, which is well-established in the plastics industry, is an attractive option for next-generation bio-based product manufacturing because it offers high dimensional accuracy, reproducibility, fast cycle times, and compatibility with automation (Pacheco *et al.* 2023). However, as traditional injection-molding machines are optimized for thermoplastic resins, they are inherently incompatible with lignocellulosic materials (Tsegaye *et al.* 2024; Montanes *et al.* 2019). Unlike synthetic polymers, cellulosic fibers degrade at elevated temperatures and flow poorly in molten states, necessitating innovative material formulations and process designs (Tsegaye *et al.* 2024).

Recent technological advances have promised the integration of natural fibers into injection-molded components (Rabbi *et al.* 2021). UPM's ForMi composites (Create Green, n.d.), PulPac's dry-molded fiber technology (PulPac, n.d.), and Sulapac–Nissha's biodegradable molding materials (Sphera 2023) exemplify the adaptation of lignocellulosic fibers to thermoplastic-compatible molding processes. Nonetheless, these methods often require thermoplastic matrix polymers and are prone to incomplete fiber dispersion, fiber–matrix incompatibility, thermal degradation, and process inefficiencies (Zhang *et al.* 2022). These shortcomings have limited the adoption of fiber-based molding in applications requiring performance and environmental sustainability (Bordón *et al.* 2022; Prasad *et al.* 2024).

To bridge this critical gap, the authors have engineered a dedicated injection-molding platform for fiber-dominant, lignocellulosic feedstocks. Unlike hybrid composites that rely heavily on thermoplastics, this approach pairs the high-value utilization of waste-derived cellulosic fibers with natural, biodegradable binders and functional additives, thereby enabling a fully bio-based injection-molding process. The aim is to upcycle low-recycling-rate materials such as used paper cups, poly-coated milk cartons, and aseptic beverage cartons into functional injection-molding feedstock, in this way diverting them from landfill or incineration streams and creating added value within a circular economy framework.

To develop a fiber-based injection-molding process, it is necessary to identify suitable fiber–binder formulations with favorable rheological properties, high mechanical strength, strong water resistance, and high thermal stability. Bio-based binders such as cornstarch (Maulana *et al.* 2022) can be combined with reinforcing agents such as microfibrillated cellulose (MFC) (Tayeb *et al.* 2018), potentially yielding composites with strong internal bonding, structural integrity, and moldability (Hossain and Tajvidi 2024). However, the interaction among these components within the context of injection molding has been rarely studied, limiting the upscaling of such materials for industrial use.

Accordingly, this study evaluates the feasibility of using wastepaper (used paper cup)-derived fibers, natural starch binder, and MFC as the key components in injection-moldable bio-composite formulations. To study the feasibility of using this selection of materials in a future developed injection-molded composite, handsheets were prepared from paper cup powder, cornstarch solution, and MFC slurry at various mixing ratios. The physical properties of the handsheets were assessed to identify the promising formulation candidates. These results will inform the design of a pilot-scale fiber-injection-molding system and broaden our understanding of sustainable material designs for biodegradable, high-precision molded products.

EXPERIMENTAL

Materials

The primary raw material, commercially available paper cups, which were coated with polyethylene on the inside, were purchased from a Korean market as shown in Fig. 1. Hardwood bleached kraft pulp (HwBKP) from Moorim P&P (Ulsan, Republic of Korea) was used to prepare MFC in the laboratory to serve as a reinforcing material of the proposed composite and as the primary material in one handsheet. Cornstarch (Moorim Paper Co. Ltd., Jinju, Republic of Korea) functioned as a binder, and a commercial MFC manufactured by Moorim P&P Co. Ltd. (Ulsan, Republic of Korea) was the control to set the specifications of laboratory-prepared CMF. Tables 1 and 2 show the specifications of cornstarch and commercial MFC, respectively.



Fig. 1. Image of the type of paper cup used in this study as a source of material

Table 1. Specifications of Cornstarch Used in this Study

Appearance	Purity	Moisture content	Solubility	Viscosity
White powder	≥ 99% starch	≤13.5%	Insoluble in cold water	Medium to high

Table 2. Specifications of the Commercial MFC Used in This Study

Source material	Diameter	Length	Average particle size
Natural pulp (HwBKP)	10-40 nm	> 1 μm	25.0 μm

Methods

Preparation and characterization of paper-cup powder

The paper cups were manually cut and ground for 20 min using a grinder (SHMF-3000S, Hanil Electric, Bucheon, Republic of Korea). The dry powder samples were collected at 1 min intervals, and their crushed state was visually checked. For an analysis of their fiber characteristics, they were diluted to 0.05%, as specified in the Technical Association of the Pulp and Paper Industry (TAPPI) 271 standard, and their fiber length and width were measured using a fiber length meter (FQA-360, OpTest Equipment Inc., Hawkesbury, Canada). The final powder was selected according to fiber length.

Preparation and characterization of micro-fibrillated cellulose

MFC was prepared by refining and homogenizing HwBKP. Specifically, 1.57% HwBKP was soaked in tap water and refined to 300 ± 5 mL Canadian standard freeness using a laboratory Hollander beater. The refined pulp slurry was diluted to 0.7% consistency and then fibrillated using a high-pressure homogenizer (MN400BF, Micronox, Gwangju, Republic of Korea). The pressure was adjusted to 20,000 psi, the pulp slurry was fed into the homogenizer, and fibrillation was performed three, four, and five times.

The particle size of the 0.2% MFC slurry was measured using a particle analyzer (1090 LD, CILAS, Orléans, France). Particle size measurement based on laser scattering is not a perfect method for detecting fiber dimensions because MFC has a high aspect ratio (Gantenbein *et al.* 2011). However, particle size data indirectly indicates the size differences between MFC types (Park *et al.* 2018). The 1.0% MFC slurry viscosity was determined using a low-shear viscometer (DV-IP, Brookfield Engineering Laboratories, Middleborough, USA) at 25°C. The commercial MFC (control) was in powder form, so its average particle size only needed to be measured.

The fiber width of the prepared MFC was analyzed using field-emission scanning electron microscopy (FE-SEM; JSM-7610F, JEOL, Tokyo, Japan) to ensure that it was fibrillated to the microscale after homogenization. Wet MFC pads were prepared as test specimens, whose fiber widths were measured using a vacuum filtration system. The pads were dried *via* solvent exchange using ethyl alcohol, acetone, and n-hexane to prepare the test specimens. Afterward, FE-SEM images of the pads were captured, and their fiber widths were measured *via* image analysis using a three-dimensional (3D) image software (MP-45030TDI, JEOL, Tokyo, Japan).

Manufacture and measurement of physical properties of handsheets

The ingredients of the injection-molded composite—paper cup powder, cornstarch, and MFC—were used to prepare handsheets. Figure 2 shows the handsheet preparation process and manufacturing conditions. The powder was obtained by grinding used paper cups. A starch solution with a concentration of 10% was prepared by cooking cornstarch powder at 90 to 95 °C for 30 min and then cooling it to room temperature. The final starch solution was diluted to 3% concentration for handsheet preparation.

The paper cup powder and starch solution with a concentration of 3% were initially mixed at the ratios in Table 3 to prepare handsheets and determine the optimal starch content. For an analysis of the strength enhancement provided by MFC, additional handsheets were produced by mixing the powder, starch solution, and MFC slurry according to the formulations in Table 4.

The handsheets were prepared as follows: the powder was dispersed in tap water, the starch solution and MFC slurry were sequentially added to the powder slurry, and this mixture was stirred for 2 min at 600 rpm. Handsheets with a grammage of $60 \pm 5 \text{ g/m}^2$ were then produced following the TAPPI T205 sp-06 (2006) standard. The wet handsheets were pressed at 345 kPa for 5 min using a laboratory wet press (Model 326; Wintree Corporation, Osaka, Japan) and subsequently dried at 120 °C using a cylinder dryer (Daeil Machinery Co. Ltd., Daejeon, Republic of Korea).

The handsheets were conditioned at 23 °C and 50% relative humidity to maintain their moisture content at 8%. The tensile strength (TAPPI T494 om-06, 2006) and burst strength (TAPPI T411 om-10, 2010) of the handsheets were also measured.

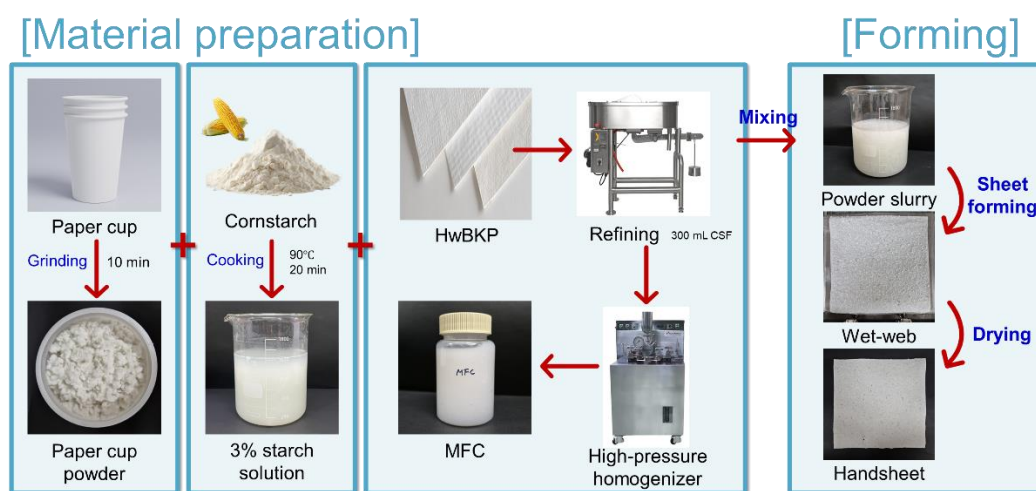


Fig. 2. Flow diagram of handsheet preparation

Table 3. Paper Cup Powder–Starch Mixing Ratios for Handsheet Preparation

Paper Cup Powder (wt %)	Starch (wt %)
50	50
60	40
70	30

Table 4. Paper Cup Powder–Starch–MFC Mixing Ratios for Handsheet Preparation

Paper Cup Powder (wt %)	Starch (wt %)	MFC (wt %)
60	39	1
	37	3
	35	5

RESULTS AND DISCUSSION

Characteristics of the Ground Paper Cup Powder and the Prepared MFC

Figure 3 presents images of powder samples produced after different grinding durations. During the initial 1 to 3 min of grinding, the process was not smooth, and the fiber size was not uniform, hindering the analysis of fiber characteristics. However, as the

grinding duration exceeded 4 min, the process became smoother, the particle size decreased, and fiber size uniformity improved. The fiber analysis results further confirmed these observations.

Figure 4 shows the fiber length and width of the powder produced after different grinding durations. The initial increase in fiber length at the beginning of the grinding process indicated that the grinding was not proceeding smoothly, causing the fibers to entangle and increase the fiber length. The fiber length continued to increase after 4 min of grinding, but it stabilized after 7 min. The average fiber length of the powder ground for 7 min was approximately 0.75 mm. This can be considered relatively short compared to typical virgin and recycled pulps employed in various fiber-based applications such as injection-molded composites and pulp molded packaging. This suggested that pulverized paper cup fibers might still be suitable for use in molded pulp applications, particularly where high strength is not the primary requirement. Therefore, the powder ground for 7 min was used for handsheet preparation.

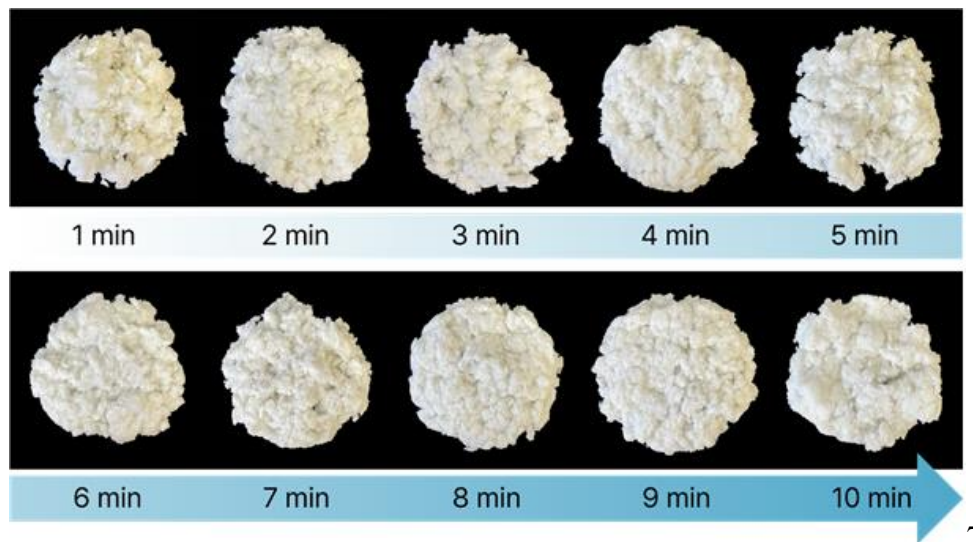


Fig. 3. Images of powder samples for various grinding durations

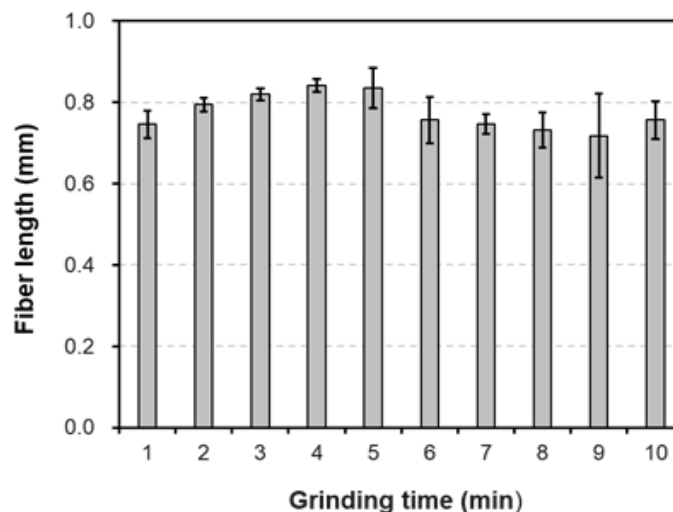


Fig. 4. Powder fiber length depending on the grinding time (Tak *et al.* 2025)

MFC samples were collected for each number of fibrillation rounds during homogenization (three to five). Table 5 summarizes the characteristics of the MFC samples prepared using the high-pressure homogenizer. As the number of fibrillation rounds increased from three to five, the low-shear viscosity increased from 650 cPs to 1,076 cPs, the average particle size decreased from 40.8 to 23.1 μm , and the fiber width decreased from 39.8 to 30.2 nm. These trends were attributed to the homogenizer mechanism: HwBKP fibers were compacted at a high pressure and mechanically broken down at the microscale, which increased their viscosity and decreased their particle size and fiber width.

The average particle size of the commercial MFC, which was in powder form, was also measured to select an appropriate laboratory-prepared MFC for handsheet preparation. Its average particle size was 25.0 μm . Therefore, the MFC sample fibrillated four times, which exhibited a similar particle size, was selected as the reinforcing agent in this study.

Table 5. Characteristics of MFCs Depending on the Fibrillation Number of Homogenization

Fibrillation number	3	4	5
Low-shear viscosity (cPs)	650	883	1,076
Average particle size (μm)	32.8	26.0	21.3
Fiber width (nm)	39.8	36.0	30.2

Measurement of Physical Properties of Handsheets

Handsheets were made using powder and starch at mixing ratios of 50:50, 60:40, and 70:30, and the changes in their strength depending on the mixing ratio were investigated. Figure 5 shows a comparison between a handsheet made from powder and starch and one made solely from HwBKP. The formation of handsheets using only starch (without the complete disintegration of the paper cups) indicated the feasibility of fabricating composites via injection molding. However, compared with the handsheet composed exclusively of HwBKP, the handsheet derived from the paper cups exhibited noticeably lower uniformity, as visually observed.

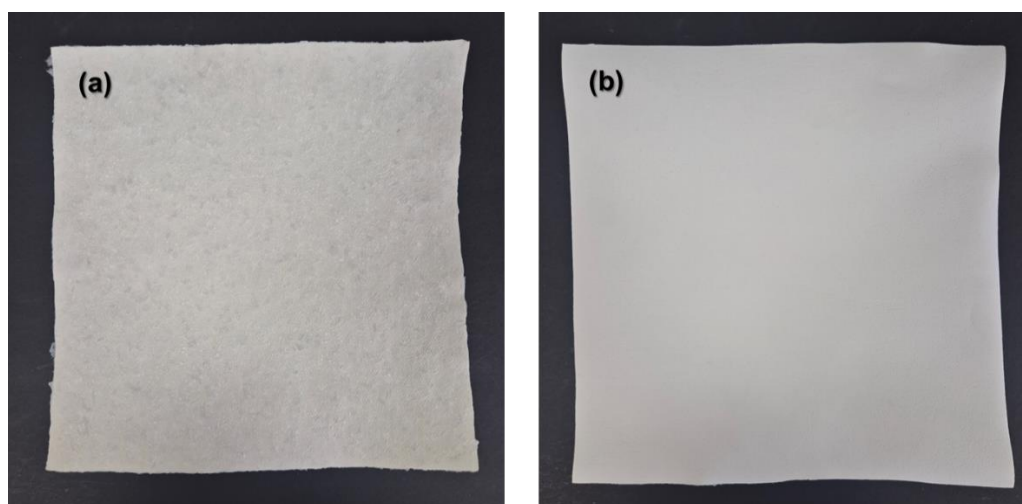


Fig. 5. (a) Handsheet made from paper cup powder and starch for modeling of injection-molded composite and (b) handsheet made from HwBKP

The tensile strength and elongation at break of the handsheets are presented in Figs. 6 and 7, respectively. As the powder content decreased and the starch content increased, the handsheet tensile strength and elongation tended to decrease. This was because paper-cup fibers, which are primarily composed of cellulose, have a strong network structure that provides mechanical strength and flexibility. As the fiber amount decreased, this reinforcing network weakened, lowering the tensile strength. Although starch acted as a binder, it is generally more brittle and less flexible than cellulose fibers. The increased starch content introduced rigid, brittle regions into the handsheet, reducing its ability to stretch (lower elongation) and resist fracture (lower tensile strength). The burst strength of the handsheets showed a similar trend to that of their tensile strength (Fig. 8). Moreover, as the starch content increased, the material started adhering to the equipment, such as dryer rolls, suggesting that a higher starch content may compromise workability in manufacturing. Therefore, the 60:40 ratio, which did not exhibit such issues, was considered the most appropriate condition for the next experiment.

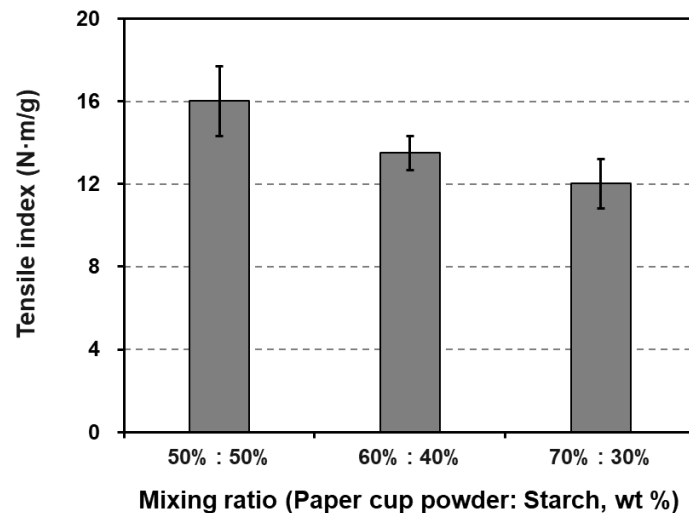


Fig. 6. Tensile index of handsheets depending on paper cup-starch mixing ratio

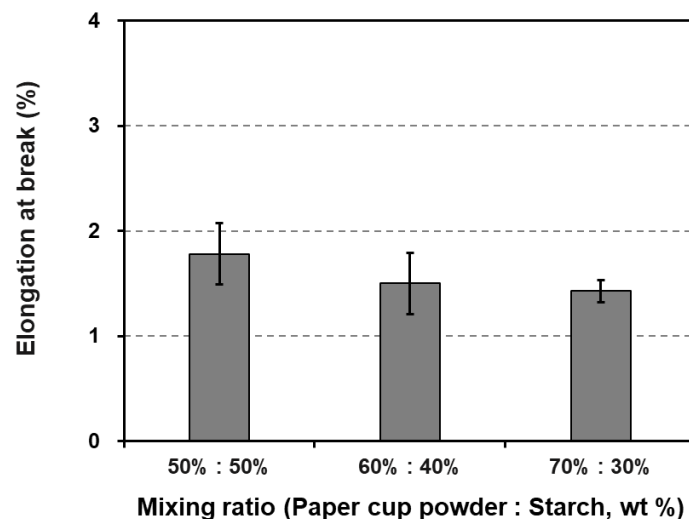


Fig. 7. Elongation at break of handsheets depending paper cup-starch mixing ratio

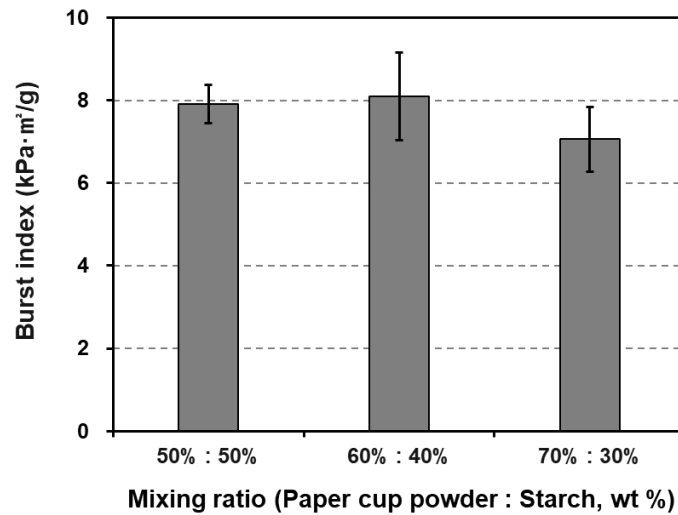


Fig. 8. Burst index of handsheets depending on paper cup-starch mixing ratio

Figures 9, 10 and 11 show the tensile strength, elongation at break, and burst strength, respectively, of the powder–starch–MFC handsheet. As the starch content decreased and the MFC content increased, the tensile strength, elongation at break, and burst strength linearly increased. The improvement in tensile and burst strengths was attributed to the enhanced fiber–fiber bonding facilitated by the addition of MFC. Furthermore, the reduction in starch content, which typically increased the brittleness and decreased the flexibility of the handsheets, may have helped enhance elongation. However, when the MFC content exceeded 5%, the dewatering time during wet-sheet formation was prolonged to over 20 min. Therefore, considering both mechanical strength and dewatering efficiency, the MFC content should be maintained at or below 5%.

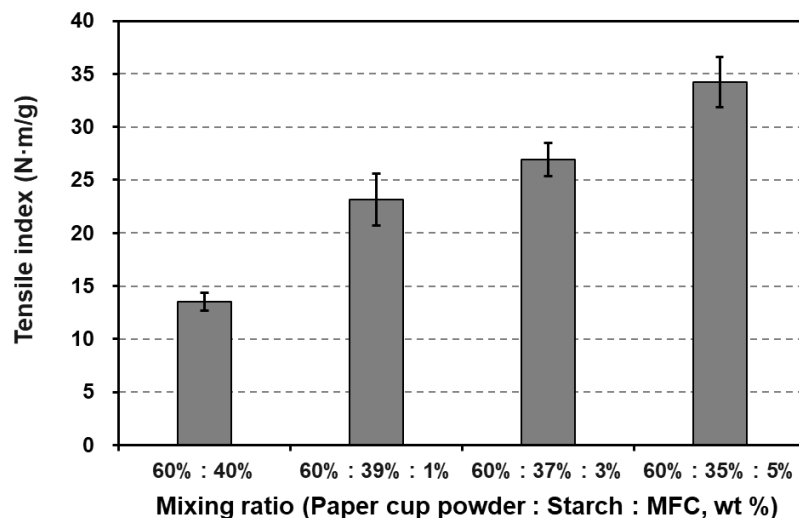


Fig. 9. Tensile index of the sheets depending on paper cup powder-starch-MFC mixing ratio

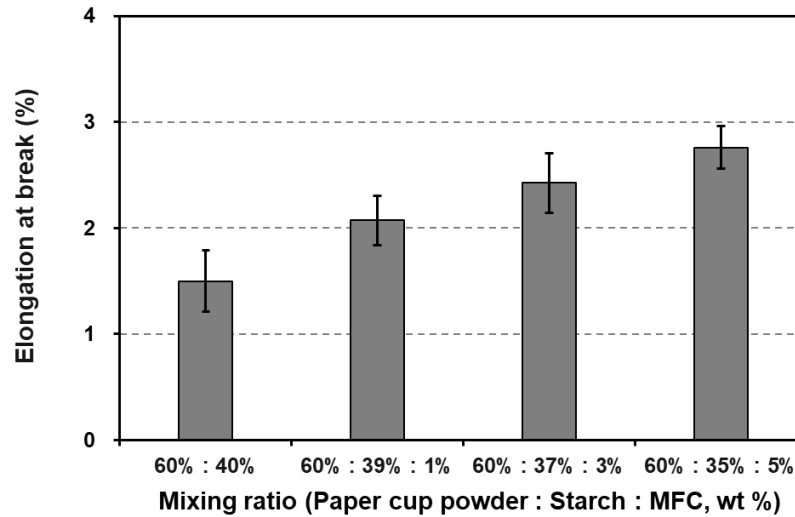


Fig. 10. Elongation at break of the sheets depending on paper cup powder-starch-MFC mixing ratio

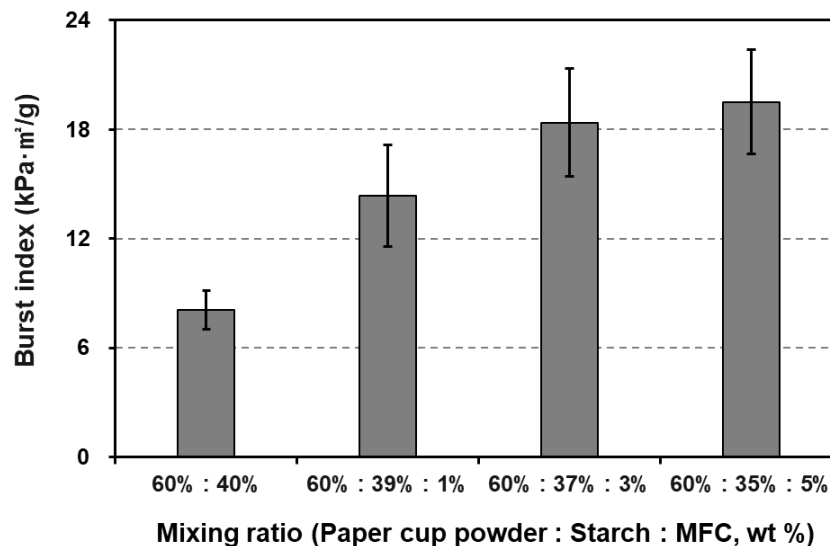


Fig. 11. Burst index of the sheets depending on paper cup powder-starch-MFC mixing ratio

CONCLUSIONS

1. This study demonstrated the feasibility of recycling used paper cups into eco-friendly injection-molded composites by incorporating starch and microfibrillated cellulose (MFC). Optimal powder preparation was achieved by grinding paper cups for 7 min, producing a uniform particle size distribution suitable for composite manufacturing. The particle size of the laboratory-prepared MFC, processed through four passes in a high-pressure homogenizer, was similar to that of a commercial MFC product.

2. A 60:40 paper cup powder–starch mixing ratio provided the best mechanical properties, balancing tensile strength, elongation at break, and burst strength, while avoiding workability issues, such as adhesion to equipment parts during drying. Increasing the starch content beyond 40% compromised the mechanical properties and workability of the material.
3. The addition of MFC improved the tensile strength, elongation at break, and burst strength linearly up to an MFC content of 5%. However, beyond this percentage, the dewatering time during handsheet formation exceeded 20 min, indicating a processability constraint.
4. In conclusion, combining paper-cup powder, starch, and up to 5% MFC is an effective strategy for producing injection-moldable composites with enhanced mechanical properties.

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