Changes in Fold Cracking Properties and Mechanical Properties of High-Grammage Paper as Affected by Additive and Fillers

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Fold cracking, which reduces the economic feasibility of paper-making, is a localized surface deformation caused by extreme bending stress. Most paper products, such as base paper and coated paper, generate fold cracking during folding processing. To control fold cracking, the mechanical properties of the base paper can be strengthened, and the flexibility of the structure can be increased by controlling the modification in pulp fibers and stock preparation conditions. This study analyzed the changes in the mechanical properties of high-grammage paper in response to the addition of precipitated calcium carbonate (PCC) and cationic starch (C-starch). The application of inorganic filler (PCC) drastically reduced the internal bond strength and tensile strength, causing fold cracking, whereas C-starch increased the bond strength between fibers, which improved the tensile strength, internal bond strength, and elongation. However, when applied independently, fold cracking occurred because of extreme increase or decrease in strength. Therefore, the combined application of C-starch and PCC made it possible to form a paper-based structure with high fold cracking resistance. Moreover, when the fold cracking resistance was excellent, the mechanical properties were balanced without being biased in one direction even under conditions of relatively low mechanical properties.

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INTRODUCTION

High-bulk and high-grammage papers are widely used for packaging and printing purposes because of their suitability in packaging and their unique texture, respectively. Folding processing is performed to produce packaging boxes from paper. Fold cracking, which is caused by high mechanical pressure, limits the economic feasibility of the paper-making industry (Kainulainen 2013).

Folding is an extreme bending mechanism that causes localized surface deformation. Depending on the direction of the fold, the stresses generated during folding are: (1) extensional stress (outside) and (2) compressive stress (inside). Collapse of the paper structure caused by external elongation stress is the main defect of fold cracking. Efforts have been made to find the cause of fold cracking and improve it. Both sides of base paper and coated paper are being investigated, and fold cracking has been found on all sides (Oh *et al*. 2013; Pál *et al*. 2013).

Fold cracking can be made more severe by increasing the amount of inorganic mineral particle filler in the coating solution or by using brittle starch binders (Breskvar *et al*. 2021). To improve the fold properties in various types of coating structures, the flexibility and mechanical properties of the coating layer are improved by applying nanomaterials and various biopolymers (*e.g*., polysaccharide, protein, and polyester) (Rastogi and Samyn 2015; Du *et al*. 2016; Sharma *et al*. 2020; Zhu *et al*. 2021). With regard to base paper, fold cracking can be controlled by improving the mechanical properties and increasing the flexibility of the structure by controlling the modification of pulp fibers, which are natural products, and the conditions of stock preparation. Improving the folding properties of base paper through *via* stock preparation is the most important method, because controlling of the properties of pulp fibers has clear limitations. Generally, physical properties and moisture conditions of low-quality base paper affect the fold cracking in high-grammage paper (Carbone 1999). However, the raw material composition, such as the amount of addition of wet strength agents and inorganic fillers, have a significant influence (Alam and Toivakka 2011; Sim *et al*. 2012). Jopson (1992) reported that the separation between fibers due to compressive stress during folding was considered delamination, and that the elongational stress applied to the outer surface was released by this delamination, thereby improving fold cracking. Delamination, fiber orientation, and thickness significantly influence the folding properties (Barbier *et al*. 2002, 2003). Folding cracks can be eliminated by reducing the thickness. However, in practice, the thickness is related to the structural characteristics of high-quality packaging paper. For this reason, reducing the thickness at the same basis weight reduces the quality of product usage. Therefore, research is required to improve the folding properties while maintaining high bulk properties.

To manufacture high-grammage paper of 150 g/m^2 or more, this study analyzed the effect of applying the conditions of additives (precipitated calcium carbonate (PCC) and starch) on the mechanical properties of paper in the stock preparation and explored various methods to improve fold cracking while maintaining high bulk characteristics.

EXPERIMENTAL

Materials

Handsheets were prepared using hardwood pulp and softwood pulp. Bleached kraft pulp of eucalyptus species was used as hardwood pulp, and bleached kraft pulp of mixed tree species (lodgepole pine, white spruce, sub-alpine fir) was used as softwood pulp. To control the mechanical properties of hand sheets, cationic starch (C-starch) and precipitated calcium carbonate (PCC), which are additives that affect physical properties, were used in sample preparation.

High-grammage Paper Forming

Hardwood and softwood pulp were either used individually or blended in a 9:1 ratio and beaten at 30 ºSR using a laboratory Hollander beater. After the beating process, 10% to 20% and 0.5% to 2.0% of PCC and C-starch were applied to the stock, respectively, and sufficiently dispersed at 400 rpm (Tables 1 and 2). The optimal conditions were collected, and samples were prepared based on the results of independently applying the influencing factors (Table 2). Each sample was prepared with a base weight of 200±10 g/m^2 and conditioned for 24 h under the conditions of 20 ± 5 °C and 50±2% relative humidity.

Table 1. High-grammage Paper Sheet-Making Conditions with the Addition of C-starch and PCC

Symbol	Pulp Fiber Mixing Ratio		C-starch	PCC
	Hw pulp	Sw pulp	%	%
Control-Hw	100	-	-	-
Hw-Starch-0.5	100	-	0.5	-
Hw-Starch-1.0	100	-	1.0	-
Hw-Starch-1.5	100	-	1.5	-
Sw-Starch-0.5	-	100	0.5	-
Sw-Starch-1.0	-	100	1.0	-
Sw-Starch-1.5	-	100	1.5	-
Hw-Ash-10	100	-	-	10
Hw-Ash-20	100	-	-	20
Sw-Ash-10	-	100	-	10
Sw-Ash-20	-	100	-	20

Table 2. High-grammage Paper Sheet-Making Conditions with Complex Application of Additives in Stock Preparation

Symbol	Beating Degree	Pulp Fiber Mixing Ratio		PCC	Cationic Starch
	ºSR	Hw pulp	Sw pulp	%	%
10-0	30	90	10	10	-
10-0.5					0.5
10-1.0					1.0
10-1.5					1.5
10-2.0					2.0
20-0				20	-
20-0.5					0.5
20-1.0					1.0
20-1.5					1.5
20-2.0					2.0

Evaluation of High-grammage Paper Properties and Rate of Change

The properties of the high-grammage paper under each condition were evaluated. To calculate the density, the thickness was measured using a micrometer (L&W micrometer/ABB/Swiss). The tensile strength and elongation were measured using an L&W tensile tester. Internal bond strength was measured using a Scott bond tester/ Zwick Roell; TAPPI T 541 om-21(2021)). For a better visual evaluation of the correlation between the mechanical properties and folding properties, each value was calculated as the rate of change within the range of maximum and minimum values and comparative analysis was performed. The rate of change was calculated as follows,

$$R = \frac{D - V_{min}}{V_{max} - V_{min}} \times 100 \tag{1}$$

where R is the rate of change, D is the data value (density, tensile strength, internal bond strength, elongation), $V_{\min}$ is the minimum value of total data, and $V_{\max}$ is the maximum value of total data.

To establish the V_{min} and V_{max} values, handsheets with a basis weight of $200 g/m^2$ were prepared by mixing hardwood and softwood pulp (9:1, 8:2, 7:3), beating degree control (20 – 40 °SR) and applying PCC (10, 20%) and C-starch (0 – 1.5%), and the mechanical properties values were collected. The V_{min} and V_{max} values and the corresponding hand-sheet manufacturing conditions are shown in Table 3.

Table 3. Experimental Ranges for V_{min} and V_{max} Values

	V_{min}	V_{max}	Handsheet Making Conditions	
Density (g/m^2)	0.43	0.78	V_{min}	Hw pulp 20°SR
			V_{max}	Sw pulp 40°SR
Tensile strength (kN/m)	18.31	159.27	V_{min}	Hw pulp 20°SR
			V_{max}	Sw pulp 30°SR
Internal bond strength (ft-Lb 1000)	0	346	V_{min}	Hw pulp 20°SR
			V_{max}	Sw pulp 30°SR
Elongation (mm)	0.55	3.32	V_{min}	Hw pulp 20°SR
			V_{max}	Hw pulp 40°SR

Evaluation of Fold Cracking Resistance

The evaluation of fold cracking resistance has been conducted utilizing various methods, such as image analysis or visual analysis methods, which assess the deformation of the paper-based structure (Oh *et al*. 2015). The quality criterion in the industrial field for evaluating fold cracking is based on the occurrence of the phenomenon, rather than the extent or magnitude of the damage caused by fold cracking. Thus, it is necessary to check for fold cracking under extreme conditions, such as a dry environment or severe folding processing, and to explore ways to improve it.

In this study, the resistance to fold cracking was evaluated by conducting folding tests under extreme processing conditions. The hand-sheet was adjusted to a low moisture content, and a rigorous folding process was then applied, including high pressure and speed, linear pressure, and large folding angle. The moisture content of the handsheet was reduced to 2% or lower by drying it in an oven dryer at 135 °C for 2 minutes. The folding process was performed using a laboratory rubber roll press, with a folding angle of 360 ° and a pressure of 1.96 MPa applied at a speed of 20 m/min. Thereafter, the folded lines of the handsheet subjected to folding stress were visually observed, and the occurrence of fold cracking was classified into “normal” and “crack” (Fig. 1).

The handsheets were evaluated by folding process them a total of 10 times in different locations. All of the handsheets were classified as “crack” even if only minimal fold cracking was observed.

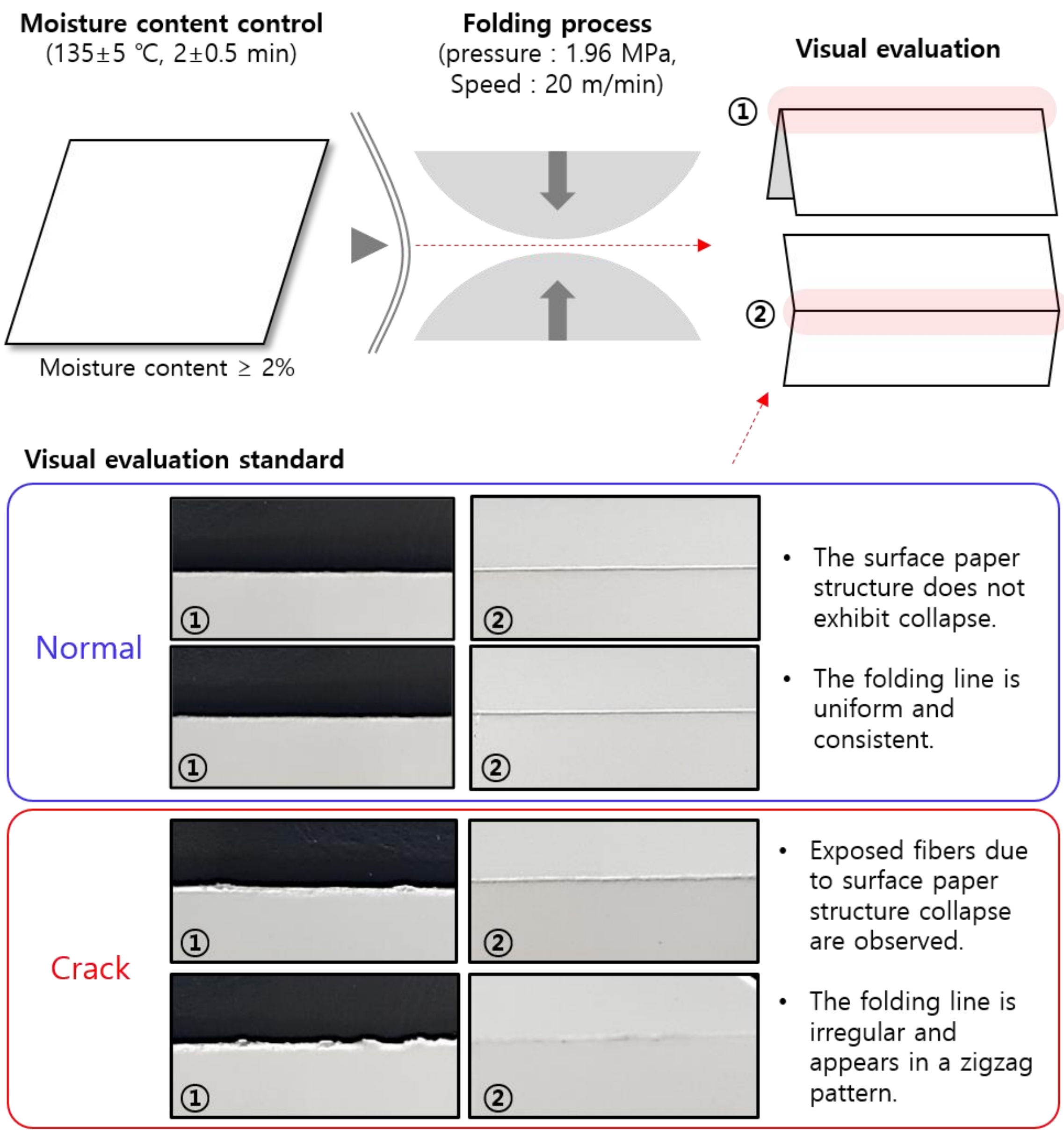

Fig. 1. Visual evaluation method of fold cracking

RESULTS AND DISCUSSION

Effect of PCC Addition on Mechanical and Folding Properties

When more than 10% of PCC was added, fold cracking occurred in the high-grammage paper composed of hardwood or softwood pulp fibers (Fig. 2). The addition of PCC reduced the internal bond strength and tensile strength, but it did not significantly impact density. The elongation increased when 10% PCC was added, but it decreased when 20% PCC was added. Despite the expectation that the addition of PCC would enhance the fold cracking resistance by alleviating the elongational and compressive stress on the folding surface caused by internal collapse due to delamination (Jopson 1992), the results were contradictory due to the extreme decrease in internal bond strength and tensile strength. The decrease in mechanical properties led to fold cracking, even at relatively small thicknesses. The application of inorganic filler to improve production cost can lead to an extreme decrease in mechanical properties, resulting in fold cracking.

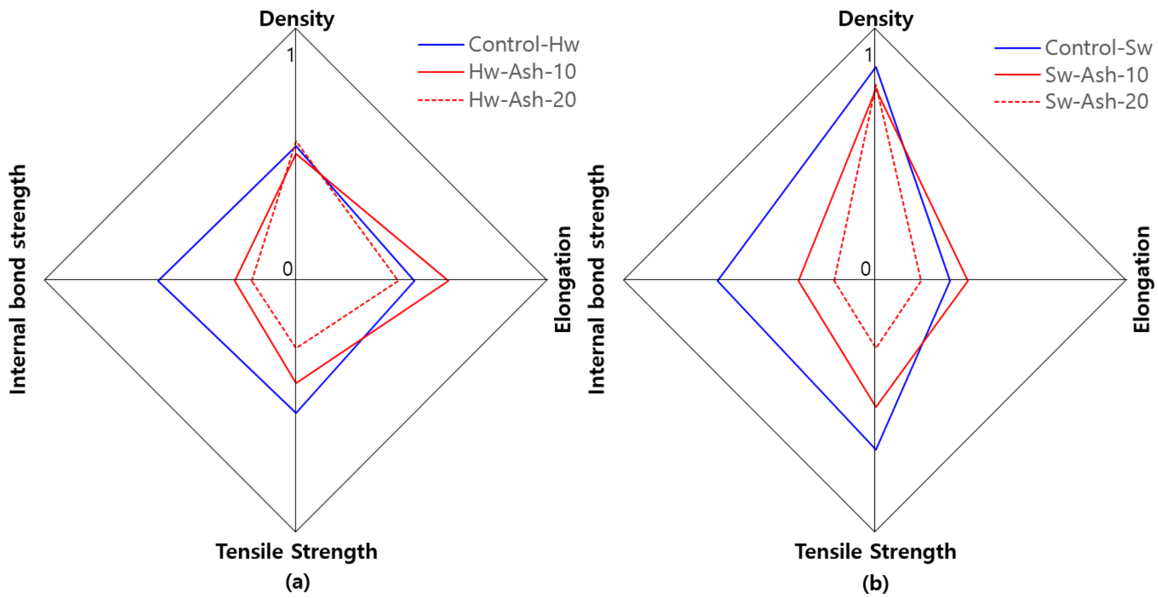

Fig. 2. Rate of change in the mechanical properties and fold cracking occurrence of high-grammage paper according to the PCC addition (a : Hardwood pulp, b : Softwood pulp)

Effects of C-starch Addition on Mechanical and Folding Properties

The mechanical properties and fold cracking according to the amount of addition of cationic starch are shown in Fig. 3. A tendency opposite to that associated with PCC application is evident. In the case of hardwood pulp, fold cracking did not occur under all the conditions (Control-Hw, Hw-Starch-0.5, Hw-Starch-1.0 and Hw-Starch-1.5), and the tensile strength and elongation increased significantly with the increasing amount of C-starch; furthermore, the internal bond strength increased with the addition of 1.5% and tensile strength and internal bond strength increased significantly in the case of softwood pulp.

The elongation increased with the addition of 0.5% C-starch, and the rate of increase decreased with further addition. Fold cracking occurred under the Sw-Starch-1.5 condition, wherein the internal bond strength and tensile strength increased significantly. This difference is attributable to the fact that softwood pulp, which has a large bonding area between fibers, is more significantly affected by C-starch, a wet strength enhancer, and exhibits relatively high tensile strength, internal bond strength, and low elongation. These results are the same as those reported in a prior study that revealed that high mechanical properties adversely affect fold cracking (Sim *et al*. 2012).

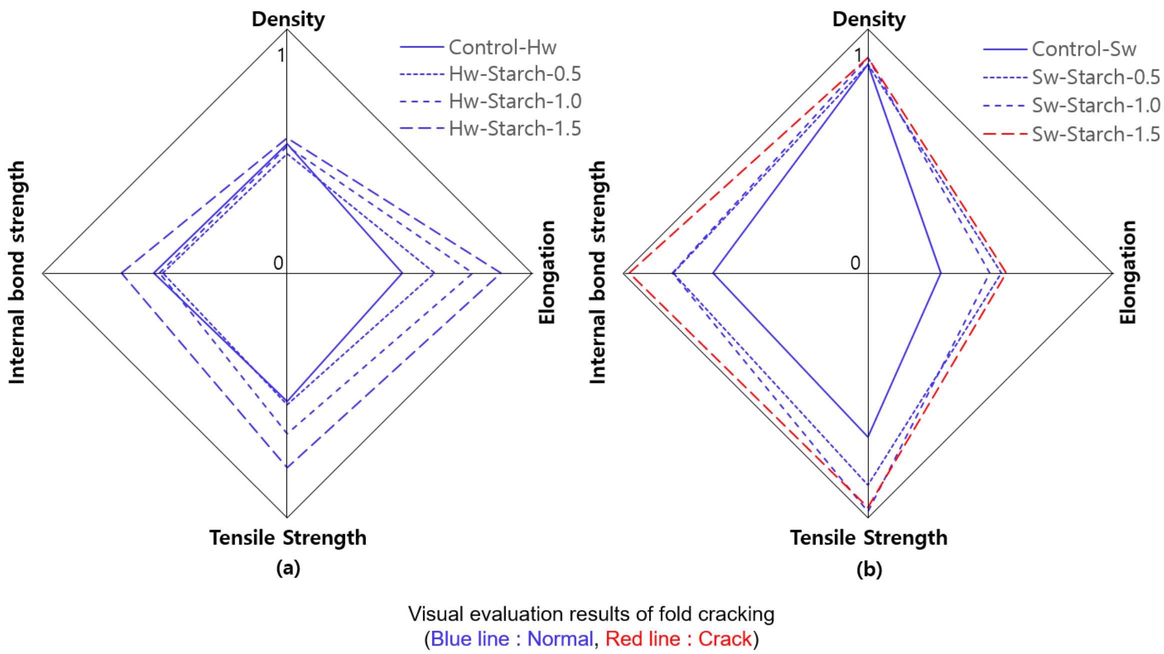

Fig. 3. Rate of change in mechanical properties and fold cracking occurrence of high-grammage paper according to the addition of C-starch (a : Hardwood pulp, b : Softwood pulp)

Evaluation of Mechanical Properties and Folding Properties with Complex Application of Additives in Stock Preparation

Based on the previous results from application of hardwood, softwood pulp, inorganic filler, and C-starch, the mechanical properties and fold cracking of samples manufactured by applying them in combination are shown in Fig. 4. At a beating degree of 30 ºSR, both hardwood and softwood pulp fibers had excellent fold cracking resistance. However, when the fiber length of the pulp fibers constituting the paper was diversely composed, it was judged to be advantageous in the folding process subjected to various stresses, so the two pulp fibers were mixed and used. In previous studies, the addition of PPC was found to negatively affect fold cracking resistance. To address both economic feasibility and fold cracking, PCC and C-starch were applied in combination. Hardwood pulp and softwood pulp were mixed in a ratio of 9:1 to achieve a beating degree of 30 ºSR, and the mechanical properties and fold cracking resistance were evaluated by adjusting the amount of added PCC and C-starch. Fold cracking does not occur when more than 1.0% of C-starch was applied under the condition of addition of 10% of PCC (Fig. 4A). The tensile strength and internal bond strength increased and elongation decreased, which is the result of the increase in the fold cracking resistance through the improvement of appropriate strength at the same density properties. Different types of changes were observed under the condition of adding 20% PCC (Fig. 4B). Similar trends were observed in the change rates of internal bond strength and tensile strength; however, the elongation tended to increase with the addition of C-starch. These results showed a similar pattern to the mechanical properties of the low-density paper in the study by Park *et al* (2020). Compared to the condition of adding 10% PCC, the PCC filled in the pores between the fibers weakened the internal fiber bonding and exhibited lower mechanical properties. As a result, it became easier to break the paper structure by tensile stress due to folding process, and the form of breaking was the type that occurred when the mechanical properties were lower than the strength of the fiber itself. The paper had high elongation properties, resulting in simultaneous improvement of the conflicting mechanical properties.

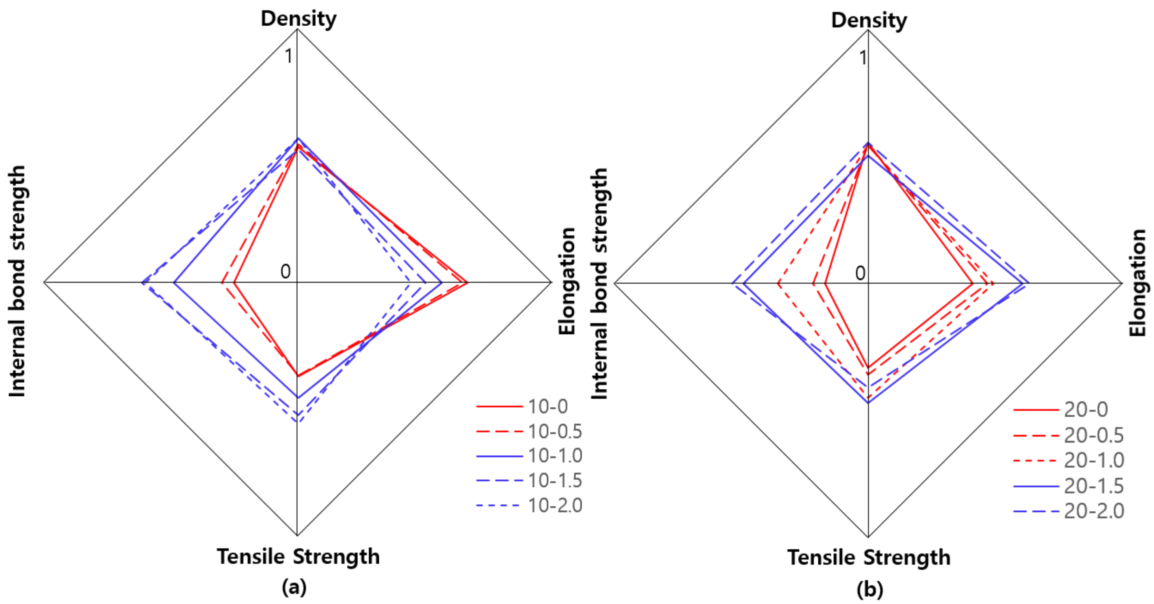

Fig. 4. Rate of change in mechanical properties and fold cracking occurrence of high-grammage paper according to the complex addition of PCC and C- starch (a : 10% PCC, b : 20% PCC)

The mechanical properties and fold cracking resistance values of all the handsheets were comprehensively analyzed (Fig. 5). The range of density for samples categorized as ‘Normal’ and ‘Crack’ was quite similar, and even if the paper's thickness decreased, the possibility of fold cracking still existed and depended on factors such as tensile strength, internal bond strength, and elongation. The tensile strength was fold cracked to be in the range of 5.84 to 9.27 kgf/15 mm or 16.01 kgf/15 mm, with the best results found in the range of 7.89 to 16.24 kgf/15 mm. The elongation was fold cracked to be in the range of 1.06 to 2.27 mm, with the best results seen at 1.39 to 3.05 mm. Tensile strength and elongation play a role in improving folding, but they are not the only factors affecting folding as they are impacted by other mechanical properties as well. The most significant mechanical property was found to be internal bond strength. The range of internal bond strength for the handsheets divided into ‘Crack’ and ‘Normal’ categories was distinct. Internal bond strength is a crucial mechanical property that determines the paper structure's flexibility or stiffness, and has a significant impact on fold cracking resistance.

Based on these results, several types of fold cracking can be identified. Firstly, there are samples with low mechanical properties (*e.g.*, Sw-Ash-10, 20). Secondly, there is a type of sample with high elongation (more than 1.5 mm), which has a flexible structure, but it is unable to endure changes in paper structure due to low tensile strength (less than 9 kgf/15 mm) and low internal bond strength (126 FT-LB 1000). Finally, samples with high mechanical properties such as high internal bond strength (346 FT-LB 1000), high tensile strength (16.01 kgf/15 mm), and high elongation (2.15 mm), but with a rigid and inflexible paper structure, may still result in fold cracking due to the lack of flexibility.

The internal bond strength and tensile strength reduced by the addition of PCCs were improved by the addition of C-Starch, resulting in improved fold cracking. Although the results of this study were limited to conditions of basis weight 200 g/m^2 and beating degree 30°SR, they can still serve as a useful reference when selecting additives (PCC and C-Starch) to improve fold cracking.

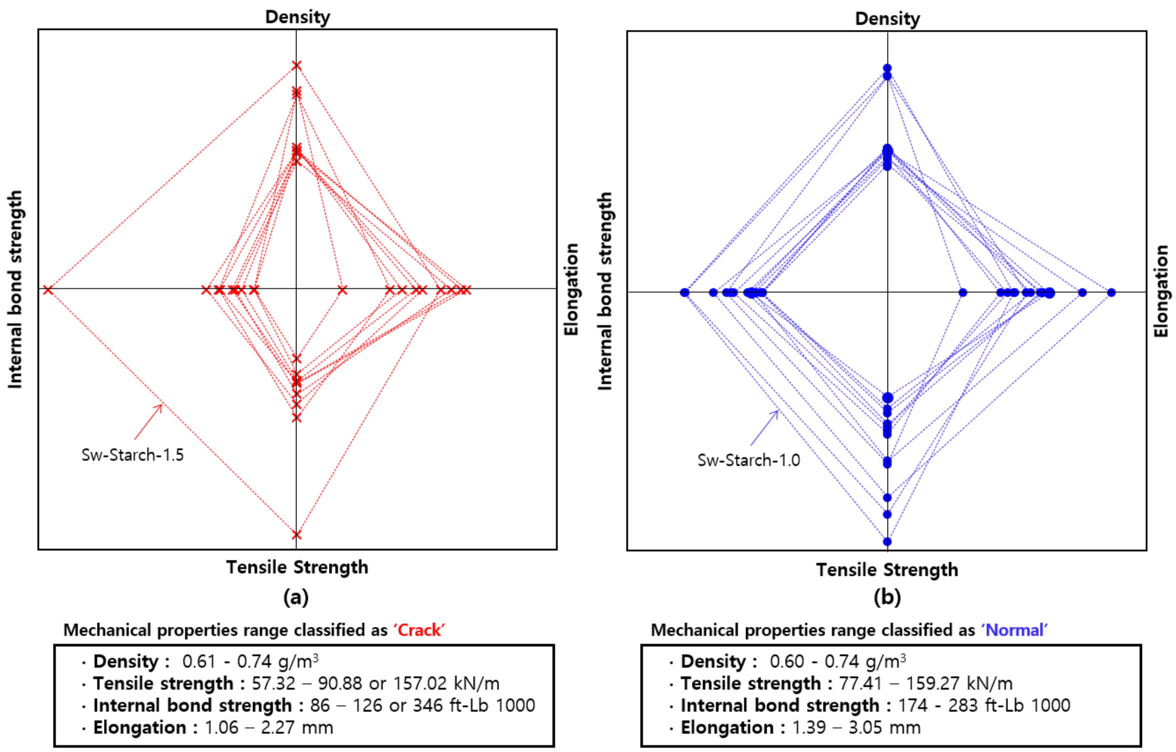

Fig. 5. Rate of change in the mechanical properties and fold cracking resistance properties of high-grammage paper (a : Crack, b : Normal)

CONCLUSIONS

1. Because the application of precipitated calcium carbonate (PCC) independently has been shown to drastically reduce the internal bond strength and tensile strength and cause fold cracking, a means to supplement the mechanical properties while increasing the economic efficiency of the product must be applied. The addition of cationic starch (C-starch) was found to improve the tensile strength, internal bond strength, and elongation by increasing the bonding strength between fibers; however, the results showed that excessive input caused a decrease in the flexibility of the paper structure, led to the occurrence of fold cracking.
2. The combined application of C-starch and PCC made it possible to form a paper-based structure with high fold cracking resistance. The tensile strength and internal bond strength increased, resulting in an increase in physical strength, and an increase in elongation, simultaneously so that the paper structure is easily deformed by folding stress, and the overall fold cracking resistance is improved.

3. Fold cracking occurred in papers with extremely low or high mechanical properties, and even when the mechanical properties were relatively low, fold cracking resistance was excellent when balanced without being biased in one direction.
4. The addition of PCC is not expected to improve fold cracking but rather to only improve the economics of the paper production process. However, the present work has confirmed that the addition of PCC can improve fold cracking while maintaining the product quality and reducing the amount of pulp used.

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