Influence of Fiber Composition and Drying Conditions on the Bending Stiffness of Paper

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Changes in thickness, elastic modulus, and bending stiffness were studied for handsheets prepared using different fiber compositions and dried under restraint or unrestraint conditions, when exposed to various humidity conditions. Four sets of experimental studies were carried out to investigate the effect of (1) different amounts of fines (or long fibers), (2) two-ply sheet forming, (3) high temperature restraint press drying, and (4) the use of recycled fibers on the thickness, elastic modulus, and bending stiffness. The results showed that thickness, elastic modulus, and bending stiffness changed depending upon the fiber composition, single or multi-ply forming, drying conditions, and recycling of fibers. Thickness change, restraint drying, and fiber hornification during recycling were the principal factors affecting the bending stiffness in cyclic humidity conditions.

Keywords: Bending stiffness; Fines content; Condebelt drying; Humidity; Thickness

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INTRODUCTION

Paper and paperboard have important roles in the packaging industry. Their advantages include abundant availability, renewability, good biodegradability and recyclability, flexibility, and ease of functionalization (Isikgor and Becer 2015; Herrera *et al.* 2017). Furthermore, cellulosic paper is safe to use and inexpensive, making it competitive for many packaging applications. Although several new applications such as polymer-fiber composites, printed electronics, and others have been suggested (Lindner 2018), packaging is still the most important and principal application for paper and paperboard. However, the hygroscopic nature of cellulosic fiber hinders the application of paper in humid conditions (Alava and Niskanen 2006).

Paper is highly hygroscopic because it is made from fibers containing hydrophilic materials such as cellulose and hemicellulose. Therefore, paper is affected greatly by changes in environmental conditions that include relative humidity and temperature. The change in relative humidity results in adsorption or desorption of moisture by papermaking fibers, which results in a substantial dimensional change of paper thickness and many other properties.

In general, strength properties are critically important properties for paperboards because they are mainly used in packaging. The compressive failure of corrugated boxes depends on changes of relative humidity (Navaranjan *et al.* 2013). It is required, therefore, to control the relative humidity in the warehouse to reduce the compressive buckling collapse of the corrugated boxes. In many cases, packaging boxes are made to have excessive strength to allow them survive in cyclic humidity environments. This is because the mechanical response of paper to outside forces in a wide range of relative humidity climates is quite different from that in the constant relative humidity environment.

Therefore, it is crucial to understand and anticipate the mechanical response of paper or paperboard to moisture change. The response of paper in variable relative humidity conditions has been explored experimentally by several researchers (Byrd 1972a,b; Fellers and Bränge 1985; Salmén 1986; Ganser *et al.* 2015). Jajcinovic *et al.* (2018) have shown that swelling or shrinkage influences the mechanical properties of pulp fibers and the fiber-to-fiber joint strength.

Stiffness is one of the next most important properties for paperboard for packaging; without stiffness, proper physical protection of packaged goods cannot be achieved. Bending stiffness is a critical parameter for the rigidity and strength of paperboard. Unlike the in-plane tensile properties, the bending stiffness depends considerably on the macroscopic thickness and layered structure of paper or paperboard (Carlsson 1986; Carson and Popil 2008). According to the fundamental mechanics, the bending stiffness of a material is proportional to the third power of the thickness and elastic modulus. Thus, bending stiffness is sensitive to the change of elastic modulus and sheet thickness. This also suggests that strategic layer structure design with different furnishes may be employed to maximize bending stiffness at minimum basis weight.

This study investigated the changes of thickness, elastic modulus, and bending stiffness of handsheets prepared using different fiber compositions and dried under restraint or unrestrained conditions, when exposed to various humidity conditions. Four sets of experimental studies were carried out to investigate the effect of (1) different amount of fines (or long fibers), (2) multi-ply sheet forming, (3) high temperature press drying, and (4) use of recycled fibers on the change of thickness, elastic modulus, and bending stiffness. The relationship among the three factors and the reversibility of the factors in a cyclic humidity environment was examined.

EXPERIMENTAL

Materials

Figure 1 shows the procedures to prepare the long-fiber fraction and fines fraction of softwood unbleached kraft pulp (UKP) stock. To produce a long-fiber fraction, the stock was first beaten to 450 mL CSF using a laboratory Valley beater. The fines content of the stock was *ca*. 14.3%. The Kajaani FiberLab™ (Kajaani, Finland) was used for analyzing fiber length. The average fiber length (length weighted) was 2.30 mm. The paper made with the long fiber stock often showed uneven mechanical properties because of the non-uniform formation of paper. To improve the formation of paper, fractionation was carried out using a Sweco Vibro-Energy® separator (SWECO, Florence, KY, USA). The fraction, which passed through a 25-mesh wire and retained on a 50-mesh wire (P25/R50), was selected as the long fiber fraction. The average fiber length of this fraction was 1.82 mm. The fines fraction was obtained after beating the original UKP stock until the average fiber length decreased to 0.29 mm. Figure 2 shows the length weighted distribution of fiber length for the long fiber fraction and fines fraction.

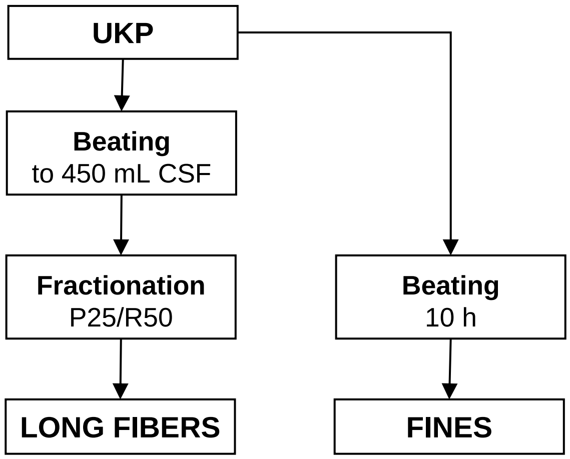


Fig. 1. Procedures for the preparation of the long fiber and fines fraction using softwood UKP stock

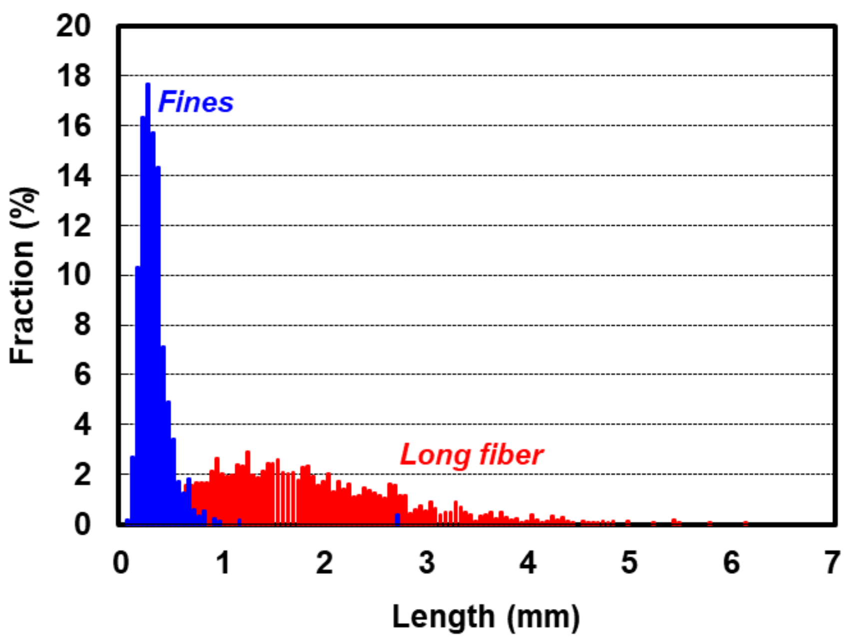


Fig. 2. Fiber length weighted distribution of the long fiber fraction and fines fraction

When OCC recycled in Korea (KOCC) was used as a raw material, slightly different procedures were employed to produce the long fiber and fines fractions. As shown in Fig. 3, KOCC was first disintegrated at 2% consistency using a low consistency disintegrator for 30,000 rev. The freeness of the stock was 400 mL CSF. The long fiber fraction was collected by fractionating the disintegrated KOCC slurry with 25- and 50-mesh wires using a Sweco separator (SWECO, Florence, KY, USA). The average fiber length of this fraction was 1.18 mm. The fines fraction was prepared after beating the fractionated KOCC slurry (P25/R50) for 3 hrs. The average fiber length of fines fraction was 0.24 mm.

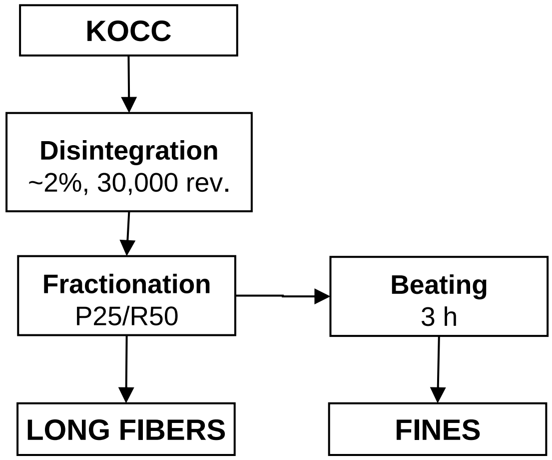


Fig. 3. Procedures for the preparation of the long fibers and the fines fraction using KOCC slurry

Handsheet Forming and Drying

Using the long fiber and fines fraction prepared, various handsheets were formed according to TAPPI T205 sp-02 (2002). Single-ply handsheets containing different amounts of fines were made using a laboratory sheet mold. The target oven dry weight of handsheets produced was 100 g/m^2. A 200-mesh wire was used as a sheet forming medium for the complete retention of the long fiber fraction. To form handsheets with controlled amounts of fines, the amount of fines fraction was varied depending on the target content of fines in the sheet. For example, to form a handsheet containing 20% of fines, a long fiber stock that gives 80 g/m^2 was prepared, and a proper amount of fines was added to get 100 g/m^2. Because a 200-mesh wire was used, complete retention of long fibers was assumed, and the proportion of fines in the handsheet was obtained from the dry weight of handsheet. The wet sheet was pressed and dried completely at 120 °C using a laboratory cylinder dryer.

Two-ply handsheets containing different amounts of fines in each ply were produced. The fines content in each ply was adjusted as described earlier. The oven-dry weights of the top and bottom plies were 50 g/m^2. After combining two wet plies, it was pressed and then dried completely at 120 °C using a laboratory cylinder dryer.

When handsheets containing 15% or more of fines were dried on a laboratory cylinder dryer, severe in-plane shrinkage occurred. For example, while the drying shrinkage of cylinder-dried single-ply UKP handsheet with 0% of fines was *ca*. 0.8%. It increased to 2.8% when the amount of fines was 20%. To restrain the drying shrinkage of handsheets, the Condebelt drying method was employed (Lehtinen 1995; Niskanen and Kärenlampi 1998), which decreased the drying shrinkage of single-ply UKP handsheet with 20% of fines to 0.8%.

Conditioning under Different Humidity Conditions

The handsheets were cut into a size of 38 ± 1 mm wide and 70 ± 1 mm long for physical testing, and preconditioned to reach an equilibrium at 51 ± 2% relative humidity (RH) and 23 ± 1 °C for at least 48 h. Then, they were exposed to different humidity conditions of 74, 94, 51, and 32% RH, as shown in Fig. 4. The moisture content of the samples at each humidity condition was determined by measuring the weight of the reference samples placed in the same humidity test chamber. Saturated salt solutions of

$CaCl_2 \cdot 2H_2O$, $Ca(NO_3)_2 \cdot 4H_2O$, $NaNO_3$, and KNO_3 were used to control the relative humidity in the chamber to 32, 51, 74, and 94 %, respectively.

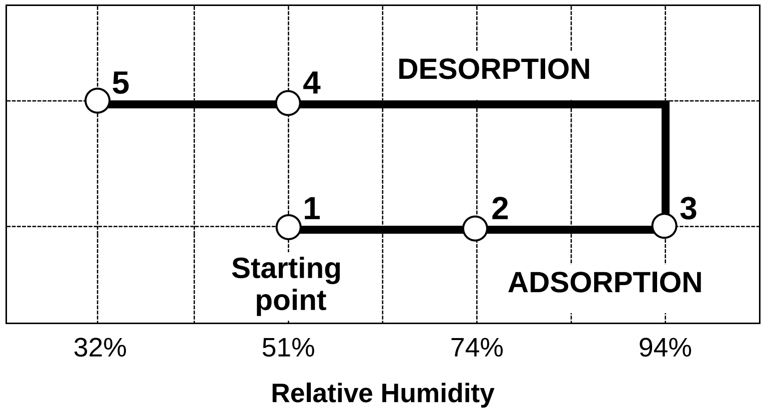


Fig. 4. Conditioning steps employed to test the effect of moisture adsorption and desorption on paper properties under five relative humidity conditions.

Testing of Handsheets

Apparent thickness was measured using an L&W micrometer (Kista, Sweden) based on TAPPI T411 om-97 (1997). Also elastic modulus was evaluated with an L&W tensile tester according to TAPPI T220 sp-01 (2001). Bending resistance of the samples was measured using an L&W bending resistance tester based on TAPPI T556 om-11 (2011). Thereafter, bending stiffness was calculated using the following Eq. 1,

$$S_b = \frac{60 \times F \times l^2}{\pi \times \alpha \times b} \tag{1}$$

where S_b is bending stiffness (mNm), F is bending force (N), l is bending length (mm), α is bending angle (15°), and b is sample width (mm). The elastic modulus, thickness, and bending stiffness were reported by displaying 95% confidence interval with error bar.

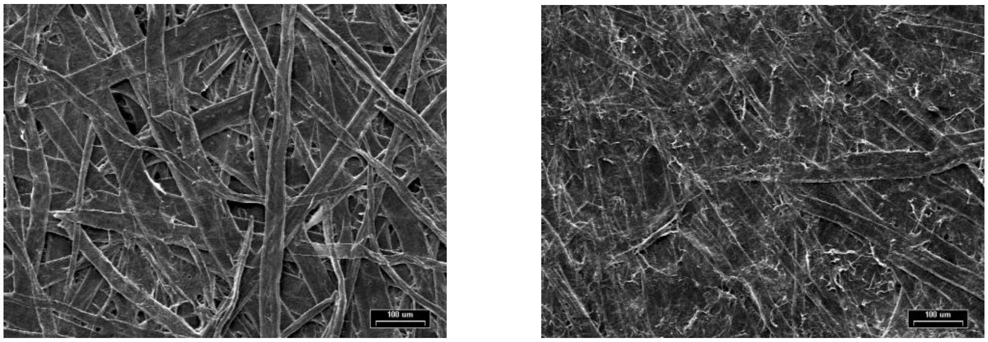
RESULTS AND DISCUSSION

Effect of Fines in the Single-Ply Handsheets

Single-ply UKP handsheets having different amounts of fines ranging from 0% to 20% at 5% increments were formed and dried. Figure 5 shows the scanning electron micrographs of two single ply handsheets containing no fines and 20% of fines. Fines addition changed the bulky sheet structure to a dense one. In addition, fines addition up to 15% increased tensile strength (Table 1).

Table 1. Tensile Strength of Single-ply Handsheets Containing Different Amounts of Fines at 51% RH, 23 °C

		S-00	S-05	S-10	S-15	S-20
Fines content (%)		0	5	10	15	20
Tensile strength (kN/m)	Average	7.55	8.51	9.01	9.68	9.72
	St. dev.	0.50	0.36	0.44	0.69	0.69



(a) S-00 **(b)** S-20

Fig. 5. SEM images for single-ply handsheets containing no fines (a) and 20% of fines (b).

Figure 6 shows the elastic modulus, thickness, and bending stiffness for single-ply handsheets containing 0% and 20% of fines as a function of moisture content.

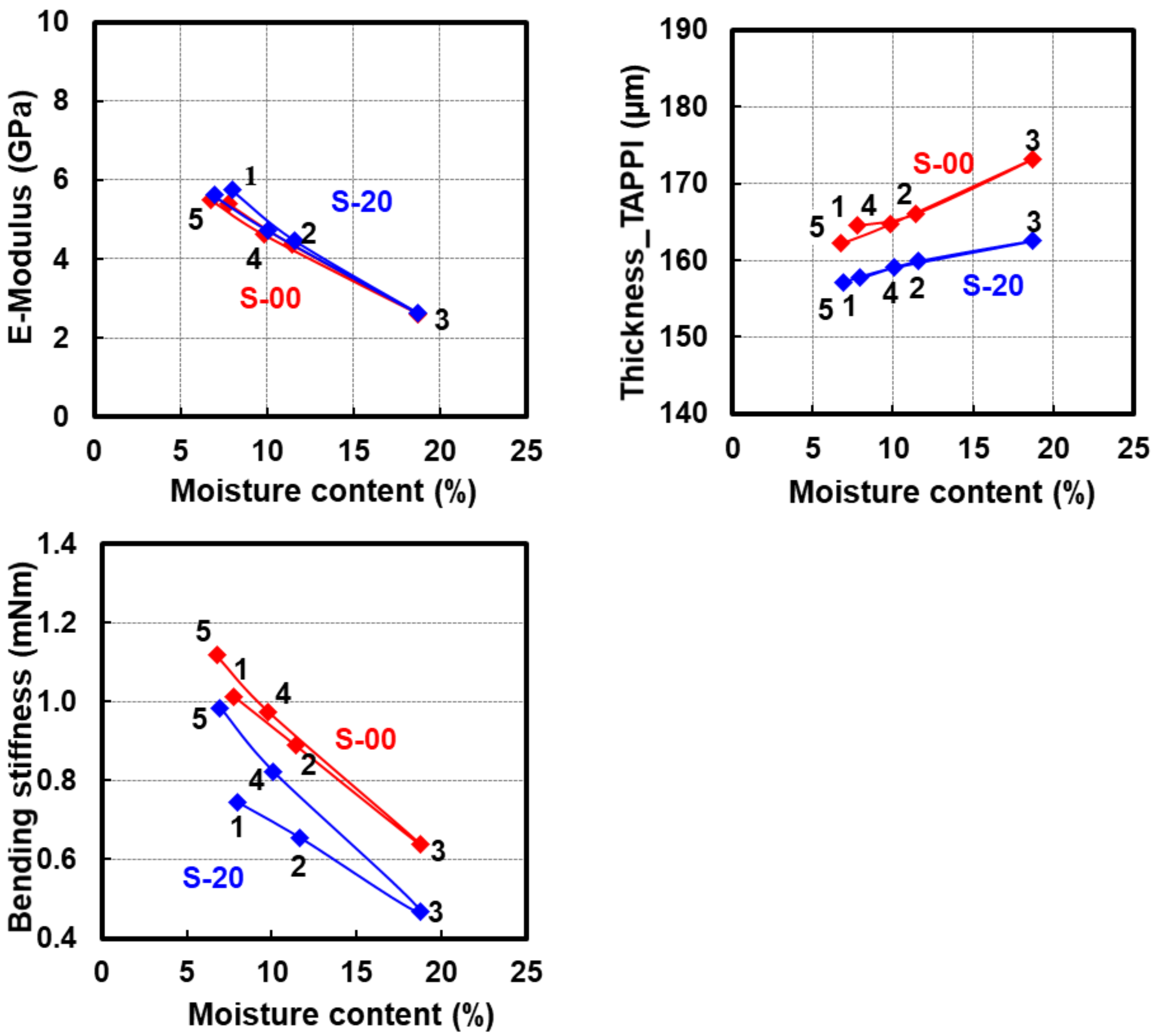


Fig. 6. Elastic modulus, thickness and bending stiffness for single-ply handsheets containing 0% (red) and 20% (blue) of fines as a function of moisture content. Numbers indicate the humidity conditions in Fig. 4.

The moisture contents at points 1 and 4 show the hysteresis effect of adsorption and desorption. With an increase in moisture content, the bending stiffness and elastic modulus of all handsheets decreased independent of the fines content. Thickness increased with an increase of the moisture content. A higher density is generally reported to increase hygroexpansion due to the increased inter-fiber contact and the effects of pore volume (Mao *et al.* 2003; Antonsson *et al.* 2009; Viguie *et al.* 2011; Erkkilä *et al.* 2015). However, the increase of the thickness for the handsheets without fines (S-00) was greater than that containing 20 % of fines (S-20), indicating that long fibers tend to expand more in the z-direction.

Single-ply handsheets showed reversible change in elastic modulus and thickness when they were subjected to cyclic humidity change irrespective of their fines contents. Bending stiffness of S-00 and S-05 also showed reversible change with the moisture content. However, the handsheets containing more than 10% of fines showed irreversible change in bending stiffness, and the irreversibility increased as the fines content increased (Fig. 6).

Bending stiffness is sensitive to the change of elastic modulus and sheet thickness. If both of these two parameters are reversible, the bending stiffness is expected to show a reversible change. In this respect, irreversible change in bending stiffness of handsheets with more than 10% of fines indicates that these two factors are not perfectly reversible or another artifact is involved. This was attributed to the fact that the apparent thickness, *i.e.* caliper, was used rather than true thickness to obtain the bending stiffness in Eq. 1. The apparent thickness is usually greater than the true thickness due to the surface roughness (Wink and Baum 1983; Sung *et al.* 2005).

Hygroexpansion of fiber mitigates the micro-compression of the crossing fiber segments and the longitudinal shrinkage of individual fibers (Uesaka and Qi 1994). Fibers, which deformed or micro-compressed by drying shrinkage, return to the original shape with increasing moisture content. If this structural recovery of fibers by adsorption remains during the next cycle of dehydration, it will affect the bending stiffness of handsheets, *i.e.*, it leads to higher bending stiffness of handsheets in desorption mode than adsorption cycle. It appears that the handsheets with greater amount of fines shrink more in the drying process and show irreversible changes of the effective thickness of handsheets upon adsorption of moisture. It appeared, however, the moisture absorption of fines did not cause much changes in apparent thickness.

Effect of Two-ply Handsheets

Figure 7 illustrates the scheme to make two-ply handsheets that contain different amounts of fines ranging from 5% to 20% at 5% increments. UKP was used to prepare two-ply handsheets, and a laboratory cylinder dryer at 120 °C was used for drying. Some important results of the elastic modulus, thickness, and bending stiffness of two-ply handsheets measured as a function of moisture content are shown in Fig. 8, and compared with the properties of single ply handsheets. In general, as in the case of single-ply handsheets, the bending stiffness and elastic modulus of two-ply handsheets decreased as moisture content increased. Thickness, however, increased with moisture content. Bending stiffness and elastic modulus showed reversible changes with moisture content in most cases.

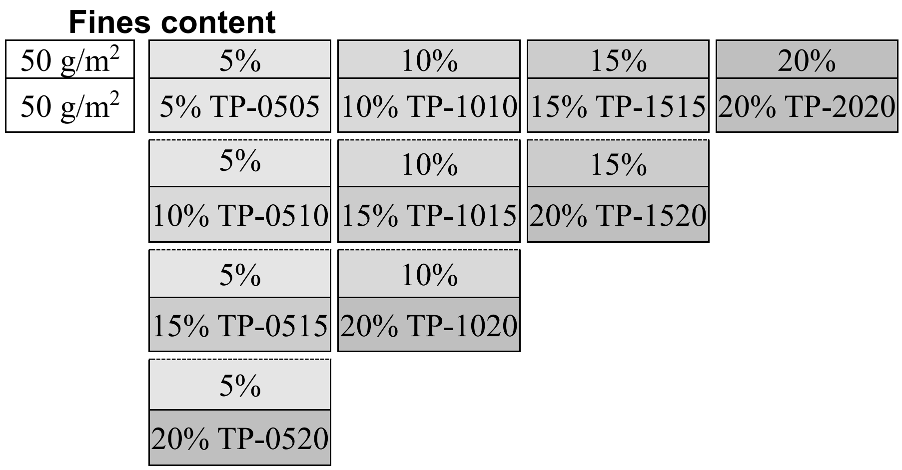


Fig. 7. The schemes to produce two-ply handsheets consisted of top and bottom plies with different amount of fines

The elastic modulus, thickness, and bending stiffness of single-ply and two-ply handsheets containing the same amount of fines, 5 and 20 %, are compared in Fig. 8. Two-ply handsheets were much thicker than the single-ply handsheets at low fines contents. However, the thickness difference between single- and two-ply handsheets decreased with an increase of fines content. For instance, the thickness difference containing 20% of fines was much smaller than that of handsheets with 5% of fines (Fig. 8), which suggested that close contacts between two plies was obtained at 20% of fines. In the case of elastic modulus, there was no significant difference between two types of handsheets. Two-ply handsheet, however, showed much greater bending stiffness than single-ply one. For example, when the moisture content was 10%, bending stiffness of single- and two-ply handsheets containing 5% of fines were about 0.8 and 1.0 mNm, respectively. This was attributed to increased thickness for two-ply handsheets. More uniform fines distribution in the two-ply handsheet may also played a role in increasing the bending stiffness. Because we have combined the top surfaces of 50 g/m^2 two handsheets to prepare a two-ply handsheet, the wire sides, which contained more fines due to the dilution controlled fines distribution in handsheets, were exposed to the surface. And this made the two-ply handsheet stiffer than single ply handsheet. This suggests that fines retention should be controlled to obtain high bending stiffness for the single-ply sheet. Two-ply handsheets containing 15% or more fines showed irreversible change in bending stiffness, but the irreversibility was much smaller for two-ply handsheet. In general, two-ply handsheets showed greater bending stiffness and better reversibility in cyclic humidity environment.

Effect of Restraint High Temperature Drying

To test the effect of restraint high temperature drying of single-ply handsheets produced with different amount of fines, a laboratory static Condebelt dryer was used (Lee *et al.* 2003). In the Condebelt process, the sheet is dried against a smooth and hot steel surface under z-directional pressure. In this experiments, the temperatures of top and bottom platens were maintained at 160 °C and 80 °C, respectively, and the handsheets was pressed between the two platens at a pressure of 5 bar for 5 sec. The basic principles and technical aspects of the dynamic and static Condebelt machines have been described (Lehtinen 1995a, b, 1998; Lee *et al.* 1999; Schlegel *et al.* 1999; Lee *et al.* 2003).

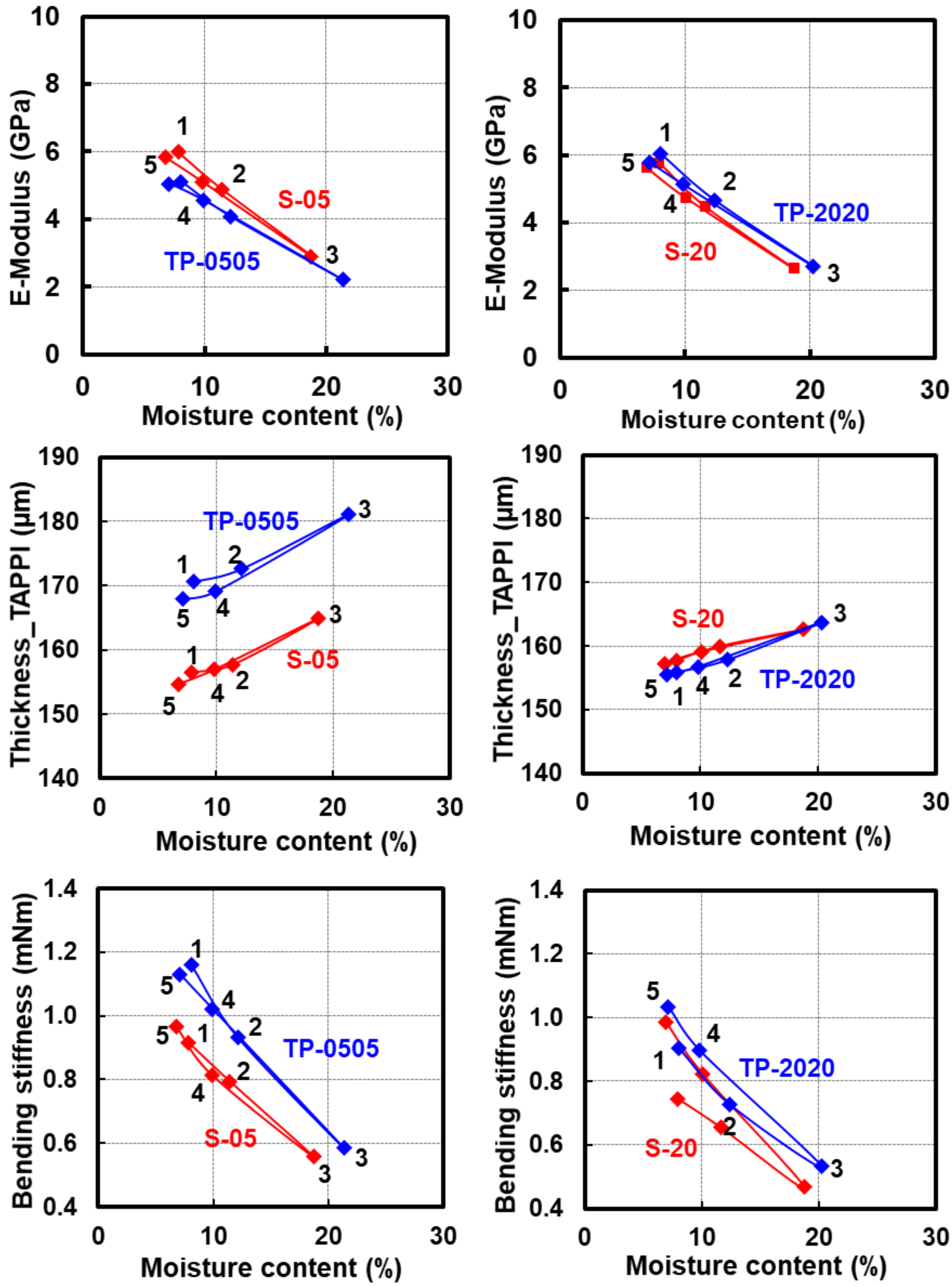


Fig. 8. Elastic modulus, thickness, and bending stiffness for two-ply handsheets containing 5% (TP-0505) and 20% (TP-2020) of fines as a function of moisture content, which compared with single ply handsheets. Numbers indicate the humidity conditions in Fig. 4.

The Condebelt drying has reported advantages in drying rates and strength improvements. A major part of the improved strength properties obtained by Condebelt drying has been explained by the increased sheet density under the restrained drying condition (Retulainen *et al.* 1998; Lee *et al.* 2003). Lehtinen (1995b, 1998) has shown that Condebelt-dried sheet has advantages in compressive strength because of the extensive fiber flattening, as well as the flow of lignin and hemicellulose. Recently, Joelsson *et al.* (2020) have shown that softening of high yield pulp fibers by hot-pressing densified the sheet by softening of lignin and produced very high dry and wet tensile strengths.

Figure 9 shows the elastic modulus, thickness, and bending stiffness as a function of moisture content for Condebelt-dried single-ply handsheets.

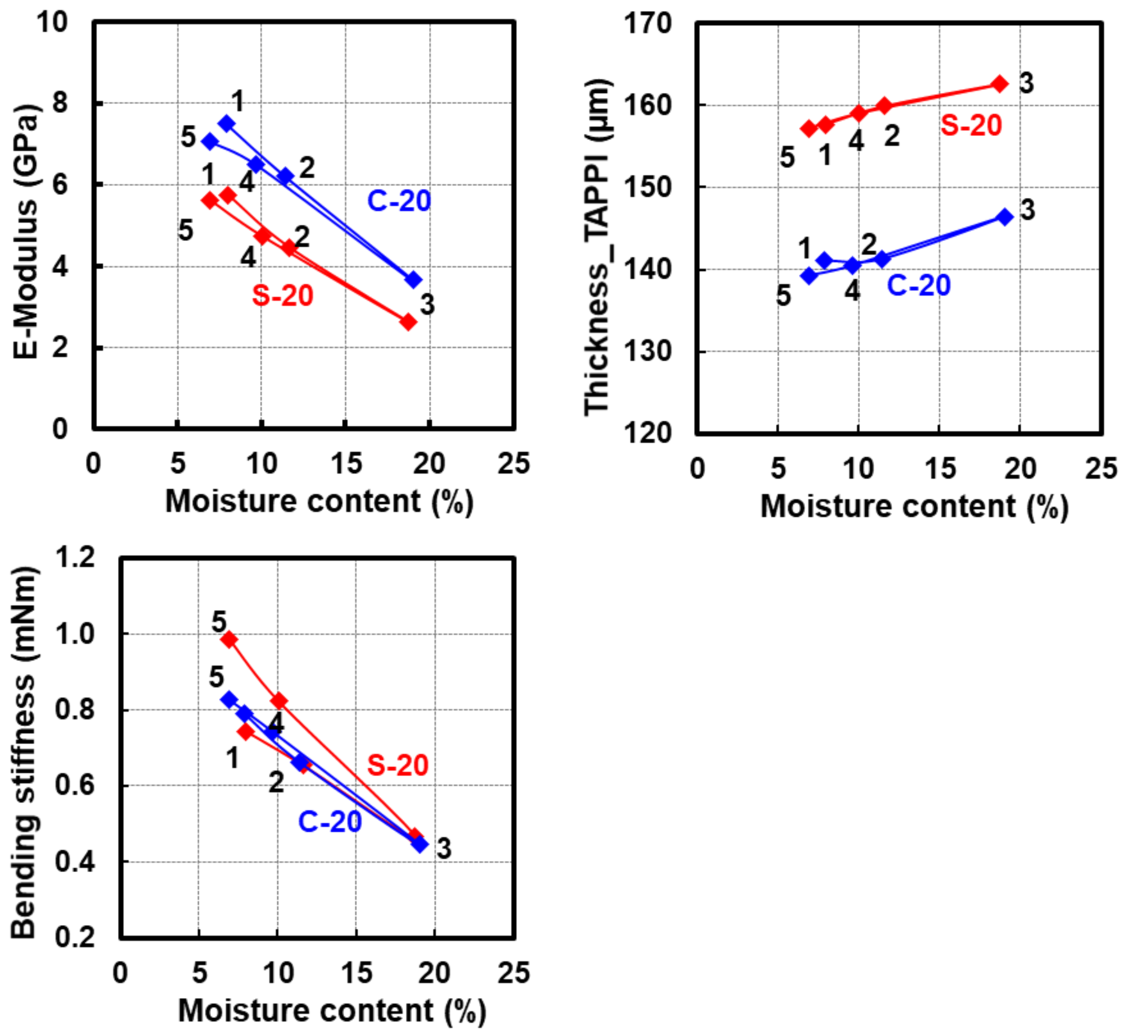


Fig. 9. Elastic modulus, thickness, bending stiffness and apparent density of Condebelt dried (C-20) and cylinder dried handsheets (S-20) containing 20% of fines.

Because of the densification effect by restraint high temperature drying, the thickness of handsheets was thinner by 20 µm than the conventional cylinder-dried ones. The increasing trend of thickness with moisture change for hot press dried and cylinder-dried sheets were almost similar. Lehtinen (1995a) pointed out that Condebelt dried paper is able to resist the adverse effects of moisture better than cylinder dried one, which improves the dimensional stability of the web. However, the present experimental results did not show this. When the densification of Condebelt dried handsheets is taken into account, the thickness increase of Condebelt-dried handsheets is far smaller than expected. Elastic modulus of Condebelt-dried handsheets was higher than that of cylinder-dried handsheets over the entire moisture cycle, which compensated the thickness reduction effect for bending stiffness.

The irreversible effect of fines on bending stiffness was also decreased (Fig. 9). While the cylinder-dried handsheet with 20% of fines showed irreversibility in bending stiffness during humidity cycle, Condebelt-dried handsheet with 20% of fines changed more or less reversibly as shown in Fig. 9. Under the initial humidity condition, Condebelt-dried handsheet showed higher bending stiffness than cylinder-dried one indicating that the effect of restraint high temperature drying is evident for the handsheets with a high fines content.

Generally, the changes of cellulosic fibrils in longitudinal and lateral direction are of the order of 1% and 20%, respectively, over the range of RH from 0% to 100% (Gallay 1973). The lateral contraction of an individual fiber induces a corresponding longitudinal shrinkage of the fiber bonded with it (Lindner 2017). Because of this transferring phenomena of the lateral shrinkage of a fiber to the longitudinal contraction of the crossing fiber bonded to it, often called microcompressions, a much greater longitudinal shortening of the fiber would result (Hartler 1995; Haslach 1996). Paper expands when the dimensional change of individual fibers transfers to the dimensions of macroscopic paper network (Navaranjan *et al.* 2013). The largest sheet shrinkage is obtained with highly beaten papers. The extent of shrinkage is different depending upon drying methods. Because Condebelt drying is a restraint-drying method, Condebelt-dried handsheets shrink less in the drying process. In other words, cylinder-dried handsheets would have more micro-compressions in the fiber crossing areas that accompanying greater longitudinal shrinkage of fibers. According to Salmén *et al.* (1987), hygroexpansive strain for kraft fibers was higher in the case of free drying than restrained drying, and in the case when the fines contents was higher. In other words, hygroexpansivity showed strong relationship with the drying shrinkage and fines contents. This explains the reason why the cylinder-dried handsheet with 20% of fines showed greater irreversibility in bending stiffness under moisture cycling than the Condebelt dried one.

Effect of Recycled Fibers

To demonstrate how recycled fibers would respond to moisture cycling, old corrugated containers (OCC) recycled in Korea was used to make single-ply handsheets with fines content ranging from 0% to 20% by 5% increments and dried at 120 °C with a laboratory cylinder dryer. The long fiber fraction of OCC had a low water retention value (WRV) of 1.19 g/g compared with the WRV of 1.84 g/g of UKP indicating a substantial loss of swelling and bonding potential of the recycled fiber.

Figure 10 shows that there were remarkable differences in thickness depending on the amount of fines for the recycled fiber. The thickness of handsheet increased as the long fiber fraction increased, which was much greater than that of single-ply handsheets made of UKP. In general, recycled fibers are stiffer and less conformable because of the phenomenon of irreversible hornification. Thus, they give weaker and bulkier sheets (Howard and Bichard 1992). The elastic modulus of OCC handsheets was considerably lower compared with that of UKP. However, OCC handsheets showed higher bending stiffness than UKP handsheet, indicating that the bulkier structure of recycled fibers compensated the loss of elastic modulus and increased the bending stiffness.

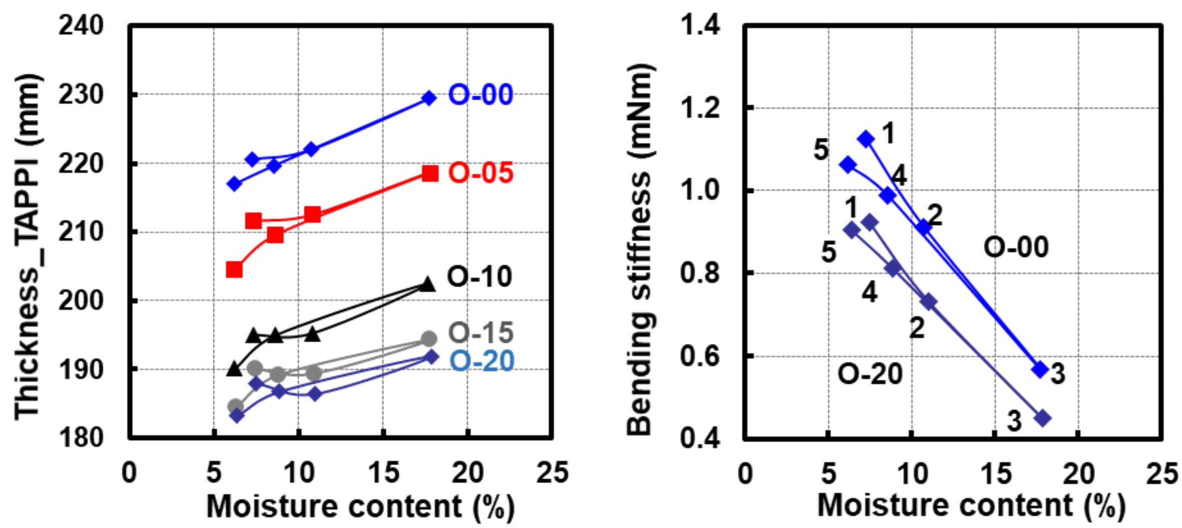


Fig. 10. Thickness of OCC handsheets with different amounts of fines, and bending stiffness of the OCC handsheets with 0% and 20% of fines

Interestingly, the elastic modulus and bending stiffness of OCC handsheets changed almost reversibly regardless of the amount of fines even for the OCC handsheet with 20% of fines. Recycled fibers that are irreversibly hornified are stiffer and shrink less than virgin fiber. The hornification of recycled fibers that collapsing the fiber and reducing the water absorbance leads to a decrease in hygroexpansion (Racz and Borsa 1997). Thus, handsheets made of recycled fibers do not have a same structural recovery potential as virgin fibers by hygroexpansion. Hence, in spite of high amount of fines, OCC handsheets showed the reversibility in bending stiffness over the entire moisture cycling range.

CONCLUSIONS

1. The effect of moisture content on the elastic modulus, thickness, and bending stiffness of single-ply and two-ply handsheets prepared using softwood unbleached kraft pulp (UKP) was investigated by exposing the handsheets to various humidity conditions. Also, the properties of Condebelt dried and old corrugated container (OCC) pulp handsheets were investigated and compared. The elastic modulus decreased, while the thickness increased as the moisture content of single and two-ply handsheets increased. For cylinder-dried single-ply handsheets made of UKP, substantial but reversible change in elastic modulus and thickness was observed when subjected to a change in moisture content. In the case of bending stiffness, handsheets with 0% and 5% of fines showed almost reversible change, but those with over 10% of fines showed irreversible change.
2. Two-ply handsheets were thicker than single-ply handsheets, which was more evident for low fines handsheets. This resulted in higher bending stiffness for the two-ply handsheets compared with the single-ply handsheet. More symmetrical distribution of fines with greater amounts on both surfaces appeared to give a higher bending stiffness for the two-ply handsheet.
3. Condebelt drying introduced the reversibility even for the bending stiffness of handsheets with 20% of fines. This suggested that restraint drying that suppressed the

micro-compression of fibers gave more reversible bending stiffness for the restraint dried handsheets.

4. The elastic modulus of OCC handsheets was considerably lower compared with that of UKP. However, OCC handsheets showed higher bending stiffness than UKP handsheet, indicating that the bulkier structure of recycled fibers compensated the loss of elastic modulus.

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