Producing Flexible Calcium Carbonate from Waste Paper and Their Use as Fillers for High Bulk Paper

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Microfibrillated cellulose (MFC) was prepared from post-consumer old corrugated container (OCC) material, which was first disintegrated in water, cleaned to remove impurities, and then fibrillated by grinding. Those processed MFCs were treated with *in-situ* formation of calcium carbonate by adding calcium oxide and injecting carbon dioxide into the mixture up to the ratio of 1:40 (MFC : calcium carbonate) by weight. The MFCs had a dark brown color initially but turned into high brightness materials similar to commercial ground calcium carbonate (GCC) after the *in-situ* formation process. The MFCs that had calcium carbonate attached on their surfaces, which were lengthy and flexible, were called flexible calcium carbonate from OCC (FCCO). Paper containing FCCO gave higher bulk, higher stiffness, and higher tensile index without lowering smoothness when compared to the paper containing commercial GCC. However, brightness was slightly lowered because of initial low brightness of the OCC. This study also demonstrated the feasibility to substitute wood fibers up to 5% with FCCO without lowering essential properties for printing paper. Benefits of the waste paper are savings of both wood resources and production cost.

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INTRODUCTION

Replacing chemical pulp with inorganic materials, such as calcium carbonate ($CaCO_3$), without loss of key paper properties in papermaking is beneficial for forest conservation and production cost savings (Svending *et al*. 2019). In reality, increasing 1% to 2% inorganic filler content in printing paper without loss of essential properties should be considered a great achievement in a paper mill. To produce highly filled paper, lumen loading of fillers by co-refining the fillers and the wood fibers (Bovin and Carnö 1977; Bown 1985; Kumar *et al*. 2011), and pre-flocculation of fillers by making flocs of precipitated calcium carbonate (PCC) or ground calcium carbonate (GCC) by applying electrolyte polymers (Gill 1993; Hjelt *et al*. 2008; Sang *et al*. 2012; Seo *et al*. 2012, 2018) was studied. The pre-flocculation and agglomeration methods decreased the surface area of the fillers per unit weight by making flocs and allowed more hydrogen bonding area between wood fibers to increase strength properties of the paper. Hybrid calcium carbonate (HCC) was prepared by producing by injection of carbon dioxide to the pre-flocculated flocs with GCC and calcium oxide, which could make semi-rigid flocs due to the newly formed PCC from calcium oxide (Jung and Seo 2015a, 2015b; Choi and Seo 2016; Choi *et al*. 2018; Kang *et al.* 2020). It was demonstrated that these semi-rigid flocs of HCC could improve the physical properties of its-containing paper remarkably and its physical properties could be as same as that of the paper containing 30% GCC (Kang *et al.* 2020).

Other approaches have been demonstrated using highly fibrillated cellulose materials. Cellulose micro- or nanofibril (microfibrillated cellulose; MFC or cellulose nanofibril; CNF) also have gained attention as the filler-modifying material due to its high surface area and bonding ability (He *et al*. 2016; Ottesen *et al*. 2016; Li *et al.* 2018; Lourenco *et al*. 2019; Song *et al.* 2022). They could improve the strength properties and smoothness of the paper, but failed to improve the paper bulk, which is the one of the critical properties of the printing paper.

Flexible calcium carbonate (FCC) was developed by forming $CaCO_3$ on the surface of cellulose nanofibrils through an *in situ* $CaCO_3$ formation method, in which the weight ratios of the newly formed $CaCO_3$ to nanofibrils ranged from 40:1 to 80:1 (Han *et al.* 2020; Kim *et al.* 2022). The FCC is flexible, is deformable, and has large size. The FCC gave extremely high bulk and high tensile strength simultaneously in paper and developed great smoothness after calendaring (Kim *et al.* 2022). The FCC is one of the promising applications of MFC and CNF for the paper industry. It enables the production of highly loaded paper without lowering product quality, and reduction of drying energy and production cost by saving the wood fibers (Han *et al*. 2020).

One of the challenges for using MFC and CNF in the paper industry is their high production costs, which is mainly attributed to the energy-intensive fibrillation process. Currently, many methods for reducing the production cost have been suggested, such as pretreatments using enzyme or chemicals, and some of them are now applied to the real production system in the paper mills (Henriksson *et al*. 2007; Pääkkö *et al*. 2007; Saito *et al.* 2009).

Using inexpensive raw material, such as recycled fiber such as old corrugated container (OCC), white ledger, old magazine (OMG), or old newspaper (ONP), is another way to reduce MFC and CNF production cost (de Assis *et al*. 2018; Rajinipriya *et al.* 2018; Copenhaver *et al.* 2021) instead of using expensive bleached kraft pulp (BKP). The resultant paper properties should be acceptable, especially with respect to brightness and strength properties. The dark brown color of recycled fibers from OCC was retained even at the MFC and CNF state, which might be a negative aspect for such applications demanding high bright color (Tajvidi *et al.* 2016).

In this study, the OCC from post-consumer old corrugated board was used for the production of MFC or CNF for producing FCC. They were cleaned and ground to the level of the fibrillated fines passing *via* 200-mesh screen and used to substitute for the cellulose nanofibrils that were normally prepared from virgin chemical pulp. The FCC from the fibrillated OCC is referred to as FCCO, and its potential to replace the FCC from the nano-fibrillated chemical pulp was examined in this study. If FCCO can replace FCC to some extent, savings in production and energy cost, and wood resources were expected.

EXPERIMENTAL

Materials and Handsheet Preparation

Omya Korea Inc. located in Hambek, South Korea donated the GCC, and its mean size was 2.0 μm. Calcium oxide was purchased from Korea Showa Chemicals Co. As a retention aid for papermaking, cationic polyacrylamide (C-PAM. MW 5 to 7 million g/mol. +5 meq/g) from CIBA Specialty Chemical (Kimcheon, Korea) was used at 0.1% based on the dry weight of the papermaking furnish. To make the handsheets containing the fillers, the authors used a mixture (20:80) of commercial softwood bleached kraft pulp (SwBKP; a mixture of hemlock, Douglas fir, and cedar) and hardwood bleached kraft pulp (HwBKP; a mixture of aspen and poplar) as the wood fiber furnish. Both materials came from Canada, and they were kindly supported from a paper mill in Korea. These wood pulps were refined together in a laboratory Hollander (valley type) beater until their freeness reached 500 mL CSF (TAPPI T227 om-99 (2003)). Then, after mixing fibers and fillers to make handsheets with the target filler contents of 20, 25, and 30 wt%, handsheets of 60 g/m^2 basis weight (TAPPI T205 sp-02 (2006)) were prepared. The ash content (TAPPI 413 om-93 1993), bulk (TAPPI T411 om-97 1997), tensile strength (ISO 1924-1 1992), Bekk smoothness (TAPPI T479 cm-99 1999), ISO brightness (ISO 2470-1 (2016)), opacity (ISO 2471 (2008)), and Gurley stiffness (TAPPI T543 om-00 2000) of the handsheets were measured according to the standard methods.

Preparation of Microfibrillated Cellulose from OCC

The disintegrated wet OCC of 4% consistency was kindly donated by a paper mill located in Ansan, Republic of Korea. The OCC was refined extensively with a laboratory Hollander beater to 200 CSF (Canadian standard freeness) as per TAPPI T227 om-99 (1999) as a pre-treatment. For removing the non-cellulosic fiber, such as inorganic material and dirt, the refined OCC was passed through a centrifugal cleaner. After cleaning, the ash content of OCC was reduced from 19.50% to 2.97% (TAPPI 413 om-93 1993). The MFC from OCC was prepared by fibrillation of OCC using a Super Masscolloider (Masuko Sangyo Co., Ltd., Kawaguchi, Japan), an ultra-fine grinding machine. The gap between the two grinding stones was kept at 120 μm from the zero position by controlling the bottom grinding stone after the pulp was loaded. Up to 10 passes were applied for the OCC in 2% consistency, and the portion that passed through the 200-mesh copper wire screen (74-μm hole size), was used for the experiment. The prepared MFC from OCC was named as MFCO. For comparison, another MFC was also prepared from HwBKP. The HwBKP was refined to 250 CSF initially and fibrillated by a Super Masscolloider with the procedure the same as for OCC except passed through 30 passes. The MFC from HwBKP was named as it is, MFC. Then, the dimensions of prepared MFCs were measured using a scanning electron microscope (SEM, S-4800, Hitachi, Tokyo, Japan).

Preparation of Flexible Calcium Carbonate (FCC)

Flexible calcium carbonate batches from MFCO and MFC were prepared according to previous reports (Han *et al.* 2020; Kim *et al.* 2022). In short, to prepare FCC, the consistency of the MFCs was controlled to 0.2% at 30 °C. One liter of MFC suspension in a 2-L beaker has 2 g MFC (dry weight basis), and 44.9 g of calcium oxide was added to make 80 g by $CaCO_3$ by the reaction with injected pure carbon dioxide (flow rate 3 L/min.). The weight ratio of $CaCO_3$ to MFC for the FCC was 40.0. The temperature was held at 30 °C initially and raised by the exothermic reaction to 33 to 34 °C at the end while stirring at 350 rpm. When the pH reached 7.0, the authors waited two more minutes to confirm the stable pH and terminated the reaction.

RESULTS AND DISCUSSION

Morphology of Microfibrillated Cellulose and Flexible Calcium Carbonate

The FCC was prepared by coating a high amount of $CaCO_3$ (40 to 80 times by MFC weight) on the surface of MFC by an *in-situ* carbonation process. *In-situ* carbonation process can improve the optical properties such as brightness of recycled fibers by increasing the light scattering and covering colored impurities with newly formed $CaCO_3$ (Seo *et al.* 2017).

Therefore, the brightness difference between FCCO and FCC could be remarkably reduced due to the fact that the newly formed $CaCO_3$ covered the surface of the MFCO and the colored impurities while bright difference between MFC and MFCO was very high. Finally, the brightness of the FCCO containing paper would reach the level of the commercial printing paper containing GCC.

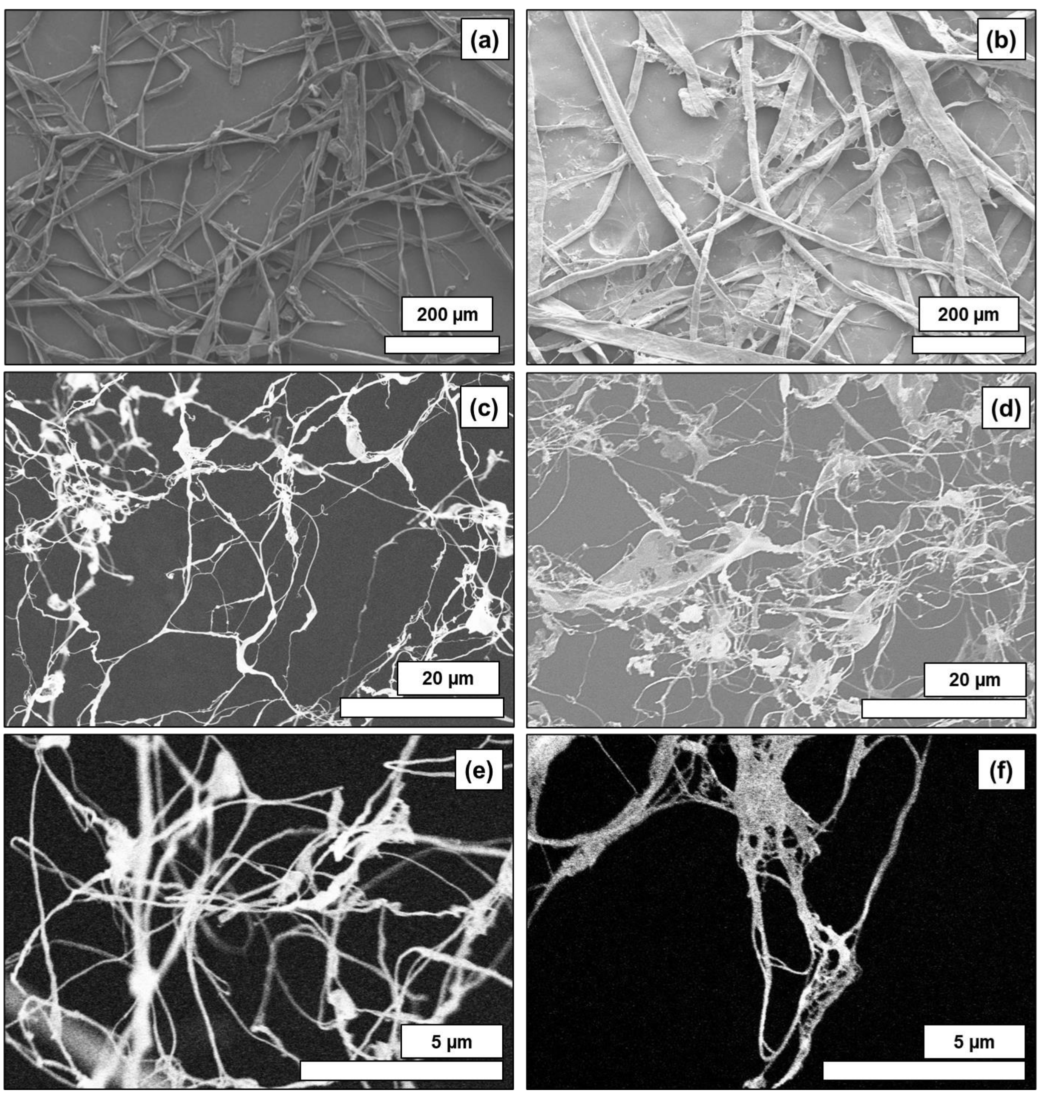

Fig. 1. SEM images of the HwBKP (a) and OCC (b) and their fibrillated samples; MFC (c) from HwBKP and MFCO (d) from OCC, respectively. Their higher magnifications were also shown in MFC (e) and MFCO (f).

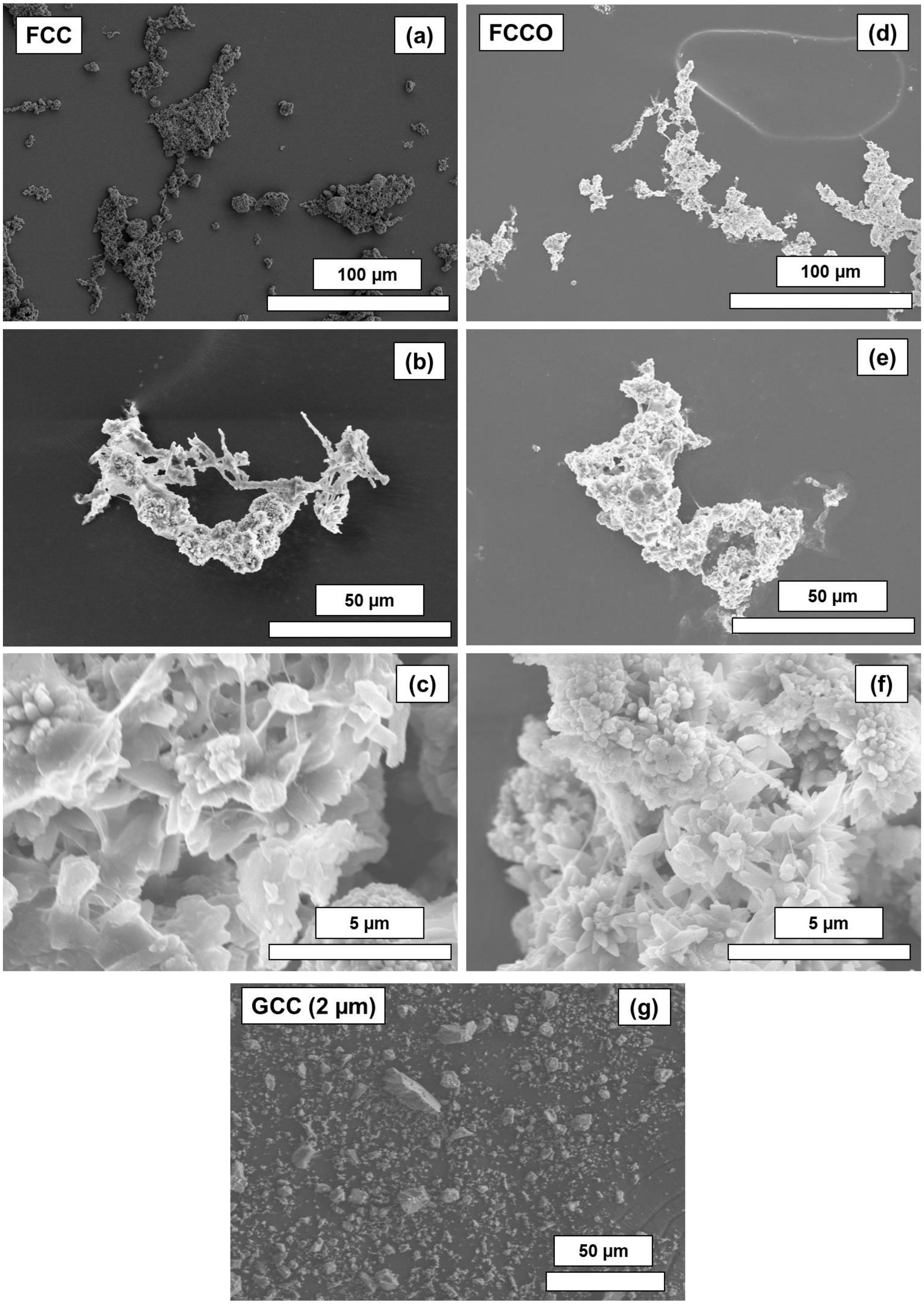

Fig. 2. Micrographs of flexible calcium carbonate from MFC (FCC, a toc), and MFCO (FCCO, d to f). and GCC (g). The images (b), (e) and (c), (f) are high magnification of 1K, and 10K, respectively for each FCC.

Figure 1 shows the morphology of HwBKP (Fig. 1a), OCC (Fig. 1b), and their MFCs. Regardless of cellulose source, cellulose fibrils (Fig. 1c to f)) that were prepared had similar width dimension of 100 to 200 nm, which was much larger than the limit dimension of 'nanofibrils' (30 nm) according to the definition of the TAPPI standard (TAPPI WI 3021 2012). Therefore, the prepared cellulose fibrils should be called microfibrillated cellulose (MFC). In SEM images, no characteristic differences were found between MFC and MFCO in their morphologies. According to Han *et al*. (2020), more fibrillated cellulose such as nanofibril could make FCC with higher aspect ratio and could improve the FCC-filled paper properties, including breaking length and folding endurance. However, even FCC from the less-fibrillated cellulose like MFC still has its extraordinary characteristics to improve many useful properties of paper (Han *et al.* 2020).

Figure 2 shows the morphology of FCCs prepared from MFC and MFCO. Regardless of their sources, both FCCs, FCC, and FCCO, had similar morphology and the dimensions of the width over 20 μm and the length over 200 μm. The morphology of GCC is presented in Fig. 2g for a comparison. While GCC exhibited a wide distribution of small-sized particles, the FCCs did not have any small particles, indicating that all $CaCO_3$ were successfully formed on the surface of the cellulose fibrils by *in-situ* carbonation process. In the micrographs of FCCO at high magnification (Figs. 2e and 2f), the $CaCO_3$ aggregates were attached on the bundle of cellulose fibrils, which indicated that the brown color of MFCO might be coated by white $CaCO_3$.

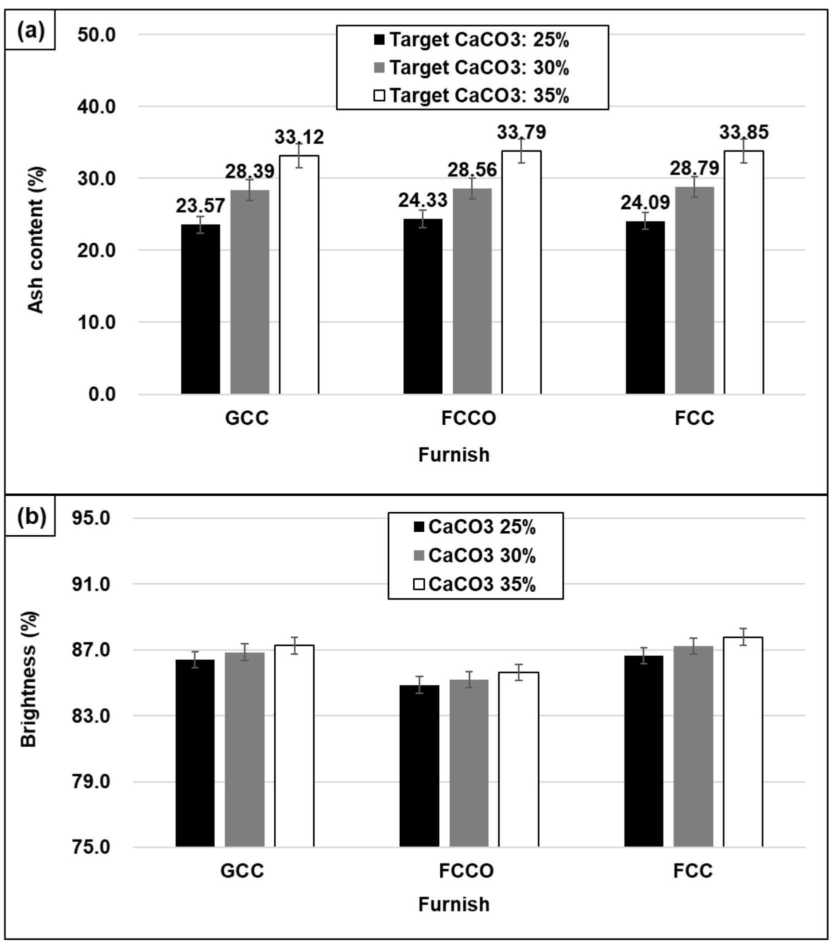

Fig. 3. Ash content (a) and brightness (b) of the papers containing three different fillers

Optical Properties of Papers

The FCC and GCC-filled papers with three different target contents of fillers were prepared. The ash contents of papers are presented in Fig. 3a. The ash content of three different filler-containing papers were similar at each target filler content. Thus, the physical properties of papers containing different filler at each target ash content could be compared.

Figure 3b shows the brightness of FCCs and GCC-containing paper. The FCC-containing paper exhibited slightly higher brightness value than GCC-containing paper at three different ash contents. The FCCO, FCC from MFCO, also showed lower values than GCC (1% to 2% lower). The OCC used for making MFCO and FCCO initially had a low brightness value of 28.9% while the HwBKP for making MFC indicated 81.5%. Therefore, *in-situ* carbonation process covering the surface of OCC with $CaCO_3$ could change the brightness of the FCCO to the level of GCC that is used for making printing paper.

Morphology of the Surface of the Filler-Containing Papers

Figure 4 shows the typical surface images of the filler-containing papers with 30% filler contents. From the image of GCC-containing paper, small GCC particles appeared to be distributed randomly in its containing paper, which would highly affect the paper properties such as paper bulk and strength. FCC and FCCO exhibited similar morphologies of the surface of their containing papers, where fillers existed as the clusters and clean fiber surfaces were observed. The large size of clusters would improve the paper bulk. It also would maintain the strength properties of their containing paper by causing less interruption of fiber-fiber bonding in the paper in comparison to GCC.

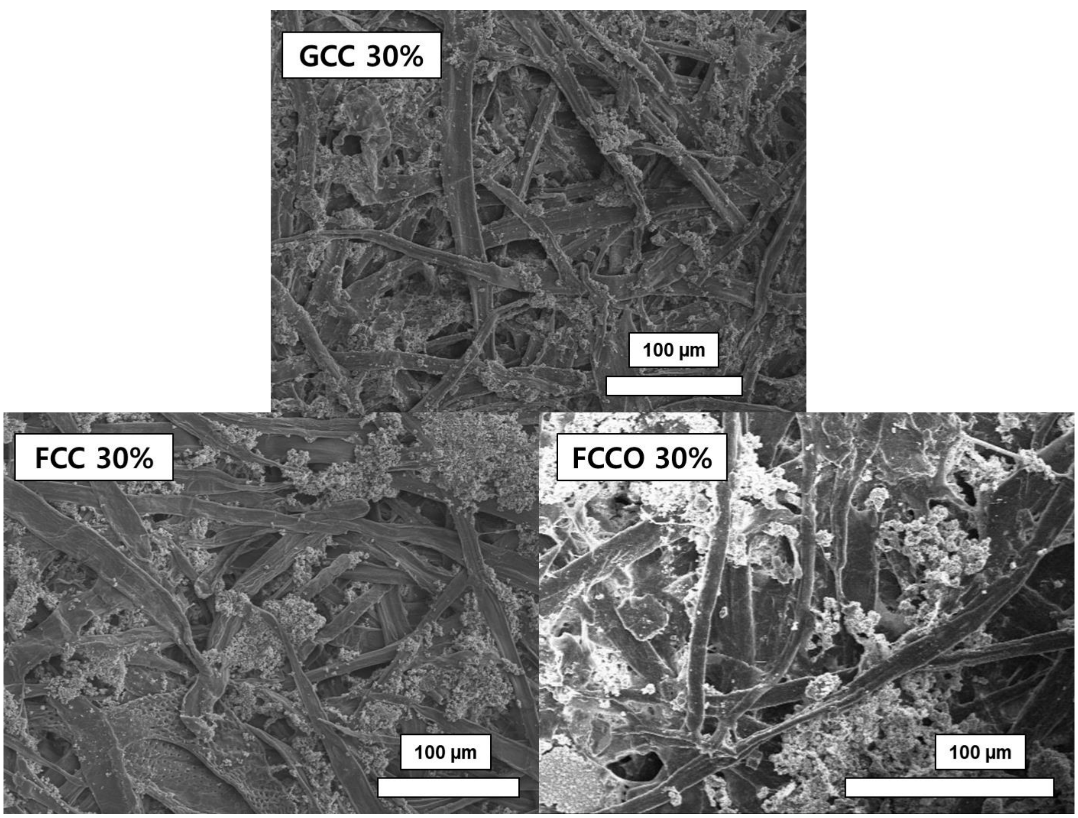

Fig. 4. Typical SEM images of GCC, FCC, FCCO-containing paper with the ash content of 30%

Physical Properties of Paper

The bulk, stiffness, and bulk smoothness of papers containing three different fillers are shown in Fig. 5. As reported previously by Han *et al*. (2020), the FCCs from both MFCs developed remarkably higher bulk of paper than GCC, and bulk increased with increasing filler content, unlike that in GCC (Fig. 5a). The high bulk of FCC was because of the large size of FCC as shown in the micrographs in Fig. 2. Even though the bulk of FCCO from OCC exhibited lower than FCC from MFC, it was still much higher than GCC.

Because the stiffness is proportional to the cube of the paper thickness (≈ bulk), high bulk can contribute to high stiffness. Indeed, high bulk of FCC resulted in high stiffness, as presented in Fig. 5b. Additionally, while the stiffness of GCC-containing paper decreased highly by increasing its ash content to 35%, the decreased rate of stiffness was much lower for the FCC-containing ones regardless of the cellulose source

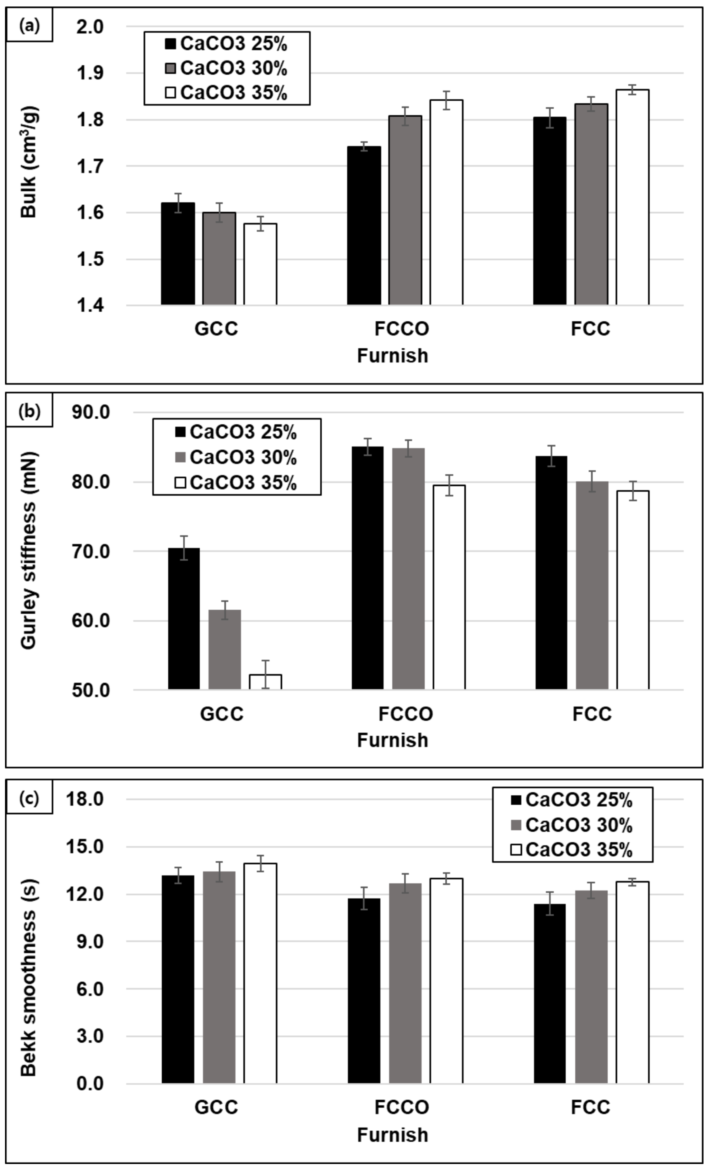

Fig. 5. Bulk (a), Gurley stiffness (b), and Bekk smoothness (c) of the papers containing three different fillers

The large size of filler, such as FCCs, can contribute to high bulk of the paper, but its smoothness might be worse and might decrease as the filler content increases (Kim *et al*. 2022). However, smoothness of FCC-containing papers was not much lower than GCC, as presented in Fig. 5c and it increased slightly as the filler content increased. As previously reported, the cellulose fibril core in the FCCs makes it flexible and deformable under pressure, which can help to maintain the smoothness (Han *et al*. 2020). This critical characteristic of FCC was not changed when using the MFCO as the core of FCC. Furthermore, Kim *et al*. (2022) showed that the FCC-containing paper had better response to the calendering pressure than the GCC-containing one to achieve a higher smoothness value.

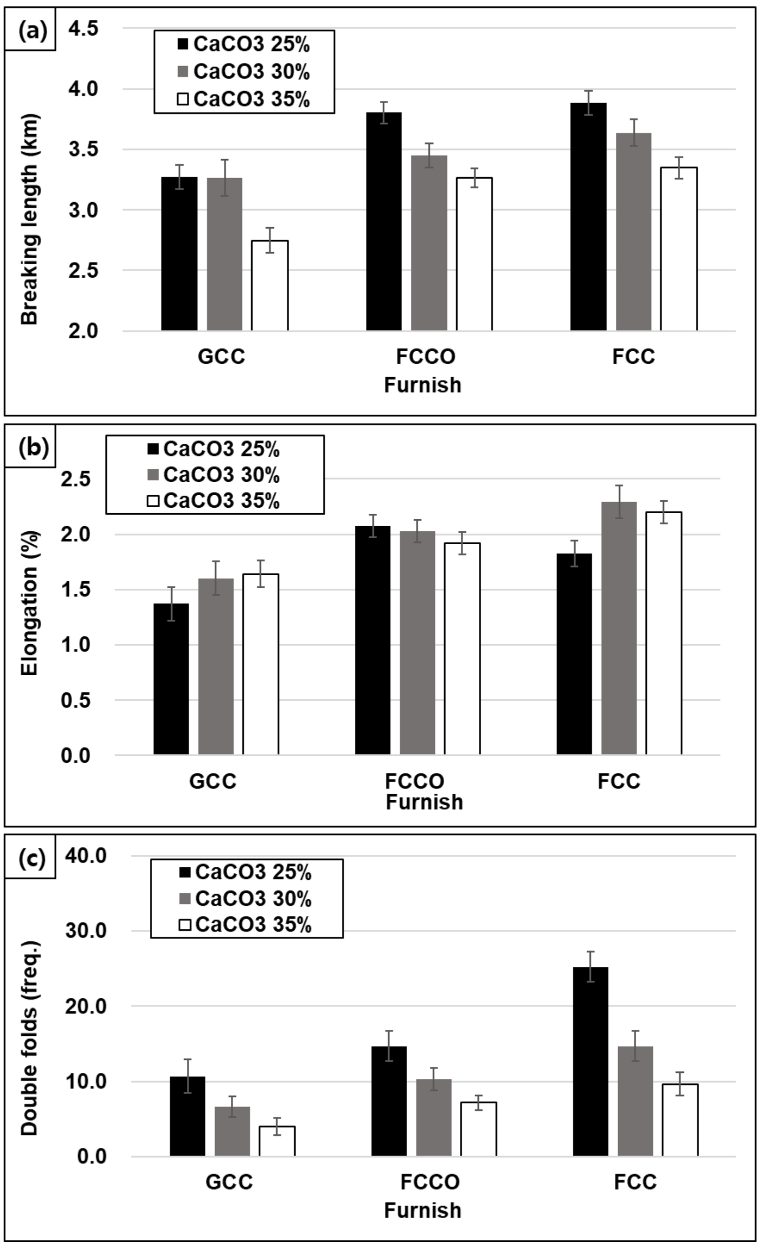

Fig. 6. Breaking length (a), elongation at break (b), and folding property (c) of the papers containing different fillers

Tensile strength and folding properties of FCCs and GCC-containing papers are shown in Fig. 6. In Fig. 6a, the tensile strength of paper is exhibited as the breaking length, which is a measure of tensile strength after compensating for the basis weight differences. The breaking lengths of FCC-containing papers showed highest values in all papers at the same ash contents. The breaking length value of FCCO was slightly lower than FCC, but it was also much higher than GCC. Higher strength of FCCs than GCC was attributed to its large size to make the total filler surface area small and less presence of small particles in FCCs as shown in Fig. 2 (Han *et al.* 2020). Lower strength of FCCO compared to FCC might have been due to the lower strength of the cellulose source, recycled fiber (Ghahrani *et al.* 2023). However, it should be studied further in detail in the future. Elongation at break in Fig. 6b also shows the same trend with the tensile strength.

The folding endurance is related to the paper cracking during folding. The FCC exhibited the highest value, FCCO was next, and GCC was lowest. Lower folding endurance value of FCCO than FCC might be related to the strength of cellulose fibril core, but this should be further studied in detail in the future.

CONCLUSIONS

1. Post-consumer old corrugated container (OCC) was used to prepare microfibrillated cellulose OCC, which is here called MFCO. The MFCO was used as the cellulose fibril core of the flexible calcium carbonate OCC (FCCO) by attaching a large amount of $CaCO_3$ on the surface of MFCO (40:1 weight ratio) by an *in-situ* carbonation process.
2. The FCCO had similar morphology to the FCC from HwBKP pulp. There was a large difference of ISO brightness between OCC and HwBKP (brightness difference of 52.6%). However, the difference was decreased to less than 3% when they were transformed to FCCO from OCC, and FCC from HwBKP.
3. The FCCO was as good as FCC in developing paper bulk, tensile strength, and stiffness but was slightly lower in brightness and folding endurance due to the difference of the original source materials. Both FCC and FCCO contributed to much higher bulk, tensile strength, and stiffness when compared to GCC in paper.
4. This study shows the utilization of waste paper for the preparation of highly functional filler, FCCO, which could lead to the reduction of the use of virgin wood fibers, lowered production cost, and energy savings during paper manufacture.

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