Effects of Precoating Color Formulation with Coarse Ground Calcium Carbonate and Porous Precipitated Calcium Carbonate on Paperboard Properties and Printability

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To identify suitable pigments for the precoating of paperboard, the rheological properties of coating colors and their effects on the surface and printing properties of coated paperboard were evaluated with respect to the type and combination of coating pigments. The investigation included porous precipitated calcium carbonate (PCC) and four types of coarse ground calcium carbonate (GCC) of different sizes. As the GCC particle size increased, the viscosity of the coating color decreased in the low-shear region, and the degree of dehydration increased. Coatings containing PCC, which comprised relatively small and highly porous particles, were found to be less dehydrated than coatings containing only GCC. The surface roughness of the coated paperboard increased as the GCC particle size increased, leading to reduced paper gloss. However, increasing the GCC particle size decreased the binder usage and increased surface strength. In conclusion, it is believed that the use of 55-grade GCC rather than the smaller size of 60-grade GCC can reduce costs and enhance surface strength by reducing binder and energy.

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INTRODUCTION

The paper industry has encountered rising production costs and stagnant sales prices due to increased expenses for raw materials, energy, and transportation, as well as inflation driven by the pandemic and war in Europe (Thitsartarn and Jinkarn 2020; Gurtu *et al.* 2022). Despite being able to leverage natural and sustainable resources, there is an urgent need for cost reduction and enhanced quality. Specifically, manufacturers of printing paper and paperboard are currently investigating diverse research paths to economize; to this end, they are exploring better use of low-grade pulp, high loading of inorganic fillers, energy-saving technologies such as heat pumps and hot presses, and improvements to base paper-coating methods (Dong *et al.* 2008; Ilkka *et al.* 2012; Abd El-Sayed *et al.* 2020; Joelsson *et al.* 2020). Among these, the use of paper-coating technology has emerged as a method to reduce production costs by utilizing existing infrastructure, making it an attractive option over other technologies.

Advancements in coating technology have been realized by exploring different coating methods, enhancing the surface properties of base paper, and optimizing binder formulation (Laudone *et al.* 2006; Li and He 2012; Aslannejad *et al.* 2019). In particular, the size, distribution, and shape of pigment particles significantly influence the paper's porosity, quality, and production costs.

In the 1990s, high-solids coating solutions were developed by using calcium carbonate slurry; the reduced drying times led to energy savings, which impacted cost reduction and productivity improvements (Nath and Kline 1995; Günthert *et al.* 1989). According to previous research (Fadat *et al.* 1986; Tsunekawa 2000; Jeong *et al.* 2018), high-concentration coating colors positively affect the surface properties of paper, printability, and print mottle enhancements. However, challenges such as diminished water retention, higher high-shear viscosity, and difficulties in coating application control stem from inappropriate high-solids coating concentration levels.

Another technology to improve coating is the multi-layer coating technique (Wang *et al.* 2009; Nutbeem *et al.* 2011; Kim *et al.* 2016). This approach, involving several coating layers with varied formulations, aims to minimize binder migration and accelerate printing speeds (Sand *et al.* 2008; Kumar *et al.* 2011; Youn and Lee 2022). Nonetheless, it necessitates precise control over each layer and careful consideration of the mechanism by which the coating layers interact during the application process. In multi-layer coating, the average particle size, specific surface area, and surface characteristics of the pigments are crucial, as they significantly impact the coating's runnability and final product's quality. Laudone *et al.* (2006) explored the structure of coating layers for a mix of two particle sizes of calcium carbonate and latexes with different glass transition temperatures; alternatively, Nechita (2020) studied the influence of binder content and pigment type on the structural properties and liquid penetration properties of the coating layer.

Common practice involves using small-particle-sized 60-grade GCC for paperboard precoating, a choice that is also prevalent in precoating for printing papers. Despite the rough surface of paperboard's top layer, which often results from recycled material, the 60-grade GCC is preferred. However, applying GCC that is coarser than 60-grade GCC in the precoating allows the rough surfaces of paperboard base paper to be filled with larger particles, thereby enhancing smoothness, reducing binder consumption, and reducing the energy required to grind pigments, ultimately leading to cost savings (Preston *et al.* 2017; Gaskin *et al.* 2019). Moreover, it has been documented that porous PCCs can crystallize, thereby altering both the chemical and physical properties of paper when used as coating pigments and fillers (Subramanian *et al.* 2007; Kumar *et al.* 2011). However, integration of coarse GCC with porous PCC for precoating layer control is an under-explored area, and systematic research on coating color properties and their effects on the product under the combined conditions is urgently needed.

This study devised suitable pigments and coating conditions for paperboard precoating by applying various particle sizes of coarse GCC and porous PCC. It evaluated the mechanism of influence of the morphological characteristics of these pigments on the coating solution and final product's quality. Four types of coarse GCC, categorized by particle size, were utilized. This study entailed measuring the rheological properties of the coating color and assessing its quality impact on the final paperboard and printing quality after applying the top-coating process.

EXPERIMENTAL

Materials

The grammage of the base paperboard was 300 g/m^2. To investigate the effects of the coarse GCC particle size in the precoating layer on the properties of paperboard, four types of GCC (GCC 60, GCC 55, GCC 50, and GCC 45, Taekyung, Goesan, Republic of Korea) and porous PCC (colloid type, amorphous shape, Taekyung, Goesan, Republic of Korea) were applied as precoating pigments. When porous PCC was applied, GCC and PCC were fabricated at a weight ratio of 9:1. GCC 95 (Taekyung, Goesan, Republic of Korea) was applied as the pigment for the top-coating color. The physical properties of coating pigments are described in Table 1. Styrene/butadiene (S/B) latex (LG Chemical Co., Yeosu, Republic of Korea) was applied as a binder. Polyacrylate copolymer rheology modifier (CV-5001F, Cheong Woo Technology Co., Ltd., Jincheon, Republic of Korea), calcium stearate lubricant (LUB.REX-55, calcium stearate type, WooJin Chemical Co., Ltd., Gunsan, Republic of Korea), and insolubilizer (PRO.WET-400G, WooJin Chemical Co., Ltd., Gunsan, Republic of Korea) were used as additives.

Table 1. Physical Properties of Coating Pigments

Pigments	Mean diameter (µm)	Brightness (%)	pH
GCC 60	1.20	91.0	9.0-10.0
GCC 55	1.62	91.8	
GCC 50	1.96	94.3	
GCC 45	2.18	90.4	
GCC 95	0.58	93.3	
PCC	0.11	95.8	

Deriving the Pre- and Top-coating Color Formulations

Tables 2 and 3 show the precoating color formulations. Precoating colors were prepared by dispersing GCC and PCC using a stirrer, followed by the addition of the latex binder and small amounts of additives. The solids content of the precoating color was adjusted to 65% by using distilled water. The precoating colors obtained by using only GCC were designated as G1, G2, G3, and G4 (Table 2). In the case of Group-GCC+PCC, GCC and PCC were mixed at a ratio of 9:1, and the precoating colors using GCC and PCC were designated as P1, P2, P3, and P4 (Table 3). In addition, as shown in Table 4, GCC 95 was used as the pigment of the top-coating color, and the solids content was adjusted to 65%, which is typically applied in the industry, by using distilled water.

Table 2. Precoating Color Formulations for the Group Only GCC

Components		Units	G1	G2	G3	G4
Pigments	GCC 60	Parts by weight	100	-	-	-
	GCC 55		-	100	-	-
	GCC 50		-	-	100	-
	GCC 45		-	-	-	100
	Porous PCC		-	-	-	-
Binder	SB-latex		8.5			
Additives	Rheology modifier		0.05			
Total solids content		%	65			

Table 3. Precoating Color Formulations for Group-GCC+PCC

Components		Units	P1	P2	P3	P4
Pigments	GCC 60	Parts by weight	90	-	-	-
	GCC 55		-	90	-	-
	GCC 50		-	-	90	-
	GCC 45		-	-	-	90
	Porous PCC		10			
Binder	SB-latex		8.5			
Additives	Rheology modifier		0.05			
Total solids content		%	65			

Table 4. Top-coating Color Formulation

Components		Units	Top Coating Color
Pigments	GCC 95	Parts by weight	100
Binder	SB-latex		9
Additives	Rheology modifier		0.1
	Lubricant		0.3
	Insolubilizer		0.2
Total solids content		%	65

Preparation of Double-coated Paperboard

Double coated paperboard was coated on one side by using a laboratory semi-automatic coater (K-control, RK Print Coat Instrument Co., Ltd., UK). The coating amounts of the precoating and top-coating layers were respectively adjusted to 30 and 10 g/m^2 by using a rod bar (precoating: rod no. 20, top coating: rod no. 4).

The coated paperboard was dried at 105 °C for 25 s in a hot air dryer (YJ-8600D, Yujin Electronics, Republic of Korea) and then calendered twice at 70 °C under a linear pressure of 2 MPa using a super calender (Beloit Corporation, USA).

Evaluation of the Coating Color Properties

A rheometer (TA Instruments Co., Ltd., USA) was used to analyze the rheological properties of the coating colors. The apparent viscosity of the coating color was measured within the shear rate range of 0.1 to 100 (*i.e.*, 1/s), and its viscoelasticity was measured at a deformation of 1% and frequency of 0.1 to 100 rad/s.

The dewatering amount was measured by using a water retention meter (ÅA-GWR, Kaltec Scientific, USA). The principle of this method is based on the measurement of the amount of water passing through a cellulose ester membrane with a pore size of 0.2 μm to the absorbent paper. Specifically, 10 mL of coating color was poured into a circular test cell with an area of 8 cm^2, and a pressure of 2 bar was applied for 60 s. The degree of dehydration of the coating color was measured, and the dewatering amount was calculated according to following equation,

$$\text{Dewatering amount (g/m}^2\text{)} = (w) \times 1250 \qquad (1)$$

where w is the degree of dehydration from the coating color. Depending on the measurement method, the dewatering amount refers to a value related to the water retention of the coating color during the measurement.

Evaluation of the Properties and Printability of Coated Paperboard

The roughness of coated paperboard was measured by using a Parker Print Surf (L&W Co., Ltd., Sweden). Furthermore, the optical properties of the coated paperboard were measured by using a gloss meter (model: T480A, Technidyne Corporation, USA) and an Elrepho 3300 system (Datacolor International, USA).

The printability of coated paperboard, including ink set-off, ink trapping, and dry pick strength, was measured by using a printability tester (RI-II, KRK, Japan) and Image-J software (Image-J 1.53a, Bharti Airtel, Ltd., USA). The surface of the coated paperboard was observed by using scanning electron microscopy (SEM, CX-200TM, COXEM, Ltd., Republic of Korea). The surfaces of the precoating layer were sputter-coated with platinum in preparation for SEM analysis, which was conducted at a magnification of 5000x.

RESULTS AND DISCUSSION

Rheological Properties and Dewatering Amount of Coating Colors

Figure 1 shows the viscosities of the coating colors according to the shear rate. Regardless of the type of pigment, at the low shear rates, the coating color viscosity decreased as the particle size increased. Furthermore, the mixing of porous PCC led to higher viscosity than that for the group Only GCC, and all coating colors exhibited shear-thinning behavior. In general, the smaller the particle size and larger the number of pores, the larger the specific surface area. Small particles with a large specific surface area generally require more energy to move within the suspension while maintaining a random distribution (Fadat *et al.* 1986).

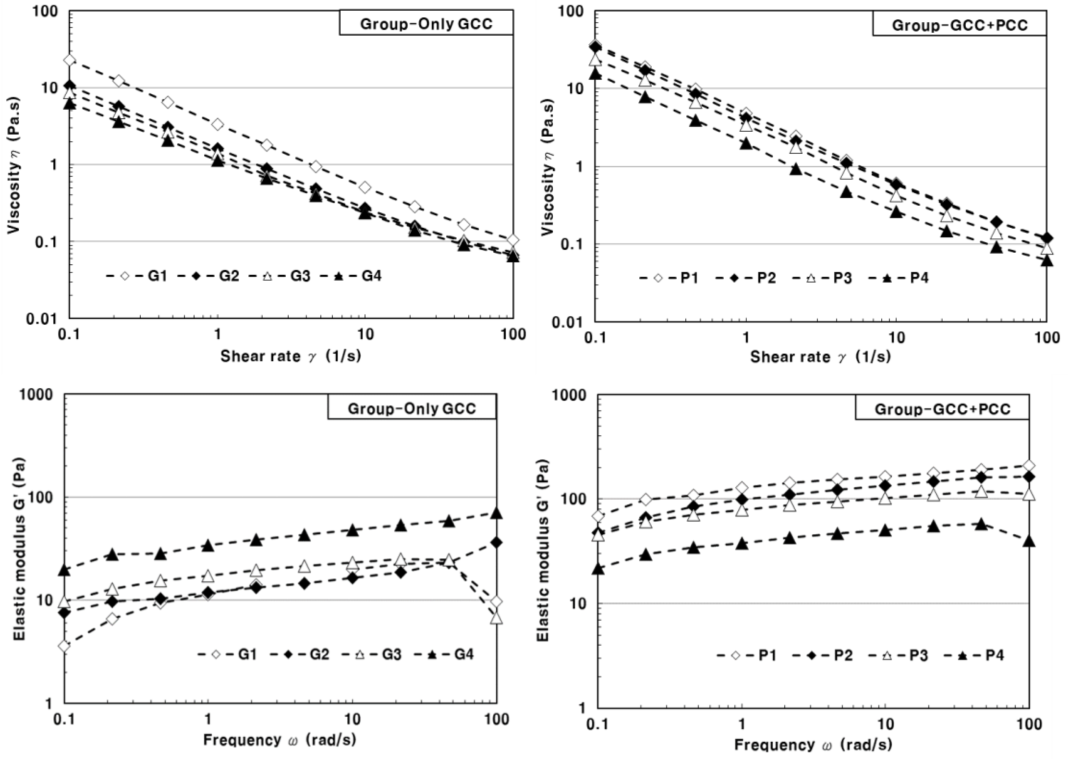

Fig. 1. Effects of pigment type and particle size on the shear viscosity and viscoelasticity of the coating colors

Because Brownian motion effects in small particle suspensions increase even at higher shear rates, the process of coating colors with small particles requires higher shear rates to exhibit the same level of shear-thinning behavior as larger particles (Fadat *et al.* 1986; Dahlvik *et al.* 2000; Bluvol *et al.* 2011). Therefore, P1, composed of small calcium carbonate particles, had the highest viscosity. Furthermore, the particle size of a pigment affects the interactions between particles because there are more particles in the same volume (Kumar *et al.* 2011). These results show that as the particle size decreases, interaction between particles is increased and viscosity increases.

The viscoelasticity of the coating color is influenced by the network formation of the components, and the fluidity of the coating color is closely related to the elastic modulus. As shown in Fig. 1, the elastic modulus of the group Only GCC increased as the GCC particle size increased. Conversely, the elastic modulus of Group-GCC+PCC decreased as the GCC particle size increased. Overall, the elastic modulus of the coating color mixed with porous PCC was high, indicating the behavior of a highly elastic material owing to the interaction between particles and strong network formation. Typically, as the particle size increases, there is less space for the pigment to move, causing the fluidity to decrease and elastic modulus to increase (Fadat *et al.* 1988; Dahlvik *et al.* 2000; Tsunekawa 2000; Jeong *et al.* 2018). However, when mixing particles of different sizes, they can be expected to exhibit different types of elastic behavior because of the differences in the packing structure and aggregation formed by the coating color.

The dewatering amount of the coating color affects the runnability of the coating process, and a high dewatering amount can affect the drying, consolidation, and uniformity of the coat's weight (Nath and Kline 1995; Bluvol *et al.* 2011). The dewatering amount of the coating color for each particle size and pigment type was measured; the results are shown in Fig. 2. The degree of dehydration of the coating color tended to increase as the GCC particle size increased, and porous PCC caused the degree of dehydration to decrease. In addition, as the viscosity of the coating color increased, the degree of dehydration decreased. Generally, when particles of different sizes are mixed, the smaller particles fill the gaps between the larger particles, resulting in a denser structure (Sandas *et al.* 1989; Thitsartarn and Jinkarn 2020). Additionally, the smaller the particles, the narrower the gap between them; thus, the cake formed by the small particles yields a filtration effect. These results suggest that when PCC with a relatively small particle size or high porosity is mixed, dehydration will be suppressed by forming an overall dense structure.

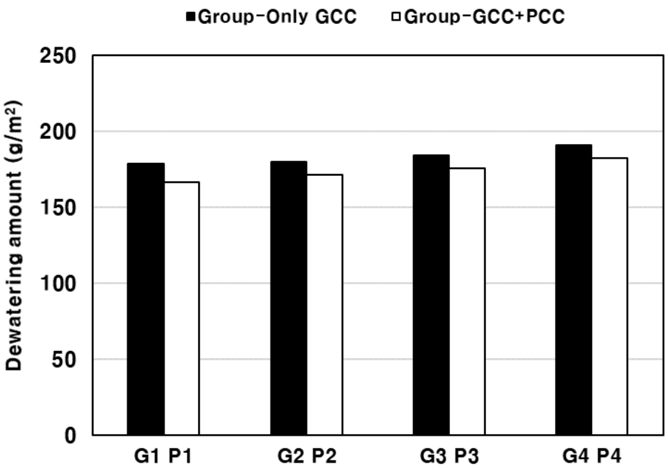

Fig. 2. Effects of the pigment type and particle size on the dewatering amount of the coating colors

Surface Properties of Coated Paperboard

Figure 3 shows the surface of precoated paperboard samples. The roughness of G1 and G2 was similar; however, the roughness increased as the pigment particle size increased. Furthermore, when porous PCC was incorporated, the gaps between GCC particles were filled with PCC to form a smoother surface. G4 and P4, which have larger particle diameters, exhibited more pores and larger particle sizes compared to other coating formulations. Variations in pigment size and morphology are likely to impact the pore size, depth, and distribution on the precoating layer surface, thereby influencing the properties of the subsequent top-coating layer on the paperboard (Li and He 2012; Kim *et al.* 2016; Lee *et al.* 2024).

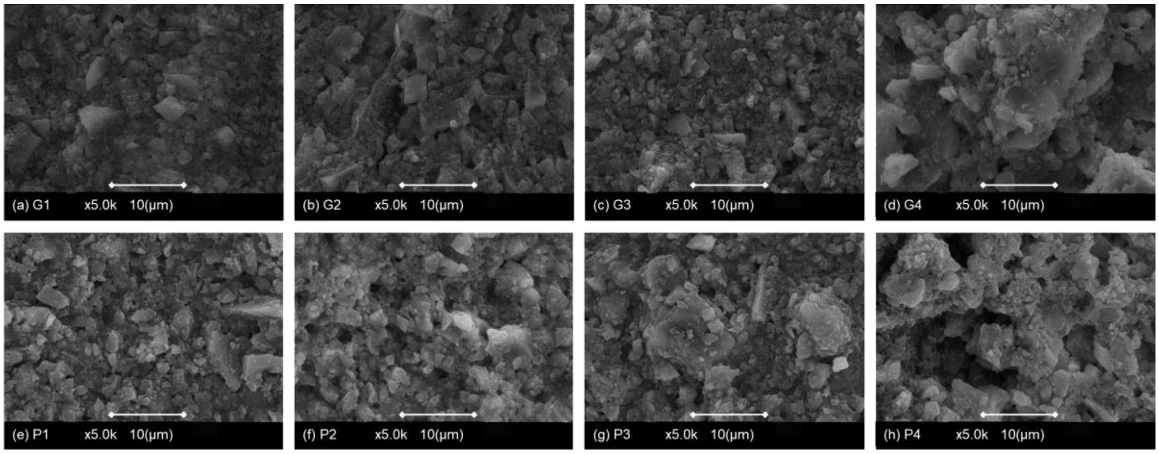

Fig. 3. SEM images of precoating surface

This trend is also reflected in the surface roughness, as shown in Fig. 4. The roughness of the paperboard subjected to the top-coating process increased as the GCC particle size increased, regardless of the PCC mixture. As the GCC particle size increased, the roughness of the precoating layer increased, which also appears to have affected the top-coating layer (Jarnstrom *et al.* 2007). In addition, the PCC mixture contributed to reduce roughness, causing small PCC particles to fill the voids between GCCs and form a smoother surface.

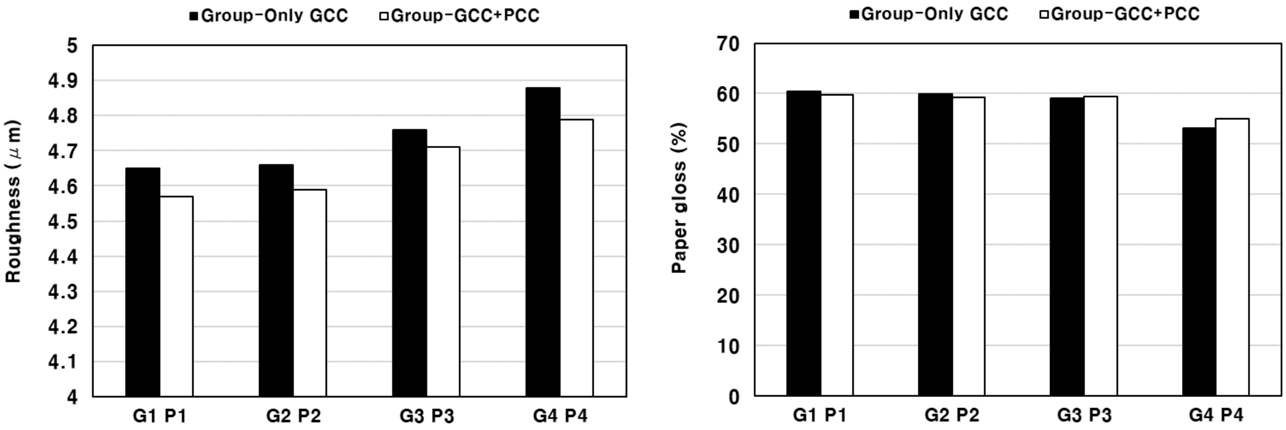

Fig. 4. Effects of the pigment type and particle size on the roughness and paper gloss of the coated paperboard

There was no significant difference in the gloss of the coated paperboard between the two groups (Fig. 4), but the gloss tended to decrease as the GCC particle size increased. Furthermore, the trend of gloss was opposite to the trend of surface roughness. Particularly, large particles formed a rough surface, and a rough surface tends to increase the amount of diffusely reflected light instead of regularly reflected light this is believed to result in less gloss (Engström, and Morin 1994; Santos and Velho 2004).

The brightness and opacity of coated paperboard are shown in Fig. 5 according to the pigment of the precoating layer. Because the same top coating was used in all conditions, the brightness was found to be significantly influenced by the brightness of the top-coating layer. Additionally, the influence of the precoating layer pigment was minimal.

Opacity is affected by the pore structure of the coating layer. Generally, as pores increase, more light is scattered and opacity increases (Günther *et al.* 1989; Kumar *et al.* 2011). However, in this study, the pigment combination was found to have no significant effect on opacity. It is assumed that the same top-coating layer offsets the influence of the precoating layer. In addition, as a result of incorporating PCC, opacity slightly increased, which is believed to be attributable to the aggregation of GCC and PCC.

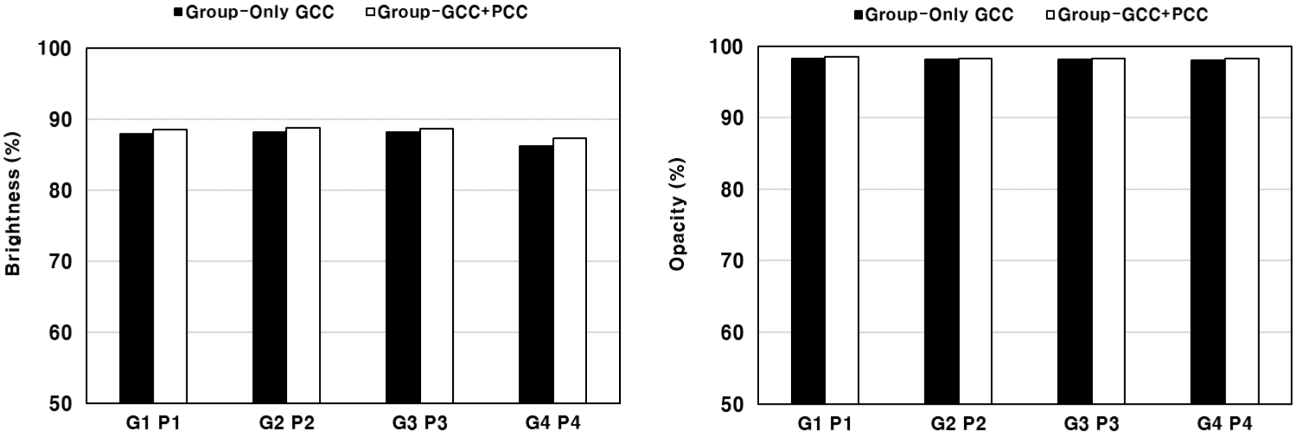

Fig. 5. Effects of the pigment type and particle size on the brightness and opacity of the coated paperboard

Printability of the Coated Paperboard

The coating layer pore structure is one of the most important factors influencing the printing ink behavior on the paper surface (Lee *et al.* 2005; Resch *et al.* 2010; Preston *et al.* 2017). Pore size and volume fraction affect ink absorption and setting. Structures containing small pores are associated with faster ink setting. Figure 6 shows the ink set-off and dry pick strength of the coated paperboard. In general, a slower ink set-off process corresponds to more smearing. Here the GCC particle size was found to have minimal influence on the ink set-off. However, when comparing the group Only GCC and Group-GCC+PCC, the coated paperboard with porous PCC in the precoating layer had excellent ink set-off properties. Thus, pigment condition-related structural differences caused alterations in capillary action, affecting the amount of ink absorption.

The dry pick strength increased as the GCC particle size increased, and it decreased when porous PCC with a small particle size was incorporated. Small-sized pigment particles had a large specific surface area and required more binder. Therefore, under the condition of the same amount of binder, the coating color comprising small-sized pigment particles seemed to have a lower surface strength because of an insufficient amount of binder. These results confirmed that the precoating layer below the top-coating layer sufficiently affects the printability of coated paper.

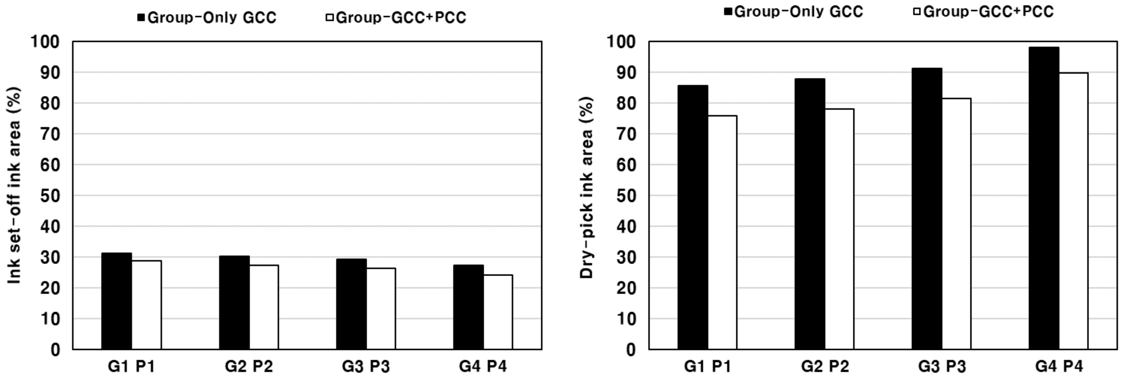

Fig. 6. Effects of the pigment type and particle size on the ink set-off and dry-pick strength of the coated paperboard

CONCLUSIONS

The effects of using coarse ground calcium carbonate (GCC) and porous precipitated calcium carbonate (PCC) as coating pigments for paperboard were investigated. The rheological properties and dewatering amount of the coating colors, as well as the surface properties and printability of the coated paperboard, were evaluated.

1. As the particle size of the pigment decreased, the increase in specific surface area tended to cause the low shear viscosity to increase. In the case of a coating color containing PCC with a relatively small particle size and porous structure, the degree of dehydration was lower than when only GCC was applied. When porous PCC was incorporated, high elasticity was measured over the entire frequency range.
2. The surface roughness increased as the GCC particle size increased, with G1 and G2 and P1 and P2 respectively exhibiting similar surface roughness. In addition, the PCC mixture contributed to reduce roughness. However, GCC particle size was not found to significantly influence the brightness of the coated paperboard; additionally, when porous PCC was incorporated, opacity increased.
3. Although the effects of GCC particle size on the ink set-off properties of the coated paperboard were minimal, incorporating porous PCC significantly improved the ink set-off properties. In addition, the dry pick strength of the coated paperboard tended to increase as the pigment particle size increased.
4. Although the top-coating layer had the same formulation, the structure of the precoating layer affected the printability of the top-coating layer. Therefore, it is expected that 60-grade GCC can be replaced by 55-grade GCC to reduce binder usage.

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