Dynamic Water Penetration Behavior of Top Coating Color and Its Effects on Structure Properties of Double-coated Layer

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The relationship between the dynamic water penetration behavior of top coating color and double-coated paper properties, including pore structure, surface roughness, gloss, and surface latex content, was investigated. Top coating was applied onto the precoated paper prepared using different pigment types, thereby having different pore structures. To investigate the effect of dwell time on pore structure and surface latex content, coating speed and the distance between two coating heads of a laboratory coater were adjusted. As the number of pores of the precoated layer increased, the amount of water that penetrated the precoated paper increased, regardless of low shear viscosity of the top coating color. The relative pore ratio and surface properties of the top coated layer were proportional to those of the precoated layer. A UV absorbance analysis was conducted to examine latex binder migration. The latex content on the surface of the coating layer was mainly affected by the dwell time to the dryer after coating.

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INTRODUCTION

Conventional paper coating is widely used to improve appearance, printability, *etc.* of the paper product by applying coating color that consists of pigment, binder, and additives on the surface of base paper. Several studies related to paper coating have focused on identifying the best pigment-binder combinations and thus the properties of the final coated product. The type and combination of the various coating color components affect the mechanical, optical, and water penetration properties of the coated paper in a complex manner (Morsy and EI-Sherbiny 2004; Ortner *et al.* 2018; Rajabi Abhari *et al.* 2018; Senden *et al*. 2000; Oh *et al*. 2019). A properly formulated coating improves diverse coated paper properties, including brightness, opacity, smoothness, surface strength, ink setting, print gloss, and so on (Al-Turaif 2006; Anthony *et al.* 2015). Not only the coating formulation, but also the water retention of coating color is critical in pigment coating because it influences the rheology and runnability of coating color as well as the pore structure of the coating layer (Sakano and Shigetomi 1999).

In the preparation of double-coated paper, precoating is conducted to improve the smoothness of the base paper and to achieve a more uniform absorptive property, thus producing the desired surface properties and printability of the top coating (Nutbeem *et al.* 2010; Resch *et al.* 2010; Lee *et al.* 2021). In general, the main purpose of the precoating layer is to improve the surface smoothness and reduce water penetration into the paper at a minimum cost. Therefore, low-grade pigments, such as coarse ground calcium carbonate (GCC), coarse grade clay, and starch with high water retention, are mainly used as raw materials to reduce the production cost of precoated paper (Dahlström and Uesaka 2009). The cost reduction strategy of a double-coated paper with a certain coat weight can be optimized by controlling the coat weight ratio between precoating and top-coating. Therefore, it is necessary to understand the influence of each coating layer on the properties of the final double-coated paper and the correlation between the properties of pre- and top-coating layers in the final coating layer.

Several studies have investigated the effect of precoated layer properties on the final coated paper quality. For instance, the effect of binder properties in a precoated layer on the surface characteristics of top-coating layer and print mottle of coated paper, *i.e.*, an uneven printing defect, was investigated (Kim *et al.* 2016). It was found that print mottle of double coated paper improves when the binder with a low T_g is used in the precoated layer. In addition, Renvall *et al.* (1990) showed that the type and particle size of pigments in a precoating color directly affect the printing quality, such as gloss, and surface smoothness, of the final coated paper. However, the coating color formulation of each layer and drying condition should be controlled properly in the double-coating process. Otherwise, double-coated papers can show more severe printing defects such as print mottling during multicolor offset printing, in comparison to single-coated papers.

Recently, Youn and Lee (2022) investigated the influences of pigment type, mixing ratio, and binder type on the dynamic water penetration behavior of precoating color and latex content on the surface of the precoated layer. They showed that when a large amount of pigment with a fine particle size was used, or as the amount of starch binder was increased, the water penetration tendency into the base paper decreased. However, the influence of precoated layer prepared using different types of pigments in the precoated layer on the property of the top coating still requires more investigation. Water retention is one of the critical coating color properties because it changes the pore structure of the coating layer (Sakano and Shigetomi 1999; Nutbeem *et al.* 2010). Furthermore, surface distribution of binders and pigments on a double-coated paper is influenced by the amount of water penetrated into the precoating layer, and this eventually affects the print mottle.

In this study, the dynamic water penetration behavior of top-coating colors was investigated, and its effect on the pore structure of double-coated paper was examined. In addition, surface properties, such as roughness, gloss, and latex content, of the top-coated layer were examined to understand the effect of the precoating layer consisted of different pigments and dwell time on the double-coated paper properties.

EXPERIMENTAL

Materials

Commercially produced base paper with a grammage of 78 g/m^2 was supplied by Hankuk Paper Co. (Ulsan, Korea). The fiber composition of the base paper consisted of 75% softwood bleached kraft pulp, 15% hardwood bleached kraft pulp, and 10% bleached chemithermomechanical pulp. Two ground calcium carbonates (GCC 60 and 95) with different particle sizes and distributions (both from Omya, Korea), and No. 2 clay (Imerys, Savannah, GA, USA) were used as precoating pigments. As top-coating pigments, GCC 95 and No. 1 clay were used. The particle size of pigments was analyzed using a particle size analyzer (Micromeritics, SediGraph 5100, Norcross, GA, USA). The percentage of pigments smaller than 2 µm in diameter for GCC 60, GCC 95, No. 2 clay, and No. 1 clay were 67.9%, 98.9%, 79.3%, and 98.8%, respectively, and the median diameter for these four pigments was 1.15 µm, 0.43 µm, 0.45 µm, and 0.29 µm, respectively. Acetate ester starch (Samyang Genex, Incheon, Korea) and styrene/butadiene (S/B) latex (LG Chemical Co., Yeochun, Korea) were used as binders for the coatings. Small amounts of lubricant (Nopcote C 104, San Nopco Korea Ltd., Pyeongteak, Korea), insolubilizer (Neowet 101H), and thickener (JT-35B, Jeong Won Chemical Co., Ltd., Busan, Korea) were included as additives. The thickener, a synthetic acrylate copolymer, was used to control the rheological characteristics and water retention property of the coating color.

Preparation of Double-coated paper

The precoating process was completed according to the method described by Youn and Lee (2022). Briefly, coating color was prepared by dispersing the clay and GCC using a high-speed stirrer, followed by the addition of the starch and latex binder. The coating color was applied onto the base paper using a laboratory sheet coater (Maiyoh coater, PM-9040, SMT, Chiba, Japan), which had two coating heads designed to allow a rod and a blade to be mounted for doctoring. A total of 3 types of precoated papers with 9.5 g/m^2 of coat weight were prepared and used for top-coating experiments. The pigment compositions of the coating color and surface properties of the precoated paper are presented in Table 1.

Table 1. Pigment Compositions and Surface Properties of Precoated Paper

Formulations	Pigment			Coat Weight (g/m^2)	Roughness* (µm)	Gloss (%)
	GCC 60	GCC 95	No. 2 Clay			
P1	100	-	-	9.52	5.13	6.1
P2	-	100	-	9.47	4.18	24.2
P3	-	-	100	9.61	4.57	22.5
* Roughness was measured by Parker Print-Surf Roughness tester (10 kg$_f$/cm^2)						

Table 2. Top-coating Color Formulations

Components		Grade	T1	T2
Pigment	GCC	GCC 95	40	40
	Clay	No. 1 Clay	60	60
Binder	Starch	Acetate ester	4	0
	Latex	S/B latex	11	13
Additives	Lubricant		1.0	1.0
	Insolubilizer		0.4	0.4
	Thickener		-	0.2

To prepare the coating color for top-coating, a thickener was first added to the pigment under high-stirring conditions, and then starch was added. The starch was cooked for 30 min at 95 °C at 10% consistency and cooled down to 50 °C before use. After stirring for 5 min, lubricant, insolubilizer, and latex were added to the pigment in that order, and stirred for 20 min. The solids content of each coating color was readjusted to 64% with distilled water. Table 2 shows the top coating color formulations.

To measure the dynamic water absorption of the top-coating color to the precoated paper, the first rod coating head was used to apply the coating color with an approximately 20 g/m^2 of coat weight, in which Rod No. 12 was used and the pressure was adjusted to 0.15 MPa. Then, the right half of the coating was scraped off using a blade mounted on the second coating head. The solids content of the scraped-off coating color was then measured. The left half of the coated surface was used to determine the coat weight. The amount of water that penetrated into the precoated paper was determined according to Eq. 1 (Sankano and Shigetomi 1999), where C_0 is coat weight (g/m^2), S_0 is the solids fraction of the original coating color, and S_t is the solids fraction of the scraped off coating color.

$$\text{Absorbed water (g/m}^2\text{)} = (C_0 / S_0) - (C_0 / S_t) \quad (1)$$

To produce double-coated paper, approximately 60 g/m^2 of the top-coating color was applied onto the precoated paper using the first rod coating head mounted with a No. 16 coating rod. The pressure of the coating rod was adjusted to 0.03 MPa. The coating was then doctored using the second blade coating head to obtain 9.5 g/m^2 of coat weight. The dwell time was controlled to 0.06 s by adjusting the coating speed and distance between the two heads to 150 m/min and 0.15 m, respectively. The double-coated paper was then dried using a hot air dryer at 150 °C. To prevent curling of the paper during the drying process, the coated paper was held down on a flat support. The coated paper was calendered twice using a laboratory calender after conditioning at a constant temperature and humidity (23 °C, relative humidity 50%). The line pressure and temperature applied in the calendaring were 90 kg/cm and 45 °C, respectively.

Properties of Double-coated Paper

The pore volume and average pore diameter of double-coated paper were analyzed using mercury porosimeter (AutoPore III, Micromeritics, York, PA, USA). The gloss of the coated paper was examined at an incident angle of 75° according to TAPPI T480 om-15 (2020) standard. The surface roughness of the coated paper was measured using a 3D noncontact surface profiling device (NanoScan E, NanoSystem & Technology, Daejeon, Korea). A UV absorbance analysis was conducted to evaluate content of latex on the surface of topcoated layer because S/B latex contains UV absorbing double bonds (Copyright Kenttä *et al.* 2000).

RESULTS AND DISCUSSION

Dynamic Absorption of Top-coating Color

The runnability of coating color and quality of the final product is strongly affected by the dewatering of coating color during application (Jäder *et al.* 2005). Thus, dynamic water absorption behavior depending on the pore structure of pre-coating layer was examined using different top-coating color formulations. Three precoated papers were prepared using pure GCC 60, GCC 95, and No. 2 clay as pigments. The structural characteristics of the pore in the three precoated layers is shown in Table 3 (Youn and Lee 2022).

Briefly, the coarse GCC 60 precoated layer showed the lowest total intrusion volume compared with the two other precoated layers using GCC 95 and No. 2 clay. This is because a less porous coating structure is formed when pigment with a large particle size distribution is used (Preston *et al.* 2008; Resch *et al.* 2010).

Table 3. Structural Parameters of Precoated Layer as a Function of Pigment Composition (Youn and Lee 2022)

Formulations	Pigment Type	Total Intrusion Volume (mL/g)	Average Pore Diameter (μm)	Relative Pore Number	Porosity (%)
P1	GCC 60	0.0267	0.069	100	6.45
P2	GCC 95	0.0338	0.070	132	7.58
P3	No. 2 Clay	0.0353	0.065	158	7.85

Table 4 shows the properties of top-coating color with different binder systems. The coating color formulated with 100% latex binder (*i.e.*, T2) showed lower viscosity and water retention than T1 coating color, in which starch binder was mixed with latex. This was due to hydrophilic and water holding properties of starch.

Table 4. Properties of Top-coating Color

Formulations	Solids Content (%)	Viscosity (cPs)	pH	Dewatering Amount (g/m^2*)
T1	63.8	2,609	9.3	97.0
T2	64.2	1,516	8.7	161.3
* This value was calculated by a gravimetric water retention meter (AA-GWR, 1.5 bar, 120 s)				

Figures 1 and 2 indicate the effect of pigment composition of precoated layer on the dynamic water penetration of top-coating color, in which the results are depicted as a function of the square root of the penetration time of the top-coating color into the precoated paper.

For pre-coating color, the amount of water that absorbed into the base paper decreased when the proportion of clay increased. This was attributed to the tortuous pathway formation by the high aspect ratio of clay (Youn and Lee 2022). However, the highest amount of water penetration into the precoated paper in the top-coating process was achieved with pure clay precoated paper (*i.e.*, P3T1), and the water penetration rate per unit time was also fast in comparison to the other conditions. Similar results have been reported by Hattori and Yamazaki (1998). They also studied the static water penetration of top-coating color, and the result showed that the amount of water that penetrated into the precoating layer increased when the number of pores in the precoated layer increased. In addition, dynamic water absorption of the T2 formulation, which had lower viscosity and lower water retention than T1, was higher, regardless of pore structure of precoated layer (Fig. 2).

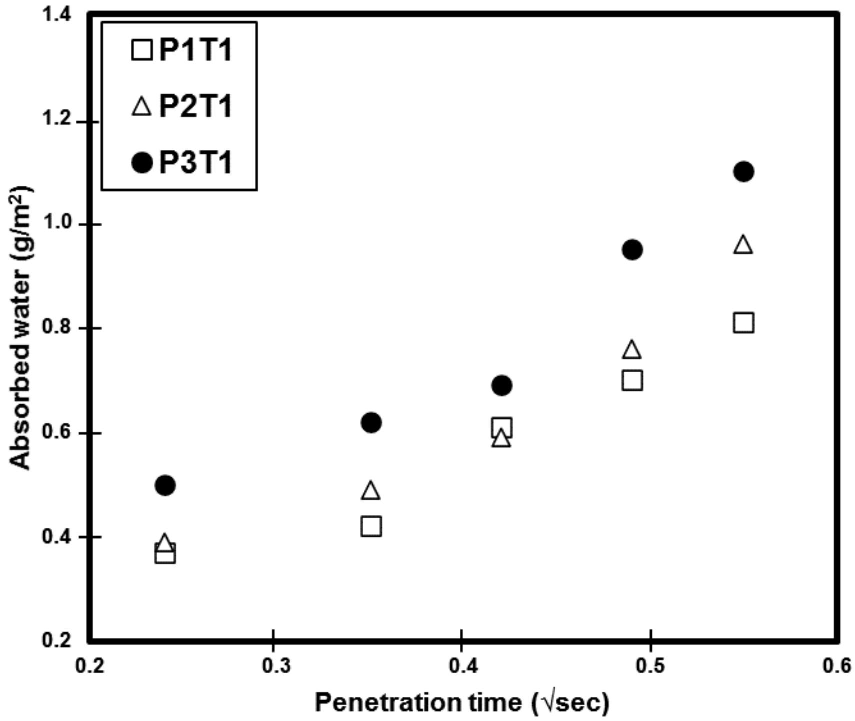

Fig. 1. Effect of pigment composition of precoated layer on the dynamic water penetration of T1 coating color

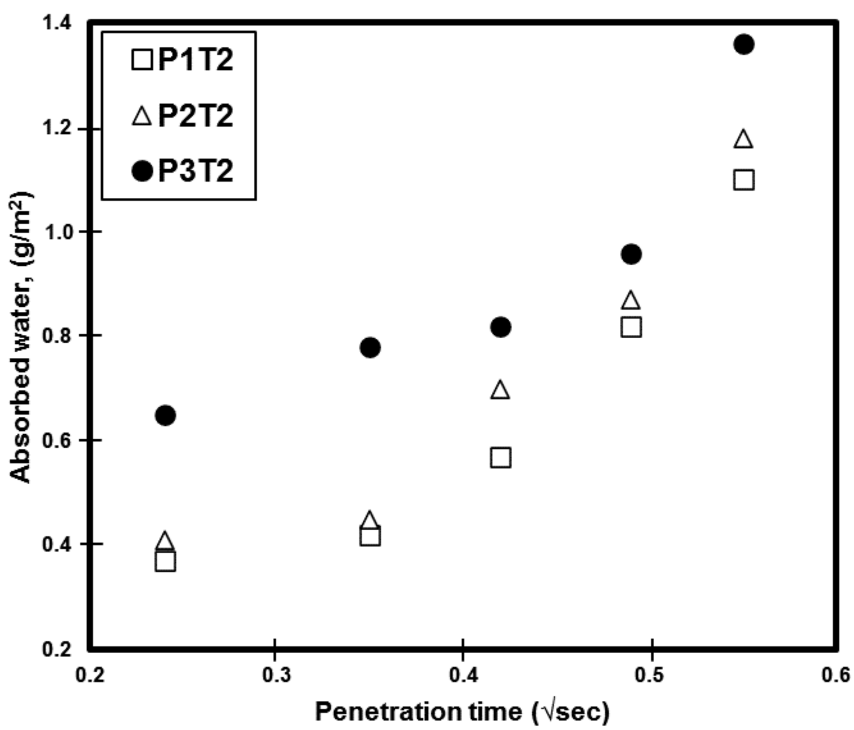

Fig. 2. Effect of pigment composition of precoated layer on the dynamic water penetration of T2 coating color

Pore Structure of the Top-coating Layer

The effect of pre-coating layer

The understanding of the pore structure of double-coated paper may be an important factor for improving the printing characteristics and quality of the final coated paper. The most effective way to investigate the effect of the pore structure of precoated layer on top coated layer is to evaluate the pore structure of the top-coating layer separately. However, it is impossible to measure the pore structure of top-coating layer independently by using a mercury porosimeter.

In this study, the top-coating color was applied onto the precoated paper with different pore structure, and then the entire pore of the double-coated paper was analyzed. Through this method, the effect of the pore structure of the precoated layer on the pore formation of the top-coated layer was indirectly evaluated.

The structural parameters of the double-coated layer are presented in Table 5. The result shows that the relative pore ratio of the double-coated layer increased in proportion to the pore ratio of pre-coated layer, indicating that the pore structure of pre-coated layer may directly influence the pore structure of the top-coated layer of a double-coated paper, which is consistent with a previous study by Gaskin *et al.* (2019).

Table 5. Structural Parameters of Double-coated Layer Depending on Pigment Composition of Precoated Layer

Formulations (Pre+Top)	Total Intrusion Volume (mL/g)	Average Pore Diameter (μm)	Relative Pore Ratio Pre : Pre+Top
P1T1	0.0405	0.0640	100 : 100
P2T1	0.0429	0.0607	132: 118
P3T1	0.0407	0.0585	158: 120

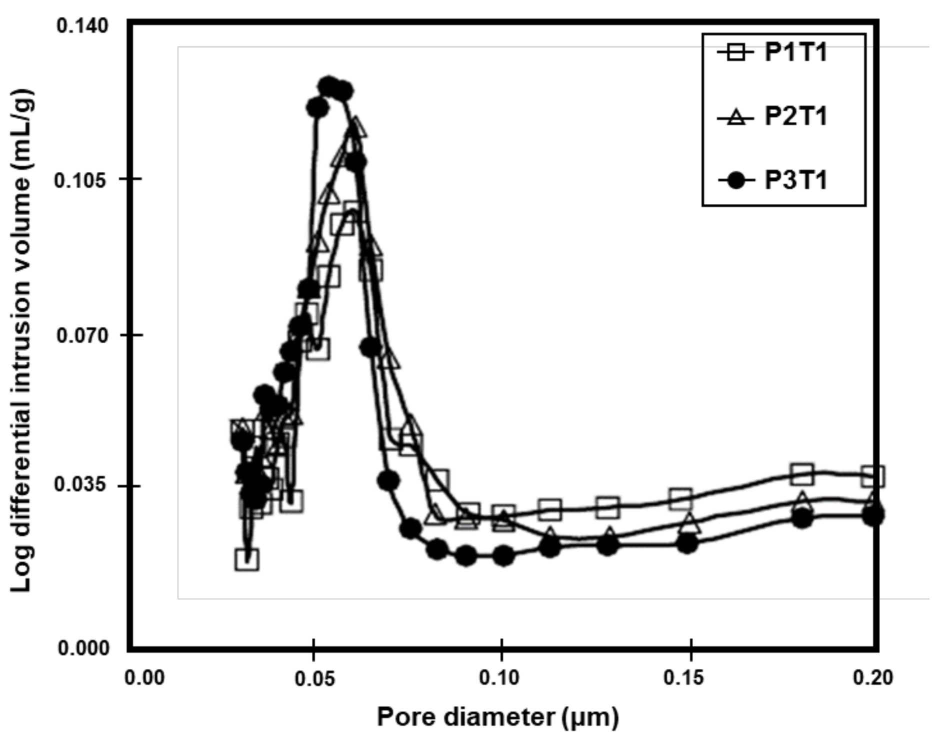

Fig. 3. Pore size distribution of double-coated layer depending on precoating layer

The standard deviation of the pore size distribution was analyzed using the volume of mercury penetrated into the pores of the coating layer (Fig. 4). As shown in Fig. 4, P3T1 had the maximum standard deviation, and the same trend was observed in the precoated layer. This result indicates that the relative pore ratio of P3T1 was higher than the other conditions, but it had a wider pore size distribution. This suggests that P3T1 is strongly likely to give non-uniform latex distribution on the surface and inside of the coating layer, and this is due to higher water penetration behavior of top-coating color (Fig. 1).

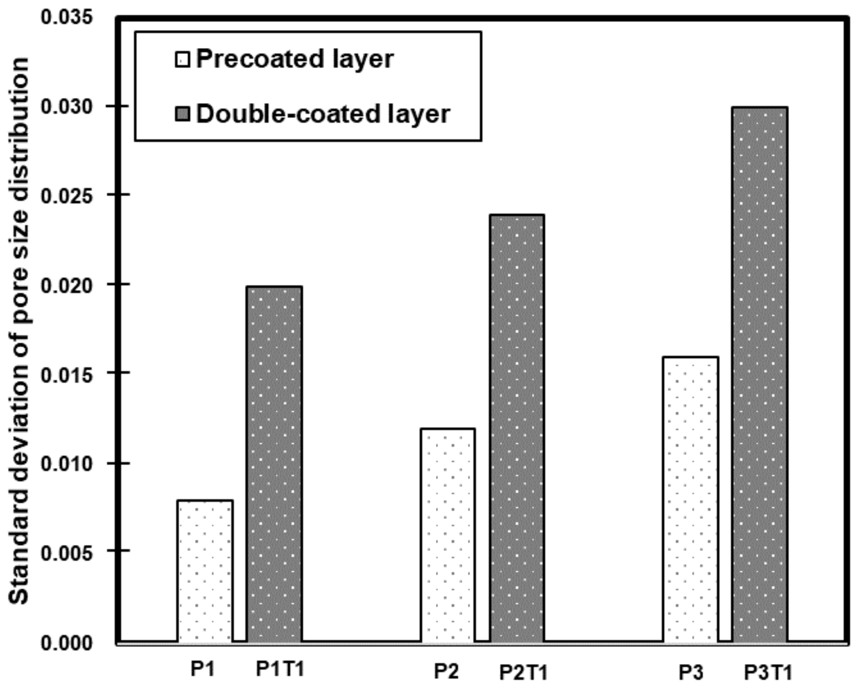

Fig. 4. Standard deviation of pore size distribution depending on pigment compositions of the precoated layer

The effect of dwell time

The coating layer applied on top of the precoated layer consolidates as water penetrates into the bottom layer or evaporates in the course of the drying process. Water penetration of the coating color begins immediately after application and continuously proceeds to reach the immobilization point (Jäder and Engström 2004).

Table 6 shows the effects of dwell time, which is the time between the application and metering of the surplus coating color by blade, on the pore structure of double-coated layer. Pre- and top-coating color formulation of P1T1 condition was used. Table 6 clearly shows that total intrusion volume was affected by the dwell time. As can be seen from the relative pore ratio, the increase of dwell time during the precoating had a greater effect on the pore structure compared with the change of dwell time in the top-coating. However, the dwell time had no noticeable effect on the pore diameter of coated layer. Generally, the printing density of coated paper depends on the coating layer thickness, latex distribution, and pore structure of the coating layer (Ozaki *et al.* 2008). Additionally, Preston *et al.* (2008) reported that non-uniform pore structure of coating layer causes non-uniform ink transfer. These indicates that the influence of the dwell time on the printability of coated paper might not be important for double-coated papers.

Table 6. Structural Parameters of the Double Coated Layer Depending on Dwell Time

Conditions	Total Intrusion Volume (mL/g)	Average Pore Diameter (μm)	Relative Pore Ratio Pre : Pre+Top
$P_{d1}T_{d1}$ *1	0.0388	0.0667	100 : 100
$P_{d1}T_{d3}$ *2	0.0407	0.0656	100 : 108
$P_{d3}T_{d1}$ *3	0.0397	0.0657	123 : 105
$P_{d3}T_{d3}$ *4	0.0402	0.0651	123 : 109

*1: Dwell time of precoating and top-coating were both 0.06 s
*2: Dwell time of precoating and top-coating were 0.06 sec and 0.30 s, respectively
*3: Dwell time of precoating and top-coating were 0.30 sec and 0.06 s, respectively
*4: Dwell time of precoating and top-coating were both 0.30 s

Surface Properties of Top-coating Layer

The effect of precoating layer

The influence of pigment particle size on the surface properties of the precoated layer, such as roughness and gloss, has been examined in a previous study (Youn and Lee 2022). The results showed that the coating surface applied by course GCC (*i.e.*, GCC 60) exhibited high roughness and low gloss, while a highly smooth and glossier surface was obtained when using pigments with fine particle size including GCC 95 and clay. The same trend was obtained for double-coated papers (Table 7), indicating the importance of surface properties of the precoated layer. In general, the target top-coat weight of a glossy paper with basis weight of 150 g/m^2 is typically 10 g/m^2. However, it seems difficult to completely cover the rough surface of the precoated layer with the coating amount of 10 g/m^2.

To investigate the effect of water absorption of the top-coating color on the latex distribution on the top surface, UV absorbance analysis was conducted (Table 7). Latex binder in the coating color was relatively small compared to the pigment suggesting that the binder migrated with water in the coating layer until the immobilization point was reached during the drying process.

The results showed that the pure GCC 60 precoated layer, which exhibited less water absorption, gave higher UV absorbance. This result indicated that a remarkable amount of latex migrated to the coating surface. In contrast, the lowest UV absorbance was obtained when top-coating color was applied on the precoated layer with pure clay formulation, indicating that the latex penetrated more into paper. Exceptionally high UV absorbance was observed when GCC 95, which formed dense surface structure, was used as a pre-coating pigment. Further research is needed to explain this phenomenon.

Table 7. Surface Properties of Double-coated Paper

Formulations	PPS Roughness (μm)	Paper Gloss (%)	UV Absorbance
P1T1	1.12	75.0	4.063
P2T1	1.05	76.9	4.413
P3T1	0.98	77.6	2.523

The effect of dwell time

Generally, a short dwell time made the binder migrate to the surface of the coating layer, while long dwell time gave uniform binder distribution in the z-direction of coating layer (Zhen and Wang 2013). This is because the amount of water that penetrates into the base paper or precoated layer increases as the dwell time increases.

Figure 5 displays the UV absorbance of the top-coated layer depending on dwell time. The result showed that the UV absorbance increased as the dwell time increased, indicating that a substantial amount of latex migrated to the surface of the coating layer rather than penetrating into the base paper or precoated layer. This happened because the change of coating speed and distance of two heads required for controlling the dwell time changed the interval required for the paper to enter the dryer after the second coating head (Table 8).

The relationship between the time required to transfer the coated paper from #2 coating head to the dryer and UV absorbance is depicted in Fig. 6. A shorter transfer time to the dryer resulted in a higher UV absorbance. It is worth noting that the latex content on the surface of coating layer was more affected by short transfer time to the dryer after coating than the dwell time.

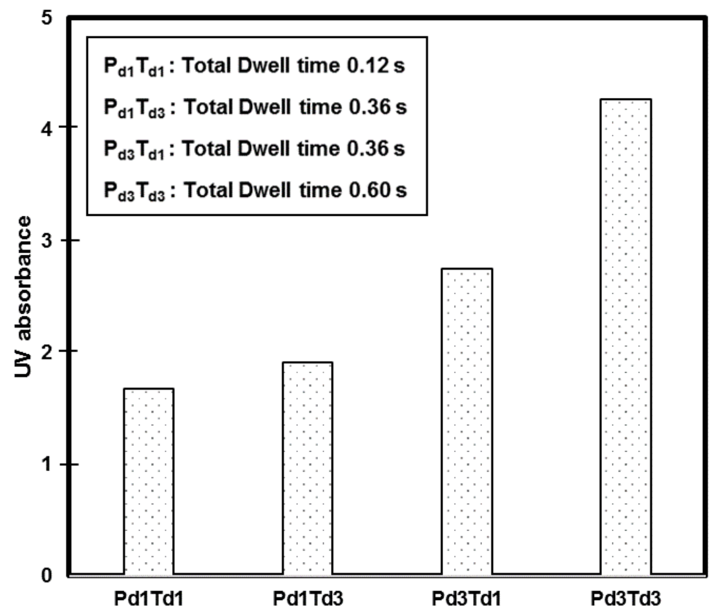

Fig. 5. UV absorbance of top-coating layer depending on dwell time

Table 8. Coating Conditions for Various Dwell Time

Conditions	Speed (m/min)		# 1-2 Head Distance (m)	# 2 Head-dryer Distance (m)	# 2 Head-dryer Interval (s)
$P_{d1}T_{d1}$	P_{d1}	150	0.15	0.575	0.230
	T_{d1}	150	0.15	0.575	0.230
Sum	-	-	-	-	0.460
$P_{d1}T_{d3}$	P_{d1}	150	0.45	0.575	0.230
	T_{d1}	90	0.45	0.275	0.183
Sum	-	-	-	-	0.413
$P_{d3}T_{d1}$	P_{d1}	90	0.45	0.275	0.183
	T_{d1}	150	0.15	0.575	0.230
Sum	-	-	-	-	0.413
$P_{d3}T_{d3}$	P_{d1}	90	0.45	0.275	0.183
	T_{d1}	90	0.45	0.275	0.183
Sum		-	-	-	0.366

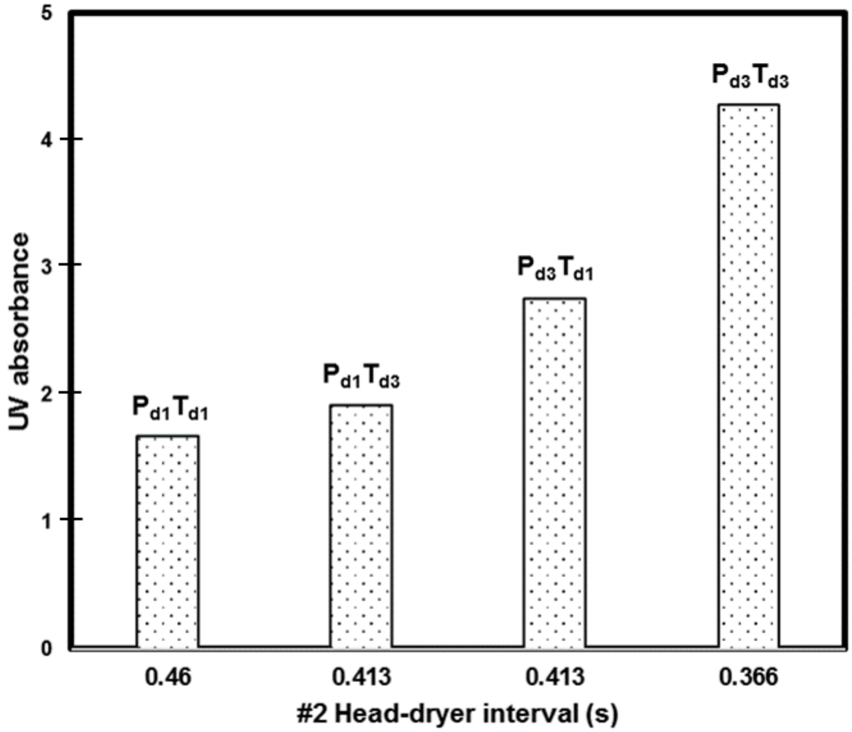

Fig. 6. UV absorbance depending on intervals between #2 head and dryer

CONCLUSIONS

The influence of the precoated layer structure depending on pigment types on the dynamic water penetration behavior of top-coating color and pore structure of the coating layer was investigated. In addition, surface properties, such as roughness, gloss, and latex content on the surface of the top-coated layer were examined to understand the effect of precoated layer and dwell time on double-coated paper properties.

1. Top-coating was applied on the precoated paper prepared by different pigment types. The results suggested that the dynamic water absorption of top-coating color increased with the increase of the number of pores in the precoated layer. Additionally, topcoating color with the lower shear viscosity showed higher water penetration to the precoated paper.
2. The relative pore ratio of the top-coated layer was proportional to that of the precoated layer. The major factors controlling the standard deviation of the pore size in the double-coated layer were the penetration rate of top-coating color and pore ratio of the precoated layer.
3. The surface roughness and paper gloss of double-coated paper were proportional to that of the precoated layer. The surface latex concentration of the top-coated layer decreased with the increase of water penetration rate of the topcoating color into the precoated layer. In addition, the degree of binder migration to the surface was proportional to the drying rates, which indicates that the concentration of surface latex increased with the increase of drying rate.

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